OUTLOOK FOR ADVANCED BIOFUELS

Vooruitzichten voor geavanceerde biobrandstoffen

(met een samenvatting in het Nederlands)

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Carlo Noël Hamelinck

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Promotor: Prof. dr. W.C. Turkenburg
verbonden aan de Faculteit Scheikunde van de Universiteit Utrecht

Co-promotor: dr. A.P.C. Faaij
verbonden aan de Faculteit Scheikunde van de Universiteit Utrecht

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“More than machinery we need humanity”

Charlie Chaplin – The Great Dictator – 1940

Contents

Summary 7

Samenvatting 11

1 Introduction to biofuels 17

2 Research focus and objectives 19

2.1 Traditional fuels 19
2.2 Advanced fuels 20
2.3 Production of the selected biofuels 24
2.4 Status of knowledge 25
2.5 Objective of this thesis 26

3 Research method 26

3.1 Large scale bioenergy supply by international transport 26
3.2 Producing methanol, ethanol, hydrogen and FT diesel 26

4 Results and discussion 28

4.1 Feedstock costs 28
4.2 Selected biofuels 29
4.3 Technological insights 32
4.4 Comparison with other studies 33
4.5 Broader comparison 33
4.6 Energy requirement for energy 37
4.7 Biofuels' use in cars 38
4.8 CO_2 emission avoided, and costs 41
4.9 Transition 43

5 Conclusion 43

5.1 Results 44
5.2 Recommendations for research and development 45
5.3 Closing remarks 46

References 47

Paper 1 53
International bioenergy transport costs and energy balance

Paper 2 83
Future prospects for production of methanol and hydrogen from biomass

Paper 3 119
Fischer-Tropsch liquids and power from biomass
via gasification: exploration of the possibilities

Paper 4 149
Production of Fischer-Tropsch transportation fuels from biomass;
technical options, process analysis and optimisation, and development potential

Paper 5 181
Prospects for ethanol from lignocellulosic biomass:
Techno-economic performance as development progresses

Appendix 217
Abbreviations and glossary 219
Resources and fuels characteristics 220
Conversion factors 222

Curriculum Vitae 227

Selection of publications 229

Dankwoord 231

Summary

This dissertation assesses the technological and economic potential of biofuels. The **Introductory essay** provides a background to the research, it summarises and compares the appended papers, and it puts the results into a broader perspective.

Modern use of biomass can play an important role in a sustainable energy supply. Biomass abounds in most parts of the world and substantial amounts could be produced at low costs. Motor biofuels seem a sensible application of biomass: they are among the few sustainable alternatives to the transportation sector and can address many of the problems associated with mineral oil.

Many biofuels are conceivable. Biodiesel (from oil crops) and ethanol from sugar beets or grains are already used in practice. However, these traditional biofuels have severe disadvantages in land use, costs, and potential to reduce CO_2 emissions. Sugar cane ethanol and advanced biofuels that are produced from lignocellulosic biomass via gasification-synthesis or via hydrolysis-fermentation have much better perspectives.

Based on the possibilities to apply fuels in the present transportation sector, the implementation ease at short and middle term, the ultimate attractiveness (e.g. fuel economy in fuel cell cars), possibilities for process scale-up, and expected economic performance on the long-term, four attractive biofuels are selected for further analysis: methanol, hydrogen, Fischer-Tropsch (FT) diesel and ethanol. Previous studies in the biofuels field, and on these four fuels in particular, did not yield comparable results, and did not give insight in the further improvement potential by the use of advanced future conversion technologies, application of large scale, process optimisation and electricity co-production.

The method followed for analysis of the four candidate fuels is globally the same. Existing and developing technologies, that could be used for the conversion were studied. Conversion concepts were developed for the production of methanol, ethanol, hydrogen, and Fischer-Tropsch diesel. The concepts incorporated new and improved technologies and were selected on potential low cost or high energy efficiency. Some concepts explicitly co-produced power to exploit the high efficiencies of once-through conversion. Biofuel production plants were modelled, optimised towards internal heat demand and supply, and surplus heat was converted to electricity. The models directly yielded the plant energy balance and flow dimensions that could be used for the economic calculations.

An important input parameter to the calculations is the feedstock costs. To supply biomass from production areas to energy importing regions, long distance international transport is necessary, implying additional logistics, costs, energy consumption and material losses compared to local utilisation. In **Paper 1**, a broad variety of chains of European and Latin American bioenergy carriers delivered to Western Europe is composed and evaluated. Different biomass feedstock production systems, pre-treatment and conversion operations, and transport of raw and refined solid biomass and biofuels are taken into account, as well as the influence of key parameters such as distance, timing and scale, on performance. 300 MW_{HHV} European biomass crops can be delivered at 70 €/$tonne_{dry}$ or 3.7 €/GJ_{HHV} when shipped as pellets. South American crops are produced against much lower costs, and

despite the long shipping distance, the costs in the receiving harbour can be as low as 40 €/tonne$_{dry}$ or 2.1 €/GJ$_{HHV}$, only 0.5 €/GJ$_{HHV}$ of this is in the actual international shipping. The relatively expensive truck transport from production site to gathering point restricts the size of the production area. Therefore a high biomass yield per hectare is vital to enable large scale systems. Methanol produced in Latin America and delivered to Europe may cost 8 – 10 €/GJ$_{HHV}$, when the pellets-to-methanol conversion is done in Europe the delivered methanol costs are slightly higher. The energy requirement to deliver solid biomass from both crops and residues from the different production countries is 1.2 – 1.3 MJ$_{primary}$/MJ$_{delivered}$.

It can be concluded that international bioenergy trade is possible against low costs and modest energy loss; and that it is not blocking the development of large bioenergy facilities. The essay continues by summarising the short- and long-term technological and economic performance of the four candidate biofuels, which has been assessed in separate papers:

Paper 2 handles the prospects for the production of methanol and hydrogen from biomass via gasification. A technology review – including promising future components – results in a set of promising conversion concepts. These were modelled to analyse the technical performance. The total energy efficiencies (biomass to fuel *and* electricity) are around 55 % by HHV for methanol and around 60 % for hydrogen production. The technical results were subsequently used for economic evaluation. 400 MW$_{HHV}$ input systems produce biofuels at 8 – 12 €/GJ$_{HHV}$, dictated by the capital investments. Although the outcomes for the different concepts are rather comparable, it can be concluded that optimised fuel production with little or no electricity co-production and hydrogen production using ceramic membranes perform somewhat above average. Long term (2030) cost reductions reside in cheaper biomass, technological learning, and scale-up to 2000 MW$_{th}$. This could bring the production costs of biofuels in the 5 – 7 US$/GJ range.

Paper 3 deals with the production of Fischer-Tropsch (FT) liquids from biomass, also via gasification. The required technology is reviewed; the most promising conversion configurations are identified and modelled. The total energy efficiencies could be 33 – 40 % by LHV for systems based on atmospheric gasification, and 42 – 50 % for pressurised. In the short term, production costs of FT liquids will be about 14 €/GJ (FT diesel 15 €/GJ) decreasing to 9 €/GJ on the longer term; the resulting diesel could then cost 10 €/GJ. Several improvement options could further reduce the costs, and more minute research could reduce the uncertainty. Therefore, the perspectives for producing FT liquids from biomass, and for their use in the transport sector could still be better.

Sequel to this paper, **Paper 4** reanalyses the system components necessary for FT diesel production from biomass and redesigns a limited set of promising conversion concepts. The main variations are now in gasification pressure, the oxygen or air medium, and in optimisation towards liquid fuels only, or towards the product mix of liquid fuels and electricity. A dynamic flowsheet model was built, allowing for direct evaluation of the influence of each parameter or device, on investment costs, FT and electricity efficiency and resulting FT diesel costs. FT diesel produced by conventional systems on the short term and at moderate scale, would probably cost 16 €/GJ. On the longer term (large scale, technological learning, and selective catalyst), this could decrease to 9 €/GJ.

Finally, **Paper 5** handles the production of ethanol from lignocellulosic biomass via hydrolysis fermentation. It reviews state of the art conversion technology and technology currently under development. Promising conversion concepts for short term and future are defined; their technical performance was analysed, and the results were used for economic evaluations. Biomass may be converted to ethanol at 35 – 48 % by HHV on short – long. The not fermentable lignin is used for electricity co-production, so that overall efficiencies are 50 – 68 %. The ethanol production costs are about 22 €/GJ_{HHV} in the short-term (5 years). A combined effect of higher hydrolysis fermentation efficiency, lower specific capital investments, increase of scale and cheaper biomass feedstock costs (from 3 to 2 €/GJ), could bring this via 13 €/GJ_{HHV} (10-15 years) down to 9 €/GJ_{HHV} in 20 years or more, if further development of the technology would be stimulated.

Back in the essay, the results for the separate fuels are presented in a directly comparable manner, by using the same input parameters. In the long term (2030) methanol and hydrogen could be produced at 9 €/GJ_{HHV}. Ethanol and FT diesel are more expensive (11 and 13 €/GJ_{HHV}). These numbers are above the current gasoline *production* costs that ranged from 3 to 7 and diesel from 2 to 7 €/GJ_{HHV} (1 €/GJ_{HHV} equals 0.0352 €/l gasoline 0.0377 €/l diesel) in the last ten years, depending on the oil market price. The uncertainties in the biofuels production costs of the four named biofuels is 15 – 30 %, this is small when considering the large uncertainty in future (2030) gasoline/diesel prices.

The four advanced biofuels are put into broader perspective with other biofuels, again using the same input parameters (when possible). Production costs for DMM, DME and synthetic natural gas might fall in the same range. In biomass producing regions such as Latin America or Eastern Europe, the four candidate fuels (methanol, ethanol, hydrogen and FT diesel) could be produced against 7 – 11 €/GJ_{HHV}. Biodiesel from rapeseed and ethanol from sugar beet or starch crops are already available, but are relatively expensive (25 – 40 €/GJ_{HHV}).

For an equal comparison the fuels' performance are expressed on a comparable basis: their cost of driving. When excluding biofuel excise duty and VAT, the driving costs are dominated by the car costs and fuel attributes about 30 % for RME in ICEVs and only about 9 % for hydrogen in FCVs. The more expensive and more efficient a car is, the less important the delivered fuel costs. At present, biofuels would be an expensive CO_2 avoidance option (100 – 300 €/tonne CO_2), but this may decrease to 50 - 100 €/tonne CO_2 which is similar to costs of other options. These costs are more influenced by the cars' costs, than by the fuels' costs.

When concerning both the costs of driving, and the potential and costs of CO_2 emission reduction, fuels from lignocellulosic crops and sugarcane are found to have the best prospects. However, with the present knowledge and data quality it is difficult to point out the absolute best fuel. Eventually, gasification based processes give a broader flexibility in fuel choice than hydrolysis fermentation. Methanol, DME and Fischer-Tropsch diesel are fuels that could gradually be introduced as blends with gasoline and diesel, and hardly require infrastructure adaptations. Sugar/starch derived ethanol blends with gasoline and rapeseed derived biodiesel with diesel may remain the most popular (and important) biofuels until 2010. Ethanol-gasoline blends could facilitate development of better ethanol processes, and the introduction of neat ethanol vehicles later. biodiesel does not have such advantages. On longer term, hydrogen, methanol and DME are the most suitable in a transition to fuel cell vehicles, if that would be desirable. The eventual shift to hydrogen then requires concurrent development of new infrastructure and FCV introduction.

Our knowledge on the performance of biofuels should be further improved, by closer assessment of several points on the route from crop to motion, and of the route as a whole. The concluding part of the introductory essay lists recommendations for further research and development. On socio-economic level this calls for increasing the knowledge on the sustainability of the required large-scale plantations, the evolvement of international bioenergy market, the role of trade barriers to this market, the transition path(s) to a biofuels future, and the effect of governmental stimulation measures and public perception on this path. Technological/economic points of R&D attention should include understanding of the limits to large-scale gasification, the development of effective gas cleaning, and improving the car end-use comparison.

In short, biofuels will certainly play a role in the future energy supply system. The weight of this role depends on the importance attached to fuel security, and on the willingness to address the increased greenhouse effect (caused by human activities) and reduce CO_2 emissions.

Samenvatting

Dit proefschrift gaat over transportbrandstoffen uit biomassa, en dan met name over de productie en de kosten van deze biobrandstoffen. Het **inleidende essay** laat de achtergrond van het onderzoek zien, vat de resultaten uit de onderliggende artikelen samen, vergelijkt ze met elkaar en met resultaten van andere onderzoeken, en plaatst de resultaten in het bredere perspectief van distributie en eindgebruik.

Het moderne gebruik van biomassa (plantaardig materiaal) voor elektriciteitsproductie, centrale warmte productie of transportbrandstoffen kan een belangrijke rol spelen in een duurzame én CO_2 (koolstofdioxide)-arme energievoorziening. Er is biomassa in overvloed in de meeste delen van de wereld en veel ervan zou tegen lage kosten geproduceerd kunnen worden. Transportbrandstoffen lijken een zinnige en aantrekkelijke toepassing van biomassa. De transportsector vertegenwoordigt 21 % van het huidige wereldenergiegebruik. Maar terwijl elektriciteit duurzaam uit andere bronnen (zoals uit wind- en zonne-energie) kan worden geproduceerd, heeft de transportsector veel minder alternatieven. Naast het verminderen van CO_2 uitstoot speelt ook energiezekerheid een grote rol. De belangrijkste olievoorraden zijn in handen van een kleine groep landen (OPEC), en de grootste verbruikers zijn West Europa en de Verenigde Staten; een allesbehalve evenwichtige situatie. De olievoorraden zijn ook eindig. Hoewel schattingen hierover uiteenlopen, is het waarschijnlijk dat aardolie op termijn duurder wordt.

Er zijn vele biobrandstoffen denkbaar. Biodiesel (uit koolzaad) en alcohol uit suikerbieten of graan/maïs is nu al te koop in Frankrijk, Duitsland, Spanje, en de Verenigde Staten. Helaas hebben juist deze ‘traditionele’ biobrandstoffen nogal wat nadelen wat betreft landgebruik. Ook dragen ze weinig bij aan het verminderen van CO_2 uitstoot en zijn ze duur. Ethanol uit suikerriet (o.a. sinds 1980 toegepast in Brazilië) en de zogenaamde ‘geavanceerde’ biobrandstoffen zijn meerbelovend. Deze laatste groep wordt uit houtachtige biomassa geproduceerd door vergassing gevolgd door synthese, of door hydrolyse en fermentatie.

Sommige van de brandstoffen zijn gemakkelijk in te passen in de huidige transportsector, doordat ze veel lijken op de huidige benzine of diesel of omdat ze (op kleine schaal) ermee vermengd kunnen worden. Anderen vragen slechts kleine aanpassingen in het distributiesysteem, aan tankstations en/of in de auto en zijn meer geschikt voor de middellange termijn. Voor de lange termijn zijn de mogelijkheden voor grootschalige productie en gebruik, en de uiteindelijke kosten van groot belang. Op grond hiervan zijn vier biobrandstoffen geselecteerd die aantrekkelijk lijken voor de middellange en de lange termijn: methanol, waterstof, synthetische diesel en ethanol. Eerdere studies in het biobrandstoffenveld, en naar deze vier brandstoffen in het bijzonder, waren vooral van verkennende aard. Ze leverden resultaten op die moeilijk met elkaar te vergelijken waren. Bovendien gaven ze weinig inzicht in welke factoren de efficiëntie en de productiekosten beïnvloeden. We willen graag weten, hoe de kosten omlaag zouden kunnen gaan door het gebruik van nieuwe productieprocessen, door afstemming van intern energiegebruik, door opschalen, en door het gelijktijdig produceren van elektriciteit uit een deel van de grondstof.

De methode die voor de analyse van de vier kandidaat-biobrandstoffen gevolgd wordt, is globaal dezelfde. Eerst zijn de productieprocessen bestudeerd, zowel apparaten en reactoren die commercieel beschikbaar zijn, als technologie die nog in ontwikkeling is. Op grond hiervan zijn fabrieken ontworpen, die waarschijnlijk efficiënt en tegen lage kosten kunnen produceren. Sommige ontwerpen zijn gericht op de optimale coproductie van brandstoffen én elektriciteit, andere alleen op brandstofproductie. Interne warmteproductie en warmtevraag zijn zoveel mogelijk afgestemd, waarbij eventuele overtollige warmte is gebruikt voor elektriciteitsproductie. De modellen leverden rechtstreeks de balansen van energie en massa binnen de fabriek, de afmetingen van stromen en apparaten. Deze gegevens zijn gebruikt om de brandstofproductiekosten uit te rekenen.

Om deze berekeningen te kunnen maken, is het nodig om de grondstofprijs van de biomassa te weten. Biomassa moet op grote schaal worden geleverd aan West Europa, vanuit productiegebieden die bijvoorbeeld in Zuid Amerika of Oost Europa liggen. Er is dus internationaal transport nodig, met als gevolg een ingewikkelde logistiek, extra kosten, energieverbruik en materiële verliezen, ten opzichte van wanneer de biomassa plaatselijk zou worden gebruikt. In het **eerste artikel** wordt een grote verzameling internationale transportketens ontworpen en vergeleken, van Europese en Latijns Amerikaanse bio-energie die naar West Europa wordt vervoerd. De analyse houdt rekening met verschillende biomassa productiesystemen (groei en oogst), voorbehandeling (drogen, verkleinen, samendrukken), met omzetting naar tussen- of eindproducten, met verschillende vervoersmiddelen (vrachtwagen, trein, schip), en met de invloed van belangrijke parameters zoals afstand, timing en schaal, op bijvoorbeeld de kosten. Op een schaal van 300 MW kan Europese biomassa, verscheept als pellets, tegen 70 €/droge Ton (1 Ton is 1000 kg) of 3.7 €/GJ bezorgd worden. Zuid Amerikaanse biomassa kan tegen veel lagere kosten geproduceerd worden en – ondanks de grote afstand – tegen lagere kosten in West Europa geleverd worden: 40 €/droge Ton of 2.1 €/GJ. Slechts 0.5 €/GJ hiervan zit in het eigenlijke internationale transport. Het lokale transport per vrachtwagen, van het productiebos naar een centraal punt, is tamelijk duur en dat beperkt de afmeting van het productiegebied. Vooral dáárom is een hoge opbrengst per hectare essentieel voor het realiseren van grootschalige systemen. Als methanol geproduceerd wordt in Latijns Amerika en als vloeistof wordt vervoerd, dan kunnen de totale kosten 8 – 10 €/GJ zijn. Als de omzetting van pellets naar methanol in Europa wordt gedaan, dan is de methanol iets duurder. Er is natuurlijk extra energie nodig om bio-energie internationaal te vervoeren; zo'n 20 – 30 % externe energie boven op de geleverde hoeveelheid bio-energie. Dit is met name transportbrandstof (diesel, stookolie) en elektriciteit. Ter vergelijking, het internationale transport van steenkool heeft zo'n 10 % extra energie nodig.

Er kan nu geconcludeerd worden dat internationale handel in bio-energie mogelijk is tegen lage kosten en met een bescheiden energieverlies. Het hoeft de grootschalige toepassing van biomassa in de energievoorziening en de ontwikkeling van grote biomassaverwerkende fabrieken niet te belemmeren. Op dit punt gaat het essay verder met het samenvatten van het korte en de lange termijn prestaties van de vier gekozen biobrandstoffen, die in de aparte artikelen behandeld worden:

Het **tweede artikel** gaat over de vooruitzichten voor productie van methanol en waterstof uit biomassa na vergassing. Vergassing lijkt op verbranding met weinig zuurstof, waardoor brandbare gassen ontstaan die voor een groot deel uit koolmonoxide (CO) en waterstof (H_2) bestaan. Deze twee moleculen kunnen worden gebruikt voor de productie van methanol (CH_3OH) door ze – op een katalysator – met elkaar te laten reageren. Door koolmonoxide met water te laten reageren, kan ook extra waterstof worden geproduceerd. Er is een overzicht gemaakt van de technologie die voor deze processen nodig is. Dit overzicht bevat ook veelbelovende maar nog niet commerciële processen. Op grond van dit overzicht zijn een beperkt aantal fabrieken ontworpen, die vervolgens zijn gemodelleerd om de technische prestatie te analyseren. De totale energie-efficiëntie (biomassa naar brandstof én elektriciteit) is ongeveer 55 % voor methanol en 60% voor waterstof. De technische resultaten werden voor economische evaluatie vervolgens gebruikt. Gebaseerd op 400 MW aan biomassa worden de brandstoffen geproduceerd tegen 8 – 12 €/GJ. Deze kosten bestaan voor een groot deel uit kapitaalsinvesteringen. De resultaten voor de verschillende fabrieken zijn redelijk gelijk. Op de lange termijn kan kostenvoordeel behaald worden door goedkopere biomassa, ervaring, en opschaling naar 2000 MW. Dit zou de productiekosten naar 5 – 7 €/GJ kunnen brengen.

Uit CO en H_2 kan – met behulp van een andere katalysator – ook synthetische diesel worden geproduceerd, door middel van het zogenaamde Fischer-Tropsch (FT) proces. Hierover gaat het **derde artikel**. Ook hier is eerst de noodzakelijke technologie bestudeerd en zijn veelbelovende fabrieken ontworpen en gemodelleerd. De totale energie-efficiëntie is 33 – 40 % voor systemen gebaseerd op atmosferische vergassing, en als de vergassing onder druk plaatsvindt kan de efficiëntie 42 – 50% zijn. Op korte termijn zullen de productiekosten van synthetische diesel ongeveer 15 €/GJ zijn, en 10 €/GJ op de langere termijn. Er zijn enkele verbeteringen mogelijk die de kosten verder zouden kunnen verminderen, en ook nauwkeuriger onderzoek zou de onzekerheden verkleinen. De verwachting is dan ook dat het produceren van synthetische diesel goedkoper kan.

Om hierop verder te gaan, wordt in het **vierde artikel** dit proces opnieuw bestudeerd. Weer wordt een beperkt aantal veelbelovende fabrieken ontworpen, met nu een variërende vergasserdruk, het gebruik van zuurstof dan wel lucht in de vergasser, en met verschillende opties voor het restgas. Er is een dynamisch model gemaakt, waarbij de invloed van iedere parameter op het eindresultaat (investering, efficiëntie, productiekosten) direct zichtbaar is. In fabrieken die bestaande processen toepassen en die een bescheiden schaal hebben, kan de diesel op korte termijn tegen ongeveer 16 €/GJ worden geproduceerd. Op de langere termijn (grote schaal, technologische ervaring, en verbeterde katalysator) zou dit tot 9 €/GJ kunnen dalen.

Tenslotte behandelt het **vijfde artikel** de productie van ethanol (alcohol) uit houtachtige biomassa door middel van hydrolyse en fermentatie. Een groot deel van houtachtige biomassa bestaat uit lange ketens van suikers. In het hydrolyse proces worden deze suikers vrijgemaakt, en in de fermentatie worden ze door gist of bacteriën in alcohol omgezet. Het artikel bestudeert de huidige processen en de technologie die op dit moment in laboratoria wordt ontwikkeld. Voor verschillende momenten in de tijd (nu, middellange, en lange termijn) zijn veelbelovende fabrieken ontworpen en ook weer geanalyseerd. Op korte, respectievelijk lange termijn kan biomassa voor 35 – 48 % in ethanol worden omgezet. Het gedeelte dat niet kan worden omgezet bestaat voor een groot deel uit lignine (geen suikerketen). Dit deel kan voor de coproductie van elektriciteit worden ingezet, zodat de totale efficiëntie dan 50 – 68 % wordt. Korte termijn (5 jaren) productiekosten zijn ongeveer 22 €/GJ. Door het gezamenlijke effect van efficiëntere organismen, lagere kapitaalsinvesteringen, grotere schaal en goedkopere biomassa (van 3 tot 2 €/GJ), zouden de productiekosten over 20 jaar 9 €/GJ kunnen zijn.

Terug naar het essay: daar worden de resultaten van de afzonderlijke brandstoffen gepresenteerd, zodanig dat een directe vergelijking mogelijk is. Op lange termijn zouden methanol en waterstof tegen 9 €/GJ kunnen geproduceerd worden. Ethanol en FT diesel zijn duidelijk iets duurder (11 en 13 €/GJ). In biomassaproducerende gebieden zoals Latijns Amerika of Oost Europa zouden de vier brandstoffen tegen 7 – 11 €/GJ geproduceerd kunnen worden. Deze kosten zijn boven de huidige brandstofproductiekosten die in de laatste tien jaar varieerden van 3 tot 7 voor benzine en van 2 tot 7 €/GJ voor diesel (1 €/GJ komt overeen met 0.0352 €/l benzine of 0.0377 €/l diesel). De onzekerheden in de productiekosten van de vier biobrandstoffen zijn 15 – 30 %, klein in het licht van de grote onzekerheden in toekomstige benzine/diesel prijzen.

De vier geavanceerde biobrandstoffen kunnen worden vergeleken met andere biobrandstoffen als (zoveel mogelijk) dezelfde aannamen worden gebruikt. De productie kosten voor DMM (dimethoxymethaan), DME (dimethylether) en synthetisch aardgas vallen in hetzelfde kostengebied. Biodiesel uit koolzaad en alcohol uit suikerbiet of graangewassen zijn betrekkelijk duur (25 – 40 €/GJ).

Voor een eerlijke vergelijking worden de brandstofprestaties op vergelijkbare basis uitgedrukt: hun kilometerkosten. Als je accijns en BTW op de brandstof niet meerekent, dan worden de kilometerkosten gedomineerd door de kosten van de auto. De brandstof veroorzaakt ongeveer 30 % in het geval van biodiesel in dieselauto's, en waterstof in brandstofcelauto's slechts ongeveer 9 %. Hoe duurder en hoe efficiënter een auto is, des te minder doen de brandstofkosten er toe. Op dit moment zijn biobrandstoffen een dure optie om CO_2 uitstoot te verminderen: het gebruik in auto's met verbrandingsmotor kost 100 – 300 €/Ton vermeden CO_2. Maar dat neemt af naar 50 – 100 €/Ton CO_2, een getal dat ook voor andere opties gevonden wordt. Deze kosten worden overigens meer veroorzaakt door de kosten van de auto, dan door de kosten van de brandstof.

Op grond van zowel de kilometerkosten als het potentieel en de kosten van CO_2 emissiereductie, komen brandstoffen uit houtachtige gewassen en uit suikerriet als besten naar voren. Toch is het met de huidige kennis en gegevenskwaliteit moeilijk om te zeggen welke brandstof op termijn absoluut het beste is. Uiteindelijk maken processen die op vergassing gebaseerd zijn een flexibelere productkeuze mogelijk dan processen gebaseerd op hydrolyse en fermentatie. Methanol, DME en synthetische diesel kunnen geleidelijk aan worden ingepast in de vorm van mengsels met benzine of diesel, en vereisen weinig infrastructurele aanpassingen. Rekening houdend met het ontwikkelen, ontwerpen, en bouwen van fabrieken en met de wettelijke procedures, kost de realisatie van een nieuwe fabriek minstens 5 à 6 jaar. Daarom kunnen alcohol gemengd in benzine en biodiesel gemengd in diesel nog tot 2010 de belangrijkste biobrandstoffen blijven. Alcohol/benzine mengsels zouden de ontwikkeling van betere ethanol processen, en de introductie van pure alcohol voertuigen later, kunnen vergemakkelijken. Biodiesel heeft dat voordeel niet. Op langere termijn zijn waterstof, methanol en DME het geschiktste in een overgang naar brandstofcel voertuigen. De uiteindelijke realisatie van een op waterstof gebaseerde transportsector, vereist zowel het ontwikkelen van nieuwe infrastructuur als het op de markt komen van de brandstofcelauto.

Om de vooruitzichten voor biobrandstoffen duidelijker te maken, moeten we zowel de onderdelen van de route die de biomassa aflegt, als de route als geheel, beter bestuderen. De conclusie van het inleidende essay somt aanbevelingen op, voor verder onderzoek en ontwikkeling. Op socio-economisch niveau betekent dit dat meer kennis nodig is van de duurzaamheid van grote plantages, van het ontstaan van een internationale markt voor bio-energie en de rol van handelsbarrières op deze markt, meer inzicht in het transitiepad of -paden naar een toekomst van biobrandstoffen, en in de invloed van overheidsmaatregelen en publieke perceptie op dit pad. Technisch-economische punten van aandacht zijn de maximum vergassergrootte, de ontwikkeling van doeltreffende gasreiniging, en het verbeteren van de eindgebruik vergelijking.

Biobrandstoffen zullen zeker een rol spelen in het toekomstige energieaanbod. Hoe zwaar deze rol zal zijn hangt af van het belang dat de samenleving hecht aan een zekere brandstofvoorziening, en van de wil om het versterkte broeikaseffect (als gevolg van menselijk handelen) tegen te gaan en CO_2 uitstoot te verminderen.

1 Introduction to biofuels

One condition for realising sustainable development is the establishment of a global sustainable energy supply [1]. The current energy supply system is responsible for several problems: exhaustion of fossil energy sources, an unacceptable supply security risk, local, regional and global environmental problems, including the emission of greenhouse gases affecting the earth's climate system. Furthermore, 2 billion people have no access to modern energy carriers. Solving these problems requires a new energy paradigm that considers the impacts of energy use at local and global scale, develops a wider portfolio of energy resources and cleaner technologies, widens access and increases efficiency, and concerns with both our present needs and the future generations' welfare [2]. Among others a shift should be made to the use of renewable energy sources such as wind, solar and biomass energy.

We confine this dissertation to analysing the potential of biomass energy. Bio energy is seen as one of the key options to mitigate greenhouse gas emissions and to substitute fossil fuels [1; 3]. Large-scale introduction of biomass energy could contribute to sustainable development on several fronts, environmentally, socially, and economic [4-6]. Sustainable growth of energy crops can improve degraded land soil quality, by sequestering carbon and restoring erosion damage. Plantations could create local employment in rural areas, and stimulate economic development. Eventually, biomass can be converted into modern energy carriers (liquid and gaseous fuels, steam and electricity) that can be used for heat and electricity consumption, and for transportation [3; 4; 7].

Abundant biomass resources are available in most parts of the world [8]. The present bio energy use covers 9 – 14 % of the global demand (about 400 EJ in 1998). Most of this use is non-commercial (estimated about 38 ± 10 EJ) for traditional, low-tech and inefficient cooking and heating in developing countries [3; 5]. Mostly in industrialised countries, modern production of energy carriers from biomass (electricity, transport fuels) contributes a lower, but significant 7 EJ [5]. Different global energy scenario studies indicate that in this century biomass may contribute much more to the world's energy supply: Some of the IPCC SRES marker scenarios attribute up to 30 % of the 2100 energy supply to biomass [9]. Analysing 17 studies, Berndes *et al.* found that the biomass contribution could strongly grow to an average 50 – 250 EJ/yr in 2060 [10]. The global (technical) potential of primary biomass in 2050, as discussed in the literature, ranges between 33 and 1135 EJ/yr [11], depending on population growth and food demand (diet), economic development, food production efficiency, energy crop productivity on various land types, competing biomaterial products, and land use choices. By assessing the land use and productivity on grid cell (0.5° by 0.5°) level, and application of IPCC SRES marker scenarios, this range may be narrowed to 300 – 675 EJ or 40 – 60 % of the energy demand in 2050 [12]. This amount could be produced on 4 – 10 % of the terrestrial surface.

Hoogwijk *et al.* [13] explore the 2050 regional and global cost-supply curves of energy crops, grown at abandoned agricultural and rest land, for four IPCC / SRES marker scenarios. They find that in the year 2050, 170 – 290 EJ_{HHV} may annually be produced at costs below 2 €[*]/GJ[†]$_{HHV}$. In Latin America, Africa, Asia and Eastern Europe significant amounts of biomass are already supplied at 0.5 – 2.0 €/GJ_{HHV} [4; 14-19]. For the first decade, forest or agricultural residues and waste will play an important role, as they are available in large amounts (30 – 90 EJ/yr in 2020 – 2050) at mostly low costs.

Currently available low cost biomass energy sources are being used for the production of modern energy carriers. In most cases this is still a local affair, but with increasing scale and decreasing production and transport costs, bio energy markets have emerged in e.g. Brazil, Sweden and Finland. Moreover, policy interventions combined with market dynamics may allow more expensive sources to be supplied over larger distances. Some world regions (like for example Latin America and Eastern Europe/Former USSR) have a much larger bio energy production potential than others, due to a combination of large land areas with good crop production potential, low population density and extensive agricultural practices. Therefore, various countries can become net suppliers of renewable bio energy, where others may be net importers [11; 20-22]. Several studies have given indications that intercontinental transported bulk wood could be delivered against acceptable prices and modest energy use [21; 23]. More and more biomass is already internationally shipped to consumers, which proves that international transport does not obstruct the large-scale implementation of bio energy.

Biomass can be converted into all main modern energy carrier types: heat, electricity and fuels for transportation ('biofuels'[‡]). The possibilities for and impacts of electricity and heat production from biomass (esp. combustion) are well known [4; 25] and widely applied in various markets. Furthermore, there are many other new or renewable technologies emerging for large-scale electricity production with low or no carbon emissions, such as (offshore) wind turbines and technologies for a cleaner use of fossil fuels. However, the transportation sector, which is almost entirely based on fossil fuel use (oil), has fewer alternatives.

Transportation represents about 27 % of the world's secondary energy consumption (21 % of primary). A share that may increase to 29 – 32 % in 2050 [9; 26; 27]. The increasing demand for transportation fuels combined with rapidly decreasing mineral oil reserves of non-OPEC states, implies that the availability of mineral oil depends on a limited number of countries [8].

Also, the transportation sector is often linked with local air pollution. Substitute use of some biofuels could reduce emissions, and individual biofuels may have specific environmental advantages. In this respect, however, modern reformulated gasoline and diesel do meet present strict requirements. Further improvement resides for a large part in engine technology.

Thus, it seems that biofuels are a sensible application of biomass. Moreover, they can play an important role in addressing many of the problems associated with mineral oil. Therefore, this thesis focuses on the potential of biofuels.

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[*] All costs in this introductory essay are in €$_{2003}$, inflation 2.5 % annually, 1 € = 1 US$.

[†] Energy is preferably expressed on Higher Heating Value basis, indicated by the subscript HHV. LHV indicates Lower Heating Value, if no subscript is given, the definition is unknown.

[‡] 'Biofuels' means liquid or gaseous fuel for transport produced from biomass [24].

2 Research focus and objectives

A few main routes can be distinguished to produce biofuels: extraction of vegetable oils, fermentation of sugars to alcohol, gasification and chemical synthesis, and direct liquefaction (see Figure 1). Many eventual fuels are conceivable: methanol, ethanol, hydrogen, synthetic diesel, biodiesel, and bio oil. All have very different properties (see Table A 1). Some of these fuels can be delivered to a central point or gas stations by existing infrastructure, while others need new tanker trucks or pipelines. Most biofuels are suitable for both present internal combustion engine vehicles (ICEVs) and future fuel cell vehicles (FCVs) using on board reforming. In some cases the fuel is direct suitable for the engine, but in most cases adaptations (fuel system materials, calibration) are necessary [28]. Tanking and on board storage (especially for hydrogen) may involve technologies that are not yet commercially available. Characteristics, advantages and disadvantages of individual fuels are summarised in Table 2.

2.1 Traditional fuels

Application of some biofuels is already reality (see Table 1). Extensive experience with alcohol (ethanol) use for transportation exists in Brazil, the USA, and some other countries. In Brazil, the cheaply available cane sugar enables a large and competitive ethanol fuel market: 11.3 % of the total primary energy consumption [30]. A National Alcohol Programme was created in 1975 (ProAlcool) to reduce oil imports, to protect the sugarcane plantation industry, to increase the utilization of domestic renewable-energy resources, to develop the alcohol capital goods sector and process technology for the

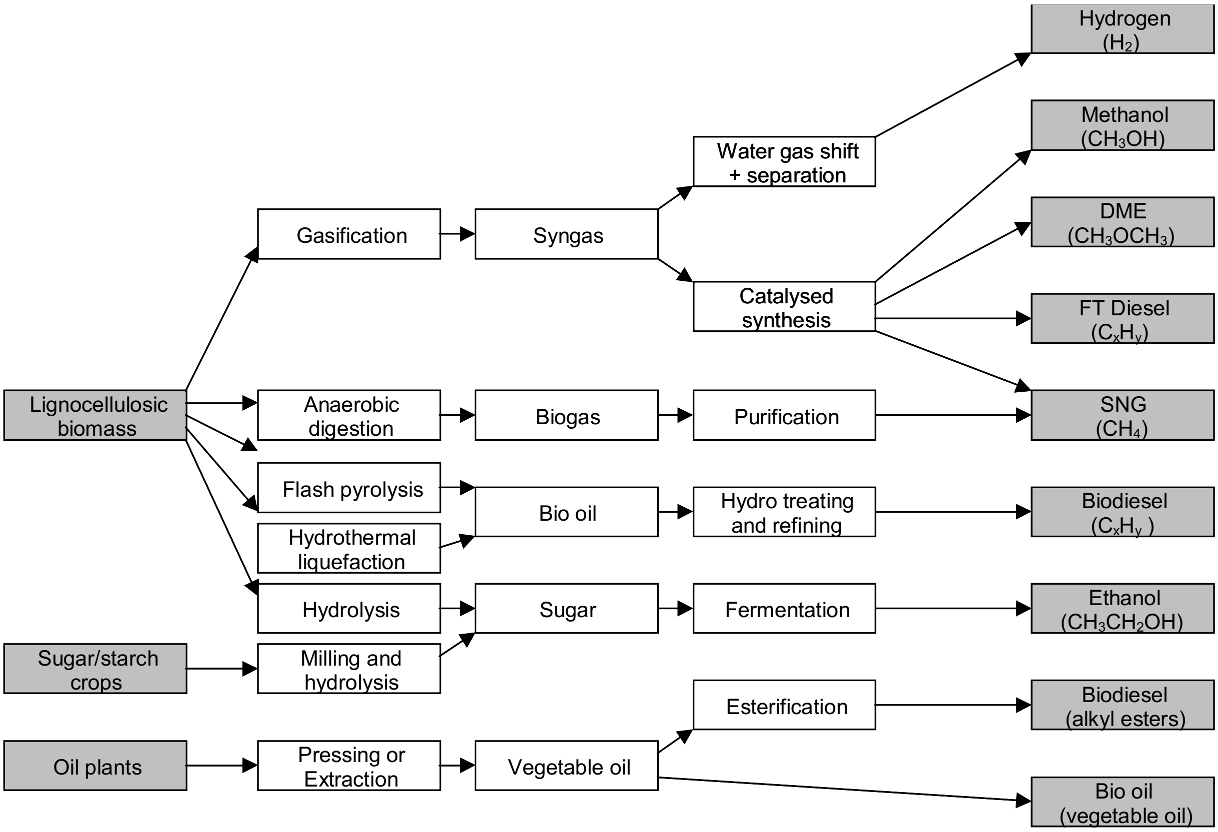

Figure 1. Overview of conversion routes to biofuels [4; 29].

Table 1. Volume and costs of present production of biofuels from different sources.

Biofuel	**Feedstock**	**Location**	**Scale** **(10^9 l/yr)**	**(PJ$_{HHV}$/yr)**	**Current costs**
Ethanol	Sugar cane1)	Brazil current	12 – 16	280 – 380	6.4 – 18 €/GJ$_{HHV}$
		Africa current	0.04	1	
	Maize2)	USA current	6	140	16 – 27 €/GJ$_{HHV}$
	Sugar beets and wheat3)	Europe current	0.27	6	25 – 39 €/GJ$_{HHV}$
Biodiesel4)	Oil seeds	Europe current	0.87	31	11 – 17 €/GJ
		USA current	1	36	

1) Sugarcane is the world's largest source of fermentation ethanol. It is one of the most photosynthetic efficient plants - about 2,5 % photosynthetic efficiency on an annual basis under optimum agricultural conditions [34]. In Brazil (and a few other countries) ethanol has historically been applied as motor fuel on a large scale. Throughout the eighties, over 40 % of the cars used "alcool" E93 (hydrous ethanol) in adapted engines. The rest used "gasohol" or E20 (20 - 24 % ethanol with gasoline), which is also called gasolina. Because of moving sugar and oil markets, and dried up subsidies, ethanol has become more expensive. In the nineties, gasohol is sometimes more like E10 and also the composition of alcool changed [33; 35]. Nevertheless, there are still 3 million cars running on neat ethanol. Ethanol market price in Brazil depends on both oil and sugar market [31; 35]. Alcohol fuels have also been pursued in a number of sugar producing African countries (esp. Zimbabwe, further Kenya, Malawi, and South Africa [34]).

2) The USA produces annually $6 \cdot 10^9$ litres bio ethanol from maize [34]; use is especially as 10 % blend in gasoline (E10). The estimated costs are 381 – 630 €/m^3 from grains (feedstock with/without subsidies) [29].

3) Ethanol production in EU (esp. France and Spain) increased from 48 ktonne in 1993 to 220 – 260 ktonne in 2003 [36; 37]. Starting from 2008 Germany plans to mix 8 % ethanol in gasoline (requiring annually $4.5 \cdot 10^9$ litres), Ethanol from grains production in Europe could be $7 \cdot 10^9$ l/yr [29]. Estimated costs are 590 – 920 €/m^3 from grains in Europe (feedstock with/without subsidies). Ethanol production is 0.32 – 0.37 litre ethanol/kg grain [29]. In Sweden, ethanol is used as 5 % and 85 % blend in gasoline (E5 and E85) [37].

4) The total transesterification capacity in the European Union (esp. France and Germany) is about 1 Mtonne/yr RME [36]. In the USA, at least five companies produce biodiesel from soybeans at commercial basis. Methyl esters require substantial subsidies to compete with diesel. Subsidies in Europe generally consist of a combination of agricultural subsidies and fuel tax exemption. Pure biodiesel is used as fuel in Germany and Austria, a 5 % blend with diesel is used in France [37; 38].

production and utilization of industrial alcohols, and to achieve greater socio-economic and regional equality through the expansion of cultivable lands for alcohol production and the generation of employment [30-32]. Ethanol is produced from maize (corn) in the USA, and on much smaller scale from wheat and sugar beets in Europe [33]. Biodiesel (methyl ester) is produced from rapeseed in Europe (especially Germany and France) and from soybeans in the USA.

The EU production and use of biofuels rapidly increased over the past 10 years: biodiesel ten folded and ethanol almost five folded. The current European biofuels contribution of some annual 37 PJ is however still negligible in the total European bio energy use (1.9 EJ) [36]. The European Union has recently set a 2 % biofuels (by energy) target for 2005, and 5.75 % in 2010 [24]. Germany plans to introduce 8 % alcohol in gasoline (in 2008) and Japan considers introducing 5 % alcohol in gasoline.

2.2 Advanced fuels

With the exception of sugarcane-ethanol, the 'short term' [37] or 'traditional' [5] biofuels have a number of severe disadvantages that are related to the feedstock. These feedstock (sugar, starch and oil crops) are in fact classic agricultural feed crops, that require high quality (valuable) agricultural land for growth. A lot of the captured sunlight is diverted to produce the seeds or store the sugar, contrary to perennial crops and grasses, and the growing season is short. The net energy yield of most annual crops (100 – 200 GJ/ha.yr on long term), therefore, is much lower than that of perennial crops (220 – 550), grasses (220 – 260) and sugarcane (400 – 500) that can be grown on less valuable land [8]. Furthermore, only part of the plants can be used for fuel production. As a result, traditional biofuels have a meagre CO_2 emission reduction potential. Moreover, their economies are poor and unlikely to reach competitive cost levels. Also the environmental aspects of annual crops are not good: they cause much more erosion (up to 100-fold) and require 7 – 10 fold pesticides compared to perennial grasses or wood [4; 10; 39-41]. Compared with sugar, starch, and oil crops, application of lignocellulosic biomass (e.g. wood and grasses) is more favourable and gives better economic prospects to the future of biofuels. Also, more types of feedstock are in principle suitable to produce a broader range of fuels than when applying traditional biofuels feedstock.

Obviously, fuels that most resemble gasoline and diesel (esp. synthetic diesel, but also HTU diesel and synthetic gasoline) are the most suitable for application in the current ICEVs. Up to a limited fraction, biodiesel, alcohols and ethers can be blend without significant difficulties in diesel and gasoline respectively. Very efficient fuel cells are candidate technologies for driving future cars. Hydrogen is the most suitable for fuel cells, but hydrogen onboard storage is still troublesome. Methanol and DME are good hydrogen sources, and much more suitable for FCVs than other hydrocarbon fuels (see later).

Infrastructure adaptations are needed for introduction of most biofuels. Fuels could be blended (if applicable) at the refinery, at the gas station or at intermediate depots. Blending at the refinery requires the least logistic adaptations, but requires a constant quality (non deteriorating) and a large scale [37]. Some fuels require extra lining or safety precautions in pipelines or road tankers, depending on the fuel's characteristics. Gasses are difficult to transport and bulky to store, although SNG could be distributed by the natural gas grid. Hydrogen in its pure form is first distributed by new or converted pipelines to depots, and further by road tanker. At the gas station a new dispensing system is required for all fuels except the diesels and blends [42].

It appears that some fuels are more suitable at present because of implementation ease, and others are more suitable for the long term, because of ultimate performance. The transformation of the current fuel supply and use system into a future one, may also be run via the introduction of advanced engines fuelled by existing fuels, or flexible fuels vehicles (FFVs) that usually can be fuelled by varying combinations of two fuels (e.g. different blends of gasoline and M85[§]). On the other hand, FFVs cannot obtain the low energy consumption levels and low emissions of dedicated engines because they have to function for all blends [28].

Ethanol seems an attractive fuel today: It can be blended to a fair percentage in gasoline, and production is likely to be attractive already at small scale, and existing ethanol can be complemented or gradually replaced by lignocellulosic ethanol. For the short- and middle-term it seems that also Fischer-Tropsch diesel is interesting because it combines direct implementability with gradual enlargement of both utilization (increasing fraction) and production scale. Also other gasification-based fuels can benefit from development of the Fischer-Tropsch process. Apart from all difficulties that the various

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[§] M85: the capital M indicates methanol, the number the percentage of methanol (by volume) in blend with gasoline. In like manner, blends with ethanol are indicated by the capital E followed by two digits.

Table 2. Overview advantages and disadvantages of individual biofuels: suitability for internal combustion engine vehicle (ICEV) or fuel cell vehicle (FCV), applicability of existing fuel supply infrastructure, ease of car storage, safety, experience, R&D status and potential success.

Biofuel	**Suitability for**		**Infra and storage**	**Safety and environment**	**Status**	**Projected cost (€/GJ) (short → long term)**
	ICEV	**FCV**				
Methanol[1]	~	++	· Severe infra adaptations · Larger tank	· Strict handling, avoid skin contact or spill · Toxic, but less than gasoline · Inflammable · Emissions decrease · Gasoline VOC (in blends) may increase	· Production technically feasible · Commodity (from NG) · Can be blended with gasoline in any ratio · Some vehicle tests	6 – 20
Ethanol[2]	+	+	· Small infra adaptations · Larger tank · Filters needed	· Biodegradable · Emissions decrease · Gasoline VOC (in blends) may increase	· Commodity (from sugar/starch) · Can be blended with gasoline in any ratio · Vehicular (ICEV) experience	21 – 40 (wheat / beet) 8 – 11 (sugar cane) 13 → 5 (cellulose)
MTBE and ETBE[3]	++	~	· Existing	· Decrease smog · MTBE may pollute groundwater	· Only use as additive · Existing business (production and use)	14 – 23
Hydrogen[4]	~	++	· New infra · Storage problematic · Range may be limited	· Explosive, though hydrogen storage systems can be engineered to be as safe as the current fuel systems	· Technically feasible · Commodity (from NG) · More renewable sources possible · Many vehicle tests	5 – 16
Synthetic diesel[5]	++	+	· Existing	· Cleaner than diesel	· Commodity (from coal and NG)	22 → 9
Synthetic gasoline[6]	++	+	· Existing	· Cleaner than gasoline	· Commodity (from NG via Methanol)	unknown
Biodiesel (oil seeds)[7]	+	-	· Existing · Short shelf life	· Safe · Biodegradable	· Existing business (production and use) · Requires large agricultural plots · Low net lifecycle energy yield	15 – 29
Bio-oil (oil seeds)[8]	~	-	· Existing	· Safe · Biodegradable	· Demonstration · Requires large agro plots	unknown
Bio-oil (pyrolysis)[9]	-	-	· Difficult	· Corrosive · Carcinogenic	· Several pilot plants · Intermediate bio-energy carrier · Probably not suitable as motor fuel	6 – 10
Biodiesel (HTU)[10]	+	-	· Existing	· As diesel	· Successful pilot plant · Requires wet biomass · Co-produces 'pure' CO_2	16
DME and DMM[11]	+	++	· Small infra adaptations	· Safe	· Commodity (from NG)	16
SNG[12]	+	-	· Difficult · By existing NG grid	· Explosive	· Small stationary applications	13

[1] Methanol can be mixed in gasoline up to 10 % without adjustments to the motor or fuel system. Larger percentages in spark-ignited engines may cause cold start problems (require preheating). Flexible Fuel Vehicles, or FFVs run on any blend of M85 and gasoline. Methanol cannot be mixed with diesel, application of neat alcohols in compression-ignition (CI) engines is difficult. Methanol may be the ideal hydrogen carrier for FCVs: it can be steam reformed at moderate temperatures. Alcohols affect lubricating oil, some plastics and metals. Methanol is biodegradable and spills last shorter than gasoline spills, but it is water-soluble and large spills may sterilize the soil. It is easy inflammable and burns with a nearly invisible flame, though fires are less severe than gasoline fires. It is toxic, but gasoline is overall more hazardous to human health. Alcohols increase the vapour pressure of gasoline in low concentration blends, which may require VOC to be removed from gasoline [28; 29; 33; 43-45].

[2] Neat, hydrous ethanol (with < 20 vol% water) can be used in special engines. Blending anhydrous ethanol (0 % water; requires azeotropic distillation or dehydration) with gasoline has less start problems. Normal vehicles can cope with up to 22 % alcohol in gasoline without motor or fuel system adjustments. Flexible fuel vehicles can process any blend of E85 (85 % ethanol with 15 % gasoline) with gasoline. As hydrogen carrier for FCVs, ethanol requires reforming at temperatures in excess of 500 °C. Existing infra can be used for distributing ethanol, but many filters are necessary to remove the dirt that is dissolved by alcohol. Lining, stainless steel, and special plastics that resist alcohol are required in tanks, pipes and fuel system [28; 29; 46].

[3] Ethyl tert butyl ether (ETBE) and methyl-tert-butyl ether (MTBE) are obtained from a reaction between isobutene and respectively methanol and ethanol. They are better gasoline substitutes than alcohols because their characteristics are closer to gasoline. MTBE (max 15 %) and ETBE (max 20 %) are often added to gasoline as oxygenates, obviating the octane number loss from aromatic and lead reductions in petrol, and reducing urban carbon monoxide emissions in winter; they are not used as fuels themselves. Since isobutene is derived from natural gas or mineral oil, ETBE and MTBE can only partly be biomass based: ETBE 45 % by mass and MTBE 35 % by mass. At present, MTBE is under debate because of ground water contamination risks [33; 43; 45].

[4] Hydrogen can be used in a spark ignition engine as a clean fuel, but requires some engine adaptations. NO_x emissions are low, and the fuel economy is high. Disadvantages may be power loss, back firing and knock. Hydrogen is widely seen as the ultimate transportation fuel to be used in FCVs. Transport, distribution and storage of pure hydrogen may be difficult and expensive (low volumetric energy density). Storage options under research are liquefied storage, in a metal hydride, or as compressed gas. The latter is the option favoured at present. Hydrogen is highly inflammable (low ignition energy and high flame propagation speed), making safety a recurring discussion issue, but with safe refuelling systems and strong storage tanks hydrogen use can be as safe as LPG or natural gas. Hydrogen could also be produced by electrolysis using green electricity (solar, wind), or by photo biological processes in algae or bacteria (at 15 and 25 €/GJ_{HHV} respectively) [28; 29; 33; 46-49].

[5] Synthetic diesel is successfully produced by the Fischer-Tropsch (FT) process from coal synthesis gas (gasification) in South Africa, and can thus also be produced from biomass via gasification. The FT process gets growing attention in recent years, especially as solution for handling associated and stranded gas. Synthetic diesel, has a very high cetane number, is fully fungible with conventional diesel [42; 50], and is thus directly usable in the present day transportation sector (ICEV CI). Furthermore, synthetic diesel is very suitable – via onboard reforming – for fuel cell vehicles, because it is very low in fuel cell catalyst poisons (sulphur).

[6] Premium gasoline can be produced synthetically from methanol by the Mobil MTG (Methanol-To-Gasoline) process [51]. The capital costs are high compared to methanol plants, since extra process units are added. As solution to stranded gas, it gets less attention than FT diesel or DME.

[7] Biodiesel properties and energy content are similar to fossil diesel and, therefore, it can be used neat or as blend in most diesel engines. It is simple to use, (fast) biodegradable, non-toxic, and essentially free of sulphur and aromatic compounds. Fuel efficiency is about the same as for petroleum diesel, or higher. On the other hand, it has poor cold start and low temperature characteristics, and a low stability (maximum 5 months shelf life). Rapeseed Methyl Ester (RME) production is established technology in the EU. The land area requirement is a serious drawback to large-scale application of biodiesel [28; 33; 52].

[8] Rapeseed oil could, in a small fraction, be added to diesel, or added to crude oil (before distillation). Addition of bio-oil to diesel is limited to a few percent in winter because of cold start problems; in summer, the fraction could be up to 20 %. A few demonstration trucks run on pure plant oil [28].

[9] Several tests have been done with pyrolysis oil in stationary diesel engines (for electricity), with promising results, although corrosion and soot problems remain to be solved. As yet, it is not suitable as transportation fuel: it contains too much oxygen and water and the H/C ratio is too low, hydro-deoxygenation requires a disproportional large amount of hydrogen. Further disadvantages of pyrolysis oil are mainly in its broad compound spectrum (several hundred different chemicals, among which acids and aromatics), its acidity, its immiscibility with petroleum fuels, and its chemical instability (decays to char) [5; 50; 53; 54].

[10] The Hydrotermal upgrading product biocrude is a mixture with a wide molecular weight distribution and consists of various kinds of molecules. It may be produced competitive with fossil crude oil, and may easily be converted to diesel, although still significant amounts of hydrogen are required. Compared to pyrolysis, more of the oxygen is removed in the process (as CO_2). Additional research is required to decrease the amount of co-produced tar/char, and decrease the costs [42; 55; 56].

[11] Dimethyl ether (DME) can be produced from methanol or (large scale) directly from synthesis gas slightly more efficient than methanol. DMM (dimethoxymethane) is produced from syngas. They can be used in a diesel engine and feature higher fuel efficiency and lower emissions (higher cetane number) than diesel. DME does not corrode metals, but may affect certain plastics. With small engine and fuel system adaptations, DME can be used in blends with diesel (10 or 20 %), with LPG (any %) and neat. Since DME is as easily reformed as methanol, it has a big potential for fuel of fuel cell automobiles in future. DME and DMM are not toxic for man, not carcinogenic, although irritating. DME burns safer than methanol (visible flame). DME vehicles may be slightly safer than LPG vehicles [28; 37; 42].

[12] Substitute or Synthetic Natural Gas (SNG) can be produced by hydro gasification of biomass and subsequent methanation of the residual CO in the syngas, or from purification of biogas (mainly methane and carbon dioxide) from anaerobic digestion [57].

fuels face, and the many different indicators one can use to compare them, higher overall (production, distribution and use) energy conversion efficiencies and lower overall costs are the key arguments for selecting biofuels for the longer term. In this respect, methanol, hydrogen and DME used in fuel cell vehicles, may perform best [5; 42; 46; 58]. The research, development and demonstration status of all these fuels varies considerably. Four fuels from lignocellulosic biomass – methanol, ethanol, hydrogen, and synthetic diesel – are selected for a detailed analysis.

2.3 Production of the selected biofuels

Methanol, hydrogen and Fischer-Tropsch diesel can be produced from biomass via gasification (biomass is more reactive than coal and thus easier to gasify [46]). Several routes involving conventional, commercial, or advanced technologies under development, are possible. Figure 3 pictures a generic conversion flowsheet for this category of processes. A train of processes to convert biomass to required gas specifications precedes the methanol or FT reactor, or hydrogen separation. The gasifier produces syngas, a mixture of CO and H_2, and few other compounds. The syngas then undergoes a series of chemical reactions. The equipment downstream of the gasifier for conversion to H_2, methanol or FT diesel is the same as that used to make these products from natural gas [46], except for the gas cleaning train. Optional are a gas turbine or boiler to employ the unconverted gas, and a steam turbine; resulting in electricity co-production.

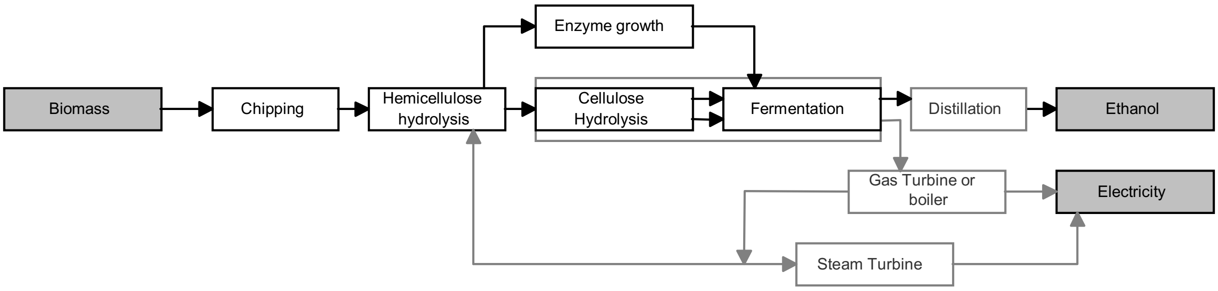

Figure 2. Ethanol production by hydrolysis fermentation schematically

Ethanol, instead, is produced via (largely) biochemical processes. Biomass is generally pretreated by mechanical and physical actions (steam) to clean and size the biomass, and destroy its cell structure to make it more accessible to further chemical or biological treatment. Also, the lignin part of the biomass is removed, and the hemicellulose is hydrolysed (saccharified) to monomeric and oligomeric sugars. The cellulose can be hydrolysed to glucose. The sugars are fermented to ethanol, which is to be purified and dehydrated. Two trends are possible for future processes: a continuing consolidation of hydrolysis-fermentation reactions in fewer reactor vessels and with fewer micro organisms, or an optimisation of separate reactions. As only the cellulose and hemicellulose can be used in the process, the lignin is used for power production.

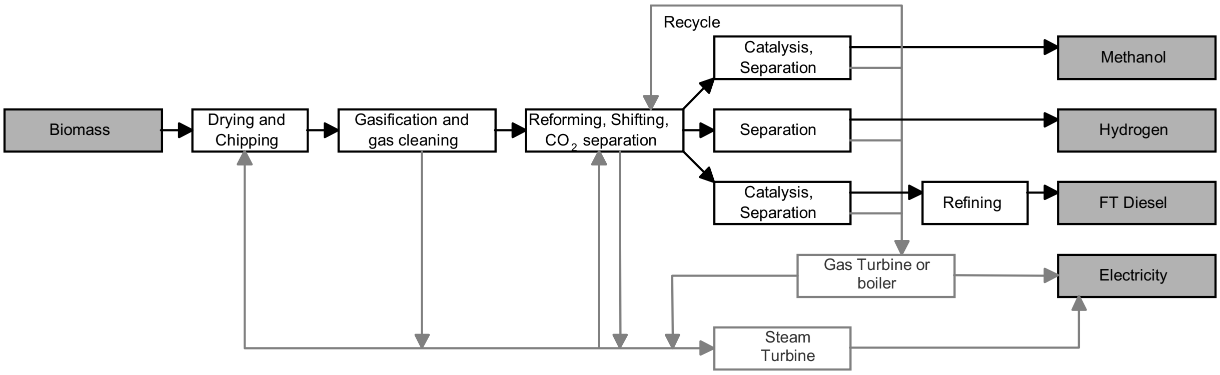

Figure 3. Generic flowsheet for methanol, hydrogen, or FT diesel production, via gasification of biomass.

2.4 Status of knowledge

Several studies exist that provide an overview of (part of) the biofuels field [37; 40; 42]. Other studies present the techno economic performance of individual biofuels [46; 58-60]. However, often these studies have been done only for small biomass input scales, because authors assume that large scale is a priori not feasible, and for existing technologies. The potential for a better performance – that could be obtained by applying improved or new (non commercial) technologies, combined fuel and power production, and increasing scale giving higher efficiencies and lower unit capital costs – has not exhaustively been explored.

Another problem is the comparability of the results. The capital analysis for biofuels producing facilities has been done in different ways. The data quality is very variable. Also, the level of detail in analyses varies enormously: from very superficial, to thorough plant analysis. In either case the influence of individual parameters (e.g. feedstock costs) on the final product price is unclear.

It can be concluded, that most existing biofuels do not perform well with respect to costs and energy efficiency. A set of advanced biofuels is expected to perform much better, but the studies that have been done so far are not comparable and do not give insight in the potential of further improvement, and in what technological or economic barriers hamper the realisation of good performance levels.

The biomass feedstock costs are a major input parameter for the calculation of the biofuel production costs. The large-scale international transport from bioenergy producing regions to Western Europe can be done via a broad variety of chains, comprising different biomass production systems, pretreatment and conversion operations, and transport of raw (chips, logs, bales) and refined biomass (pellets) by different means (truck, train, ship).

A few earlier studies have given indications that biomass could be delivered from Eastern Europe or Latin America at 4 – 6 €/GJ_{HHV}. The energy consumption for transportation may vary from modest (4 % of energy delivered) to considerable (20 %) [21; 23]. These figures are influenced by many factors that deserve more detailed analysis, especially the costs and energy use of components and the logistic organisation of supply chains, such that further optimisation of costs and efficiency is possible. Moreover, these studies focus on the short term and narrowly defined cases of biomass transport, while the long-term large-scale bio energy supply would be of interest. Key parameters such as distance, changing biomass characteristics during a supply chain, scale and timing (supply pattern) are expected to influence the overall performance, and should be explicitly assessed.

2.5 Objective of this thesis

The main research questions of this dissertation are: Which of the biofuels options have the better potential for the near term and which the best ultimate (2030) prospects? And what developments are necessary to improve the performance of biofuels production and use?

The main objective therefore is to systematically analyse the short- and long-term technological and economic performance of biofuels. This is done by assessing the four selected fuels: methanol, ethanol, hydrogen, and synthetic diesel. First, the key technologies for the production of these fuels, such as gasification, gas processing, synthesis, hydrolysis, and fermentation, and their improvement options will be studied and modelled. Then, the production facility's technological and economic performance is analysed, applying variations in technology and scale. Finally, major biofuels chains (including distribution to cars, and end-use) will be compared on an equal economic basis, such as distance costs (€/km) and CO_2 emission reduction costs (€/tonne$_{CO2}$).

It will be assessed which factors most influence the fuel production and fuel chain's performance, and which aspects are uncertain. This gives insights both in the possible barriers to implementation that need to be overcome, and in the technological improvement options that should be stimulated by Research Development and Demonstration.

A major sub-question is, whether the required large-scale long-distance transport is economically and energetically feasible. The objective is to analyse the costs and energy use of international logistic chains, and the logistic organisation of these chains.

3 Research method

3.1 Large scale bioenergy supply by international transport

A spreadsheet was developed to consistently compare a large variety of chains and assess the influence of the key parameters on cost and energy use (Paper 1). Detailed generic data was used for the technological and economic performance of equipment, conversion facilities and transport means. Biomass properties (e.g. chemical composition, density, moisture content) are changed by each conversion or operation, and influence the performance of the next step in the chain. Also matter losses and decay along chains were accounted for.

3.2 Producing methanol, ethanol, hydrogen and FT diesel

To analyse the prospects of each fuel, first a technology review was made, including existing, improved as well as new technologies within the timeframe 2000 – 2030. All the possible components were analyzed in terms of development status, costs and performance (especially efficiency). This review was used for composing and selecting promising conversion concepts. These systems were designed and modelled. The results on the technical performance were used for economic calculations.

In the production of methanol, hydrogen and synthetic diesel, many gasification technologies are or come available (atmospheric or pressurized, with air or oxygen, direct or indirect). Different types of gas cleaning could be applied. Gas conditioning by reforming and or shift can be applied to different extents. Different reactor types are available for gas separation and fuel synthesis.

In the production of ethanol, there are different technologies available or emerging for pre-treatment (hemicellulose hydrolysis) and cellulose hydrolysis. The hydrolysis enzyme can be supplied externally or be produced by using part of the feedstock. An increasing integration of functions in less reactors may be possible through time and separate (microbial) conversion efficiencies will increase. In many cases, off gas or residues allow for electricity production by a (gasifier and) combined cycle.

After reviewing the processes and technology, promising configurations for further analysis were selected. For the production of methanol and hydrogen (Paper 2), a few concepts were chosen based on expected high performance. Some concepts use conventional technology as it could be applied today, and some use advanced technology to study the long-term perspectives. The same approach was applied in exploring the possibilities for Fischer-Tropsch diesel production, but with more concepts (Paper 3). That study was of explorative nature and proposed many options that could further improve the performance of Fischer-Tropsch diesel production. Continuing on the most promising of the concepts, the influence of a number of parameters on technical and economic performance was evaluated (Paper 4). For ethanol production, the conversion technology and projected technology development led to a short-term, middle-term and long-term concept (Paper 5).

For analysing the production of methanol, hydrogen and FT diesel, detailed Aspen Plus [61] flowsheet models were made and optimised. The gasifier, reformer and gas turbine deliver heat, whereas the dryer, gasifier, reformer, and shift require steam. The supply and demand of heat is added to or drawn from the steam turbine, such that the surplus heat is turned into electricity.

Ethanol production was for the greater part modelled in the spreadsheet program Excel (except for the power isle). For each process step in the hydrolysis fermentation process, conversion extents and losses were applied, so that each step yielded intermediate amounts of sugar, ethanol and solid residuals.

The mass balances from the technical performance calculations were used for economic evaluation. For methanol and hydrogen, this was done after the technical calculations, for FT diesel and ethanol this was done concurrent. Fuel production costs are calculated by dividing the total annual costs of a system by the annually produced amount of fuel. The total annual costs consist of annual capital costs, operating and maintenance (including maintenance, consumables, labour, waste handling), biomass feedstock costs and costs of electricity supply / demand (fixed power price). The total capital investment, or TCI, is calculated by *factored estimation* [62], based on known costs for major equipment as found in literature or estimated by experts. The uncertainty range of such estimates is up to ± 30 %.

The fuels produced have been analysed by applying different degrees of detail. This eventually yields different qualities of results and insights. The methanol and hydrogen concepts were individually modelled and included far reaching heat integration. The model of the second Fischer-Tropsch study allowed for far going optimisation, by variation of gasifier oxygen level and pressure, and applying many step-by-step process configuration changes. In the Fischer-Tropsch process, pressure, temperature, and (relative) concentration of the reactants influence the product quality and yield, and this was also accounted for. The economic performance of each variation was directly visible and accounted for in the optimisation. The methanol and hydrogen concepts were rather chosen for their expected technical performance, after which they were modelled and optimised for efficiency, and eventually economically evaluated. The chosen ethanol concepts did not have much room for optimisation. Also, they were analysed at a lower level of detail than the other fuels.

Finally, several input parameters were slightly different between the separate papers, which may complicate direct comparison. Therefore, the results, summarized in the next section of this introductory essay, have been recalculated from the separate studies by using a unified set of parameters (Table 3).

Table 3. Unified set of input parameters.

Scale	400 → 2000 MW_{HHV} input (Short → long term)
Electricity price (supply and demand)	0.03 €/kWh_e
Economic lifetime (depreciation time)	15 years
Technical lifetime	25 years
Interest rate	10 %
Load	8000 h (91 % of time)
Investment path	20 % in first year, 30 % in second and 50 % in last year

4 Results and discussion

4.1 Feedstock costs

The delivered costs for biomass feedstock has been analysed in Paper 1. This paper shows that 300 MW_{HHV} (8.6 PJ_{HHV}) solid biomass in compressed form (pellets or bales) could be delivered to a Western European location from Latin America (11,000 km sea transport) at about 3.1 €/GJ_{HHV}, adding 2 €/GJ_{HHV} to the local production costs of 1 €/GJ (Paper 1), only 0.5 €/GJ_{HHV} of this is in the actual international shipping. From future Eastern European plantations (production costs 1.5 €/GJ), the delivery costs are 3.6 €/GJ_{HHV}. Scale enlargement to e.g. 1200 MW_{HHV} barely decreases costs further, because the most important operations have already past their maximum unit size. Further calculation in this section assumes that biomass in compressed form can be delivered in Western Europe at 3 €/GJ_{HHV}. In the country of biomass production, biomass can be delivered to local plants at 2 €/GJ_{HHV}.

4.2 Selected biofuels

The short- and long-term technological and economic performance of the four selected fuels – methanol, ethanol, hydrogen, and synthetic (FT) diesel – has been analysed in Papers 2 – 5. Table 4 gives the resulting key parameters for biomass to fuel conversion facilities on short- and long-term, the latter include process improvements and technological learning. At 400 MW_{HHV} input, the short-term investments range from 240 to 290 M€.

Table 4. Technological and economic performance of biomass to fuels facilities for now → future: Efficiencies to fuel and electricity, capital investment, scale factor, annual O&M costs, and fuel production costs are summarized and recalculated from Papers 2 – 5. Parameters hold at 400 MW_{HHV} biomass input. The production costs for the future include a larger scale (2000 MW_{HHV} input). Electricity buy/sell costs 0.03 €/kWh_e. Delivered feedstock costs 3 €/GJ_{HHV} (Western Europe), or 2 €/GJ_{HHV} (local in biomass producing region). The processes assume wet (30 % moisture) chipped biomass, drying to 10 – 15 % and pulverisation are included in the concepts.

Fuel		**η_{HHV} 1)**		**TCI 2)**	**R 3)**	**O&M 4)**	**Production costs 5) (€/GJ_{HHV})**		
		Fuel	**Electricity**	**(M€)**		**(% of TCI)**	**Now**	**→ future**	**Local future**
Methanol	now 6)	58.9 %	-4.0 %	235	0.79	4.0 %	12		
	future 7)	57.0 %	-0.1 %	188	0.84	4.0 %		9	8
Ethanol	now 8)	34.9 %	4.1 %	291	0.84	6.4 %	22		
	future 9)	47.3 %	4.0 %	218	0.82	3.6 %		11	9
Hydrogen	now 10)	34.8 %	16.9 %	247	0.81	4.0 %	16		
	future 11)	41.3 %	19.7 %	207	0.86	4.0 %		9	7
FT diesel	now 12)	42.1 %	3.2 %	292	0.85	4.4 %	18		
	future 13)	42.1 %	3.2 %	235	0.85	4.4 %		13	11

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1) Electricity is co-produced in most processes (Paper 2 also shows methanol concepts co producing electricity). Some processes require extra electricity.

2) From the TCI follows the TCR assuming a correction for lifetime (90.4 %) and investment path (20 %, 30 % and 50 %, in first, second and last year: 118 %). The methanol and hydrogen study (Paper 2) and the Fischer Tropsch study (Paper 4) did not include an investment path; the here presented values are therefore somewhat higher. The TCR is used for determining the annual capital costs.

3) R value found for up scaling from 400 to 2000 MW_{HHV} input, smaller R are found for downscaling.

4) O&M for the methanol and hydrogen processes is fixed at 4 %. In the Fischer-Tropsch process, O&M consists of a fixed part (4 % of TCI) and a part decreasing with scale (0.4 % at 400 MW_{HHV}, R = -0.85). O&M in ethanol production is very dependent on cellulase required.

5) The time path also incorporates a scale increase: now: 400 MW_{HHV} and future: 2000 MW_{HHV}.

6) Methanol now applies an atmospheric indirect gasifier, wet gas cleaning, steam reforming (partly fed by off gas), shift reactor, low pressure gas phase methanol reactor with recycle, and a steam turbine (methanol concept 6 in Paper 2).

7) Methanol future applies an atmospheric indirect gasifier, wet gas cleaning, steam reforming (partly fed by off gas), a liquid phase methanol reactor with steam addition and recycle, and a steam turbine (methanol concept 4 in Paper 2, with 15 % cost reduction through learning).

8) Ethanol now applies dilute acid pre-treatment, on-site enzyme production, enzymatic cellulose hydrolysis, SSF configuration (cellulose hydrolysis and C6 fermentation integrated in one reactor vessel), boiler and steam turbine (ethanol short-term in Paper 5)

9) Ethanol future applies liquid hot water pre-treatment, CBP configuration (enzyme production, enzymatic cellulose hydrolysis and co-fermentation in one reactor vessel), boiler and steam turbine (ethanol long-term in Paper 5)

10) Hydrogen now applies an atmospheric indirect gasifier, wet gas cleaning, shift reactor, pressure swing adsorption for H_2 separation, and a combined cycle (hydrogen concept 5 in Paper 2).

11) Hydrogen future applies a pressurised direct oxygen fired gasifier, hot gas cleaning, ceramic membrane with (internal) shift, and a combined cycle (hydrogen concept 3 with 15 % cost reduction through learning).

12) FT diesel now applies a direct 25 bar oxygen fired gasifier, a tar cracker, wet gas cleaning, no reforming, and once through FT synthesis at 60 bar with 90 % conversion (Paper 4).

13) FT diesel future is same as previous, but with 15 % and 5 % cost reduction (learning + process improvement).

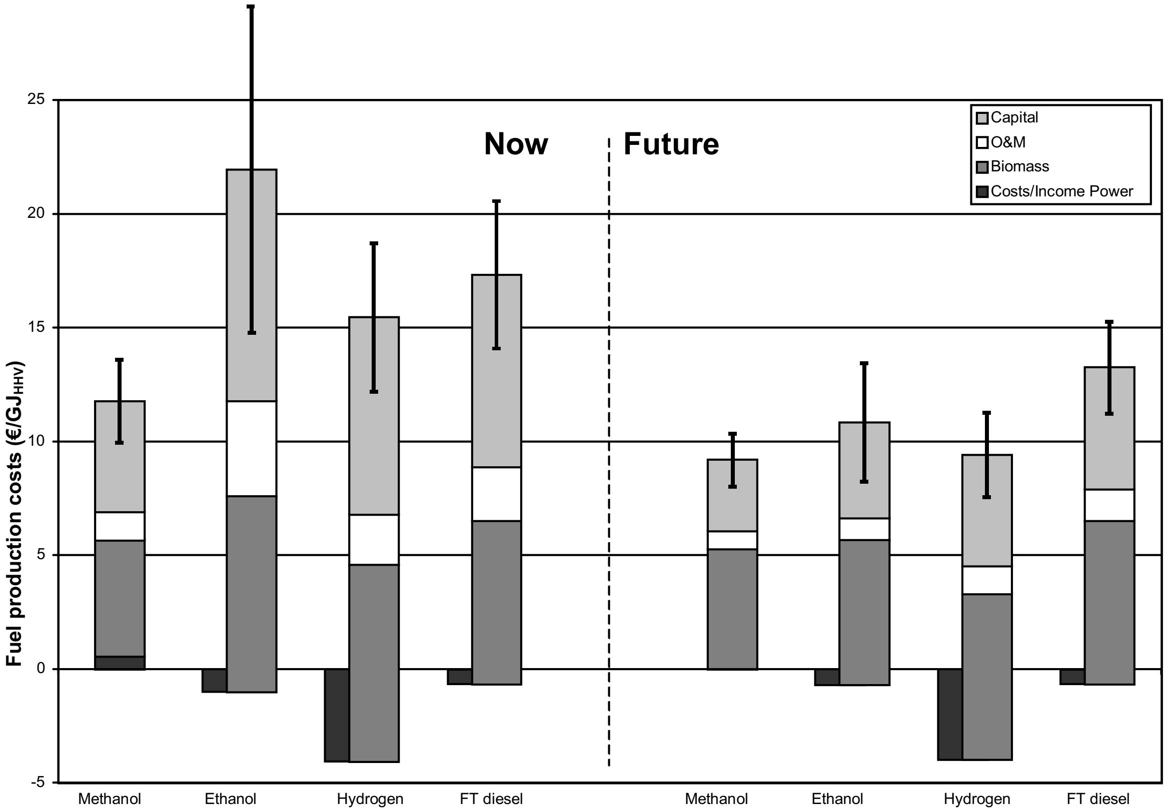

Figure 4. Breakdown of the production costs of selected biofuels (methanol, ethanol, hydrogen and FT diesel) now and in future. Feedstock costs 3 €/GJ$_{HHV}$. Time path also incorporates a scale increase: now 400 MW$_{HHV}$, middle term: 1000 MW$_{HHV}$, and ultimate: 2000 MW$_{HHV}$. Uncertainty ranges of 30 % are applied to capital (and O&M, because this is a linear function of capital), 50 % for the ethanol concepts.

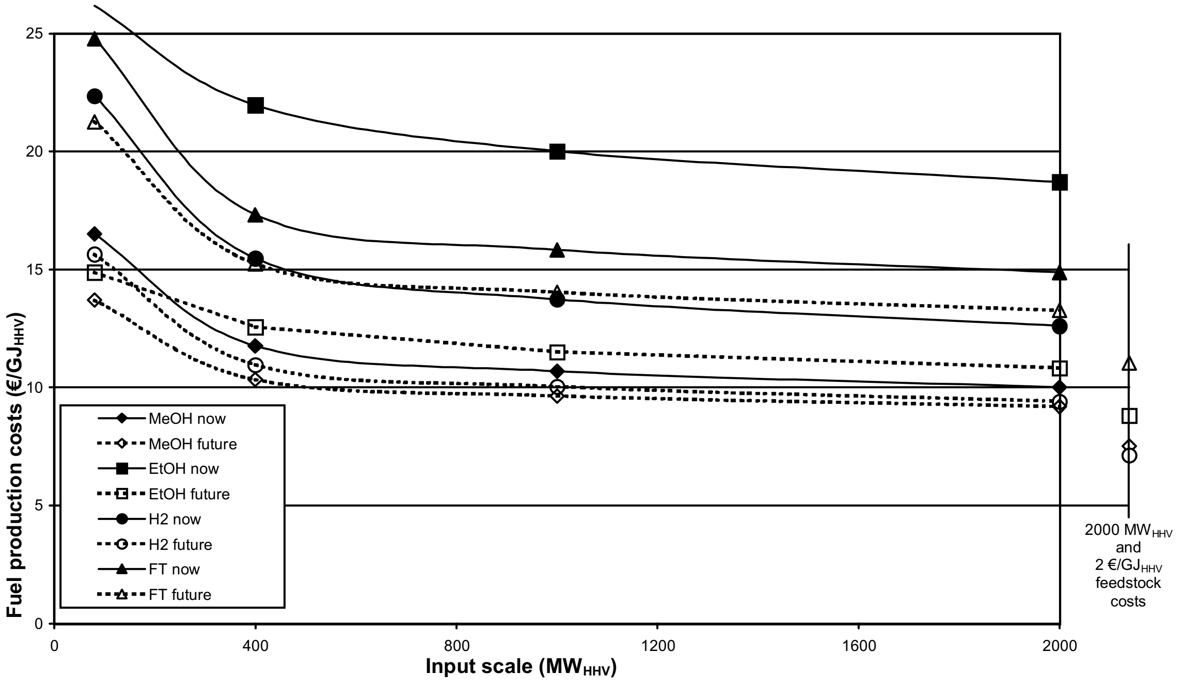

Figure 5. Influence of input scale on the production costs of selected biofuels. Feedstock costs 3 €/GJ$_{HHV}$, with a sensibility towards 2 €/GJ$_{HHV}$ at 2000 MW$_{HHV}$ input.

On the short term ethanol and FT diesel facilities are the most expensive. These processes also have the lowest total (fuel + electricity) efficiency on the short term. This, in combination with high operating costs, makes cellulose ethanol the most expensive of these biofuels on the short term. Production costs decrease with time and scale: through process improvements, technological learning and scale enlargement investment costs decrease and/or efficiencies go up.

A breakdown of the production costs into capital, O&M, feedstock and power costs is shown in Figure 4. A 30 % uncertainty should be applied to the total capital investment of methanol, hydrogen, and FT diesel concepts. The capital costs for the ethanol concepts are estimated to have a higher uncertainty (50 %). The uncertainty bars in the Figure show that the eventual influence of these uncertainties to the production costs is could be up to 30 % to 50 %.

Different overall scale factors for the production facilities' capital investments are found, and also these scale factors change over the whole 80 – 2000 MW_{HHV} range. The bare influence of scale (for all concepts of Table 4) on the biofuel production costs is made visible in Figure 5. All thermal gasification based processes experience a stronger influence of scale between 80 and 400 MW_{HHV}, than ethanol production. The hydrolysis fermentation takes place in vessels that have a small maximum size, decreasing scale advantages.

In the base situation (Figure 4) feedstock (at 3 €/GJ_{HHV}) accounts for 45 – 58 % of the total product costs. The influence of biomass feedstock price depends on the conversion efficiency from feedstock to fuel, e.g. a $\eta_{HHV,fuel}$ of 35 % (ethanol-now) implies that with every € feedstock cost reduction, the production costs reduce with 1/0.35 or 2.9 €. A much more efficient process, such as "methanol now"

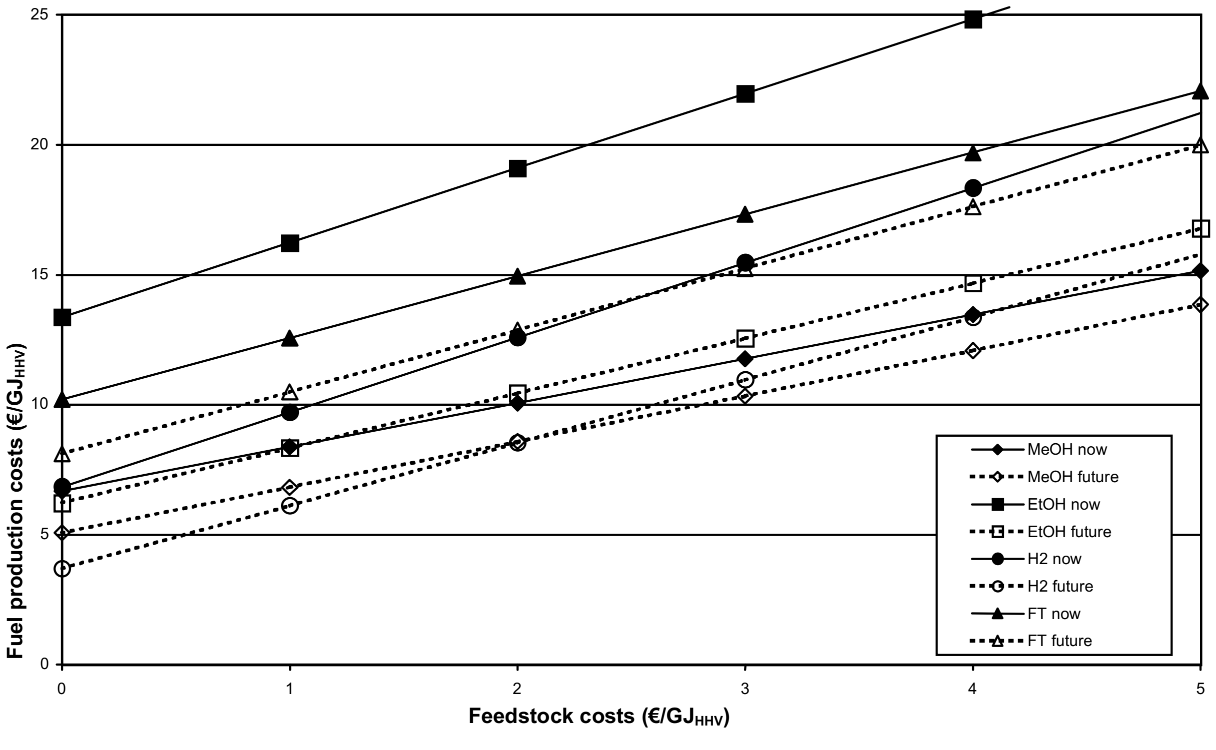

Figure 6. Influence of feedstock costs on the production costs of selected biofuels (400 MW_{HHV} input).

thus becomes relatively more attractive at high feedstock costs (refer to Figure 6). Since cheap feedstock will be used first (up to 3 €/GJ at gate), process improvements may initially focus on capital, O&M and power cost reduction.

4.3 Technological insights

Gasification based fuel production systems that apply pressurised gasifiers have higher joint fuel and electricity energy conversion efficiencies than atmospheric gasifier based systems. The total efficiency is also higher for once-through configurations, than for recycling configurations that aim at maximising fuel output. This effect is strongest for FT production, where (costly) recycling does not only introduce temperature and pressure leaps, but also 'material leaps' by reforming part of the product back to syngas. For methanol and hydrogen, however, the optimised fuel production with little or no electricity co-production performs economically somewhat better than once-through.

Hot (dry) gas cleaning generally improves the total efficiency, but the economical effects are ambivalent, since the investments also increase. A similar effect is that CO_2 removal does increase the total efficiency, and in FT production also the selectivity, but due to the accompanying increase in investment this does not result in lower product costs. The bulk of the capital investment is in the gasification and oxygen production system, syngas processing and power generation units. Especially for pressurised gasification larger scales may be more attractive than projected here and combinations with enriched air gasification (eliminating the expensive oxygen production assumed in this study) may reduce costs further.

The conversion of (hemi)cellulose to ethanol by hydrolysis fermentation is close to the stoichiometric maximum. The combination of little residual material and a high steam demand makes BIG/CC application unattractive. Developments of pre-treatment methods and the gradual ongoing reactor integration are independent trends and it is plausible that at least some of the improved performance will be realised on the middle term. The projected long-term performance depends on development of technologies that have not yet passed laboratory stage, and that may come commercially available earlier or later than 20 years from now. This would mean either a more attractive ethanol product cost on the middle term, or a less attractive cost on the long term.

Several technologies considered in this dissertation are not yet fully proven or commercially available. Pressurised (oxygen) gasifiers still need further development. At present, only atmospheric air gasifiers, operating at relatively small scale, have proved to be reliable. Consequently, the reliability of cost data for large-scale gasifiers is not known. A very critical step in all thermal systems is gas cleaning. It still has to be proven whether the (hot) gas cleaning section is able to meet the strict cleaning requirements for reforming, shift and synthesis. Liquid phase reactors (methanol and Fischer-Tropsch) are likely to have better economies of scale. The development of ceramic membrane technology is crucial to reach the projected hydrogen cost level. For Fischer-Tropsch diesel production, high CO conversion, either once through or after recycle of unconverted gas, and high C_{5+} selectivity are important for high overall energy efficiencies. Several units may be realised with higher efficiencies than considered in this dissertation: New catalysts and carrier liquids could improve liquid phase methanol single pass efficiency. At larger scales, conversion and power systems (especially the combined cycle) have higher efficiencies, but this has not been researched in depth.

In ethanol production the energy use and costs of the pre-treatment section (size reduction), and the (cost of) gypsum disposal need more attention. The investment costs for advanced hemicellulose hydrolysis methods need to be assessed more exact. Continuing development of new micro organisms is required to ensure fermentation of xylose and arabinose, and decrease the cellulase enzyme costs.

4.4 Comparison with other studies

Previous analyses on short term methanol and hydrogen production, by Katofsky [58] and Williams *et al.* (372 MW_{HHV}, 3.4 €/GJ_{HHV} feedstock, 0.07 €/kWh_e [46; 63]) yielded similar energy efficiencies (54 – 61 % by HHV), but significantly higher methanol production costs: 16 – 20 €/GJ_{HHV}. The largest difference is in the higher capital costs: higher TCI and higher annuity give 25 – 50 % higher annual capital costs. The ADL/GAVE study [42] reports 15 €/GJ methanol (feed 2.3 €/GJ, 433 MW input) largely using input parameters from Katofsky. In these studies [46; 58; 63] the efficiency towards hydrogen $\eta_{HHV,total}$ is 54 – 61 %, somewhat higher than for the short-term in this dissertation. Counterweighted by higher capital costs, the hydrogen production costs are similar: 13 – 16 €/GJ_{HHV}. Others [64] instead give much lower costs than presented here: 5.5 €/GJ_{HHV} for methanol and 5.4 €/GJ_{HHV} for hydrogen (530 MW_{HHV} biomass input). However, in that study, process efficiencies and biomass cost are not given, and a significant amount of energy is added as LPG.

The Fischer-Tropsch plant TCI is 1.7 times higher than reported by Bechtel [65], but that is explained by our application of a pressurised direct fired gasifier and air separation unit where Bechtel applies an indirect atmospheric gasifier, and by the fact that we include the power isle in the TCI where Bechtel places it outside the considered plant limits. The TCI for the FT part does compare with natural gas fed FT plants [66; 67]. Short-term production costs for biomass based FT diesel have been reported to amount 22 €/GJ [37; 42], which accords with the present findings. Bechtel do not give resulting FT diesel costs.

The resulting short- and long-term specific investments for lignocellulosic ethanol production, and also the resulting production costs are comparable with some literature [29; 60]. The much lower ethanol production costs found by Lynd *et al.* [68] can partly be explained from low biomass costs assumed, high electricity reimbursement and low annuity. Furthermore, they use very high biomass to ethanol efficiencies: 46 % by HHV in the short-term, which is high when considering the theoretical maximum of 50 – 55 %.

4.5 Broader comparison

Table 5 presents the estimated production costs for a broader range of biofuels, calculated from information on TCI, scale, and efficiencies, by using the same base assumptions as used earlier in this dissertation (refer to Table 3). For ethanol production from sugar cane a 3840 h operation window is used (limited harvest season and impossibility to store cane for longer time).

The calculated ethanol from sugarcane production costs are higher than the current prices in South East Brazil (7 – 9 €/GJ$_{HHV}$ currently without subsidies [30]). The (large) difference may be explained by the fact that many existing installations were build with subsidies in the eighties, and are at present completely depreciated, and by the direct link between the sugar market price and the decision to ferment sugar to ethanol. International alcohol shipment from Latin America to Europe will not add more than 0.5 €/GJ. If road tanker transport from an inland production location to the harbour were necessary, this would add half a euro extra per GJ per 100 km (Paper 1).

The calculated production costs of most fuels agree with literature: delivered costs are reported to amount 21 – 38 €/GJ$_{HHV}$ for ethanol from wheat, 15 – 29 €/GJ$_{HHV}$ for RME, and 13 €/GJ$_{HHV}$ for DME. Sugar beet ethanol is often projected cheaper, than calculated here, at a delivered cost of 26 – 40 €/GJ$_{HHV}$, and SNG more expensive at 13 €/GJ$_{HHV}$ [37; 40; 42; 71]. The applied method of analysis

Table 5. Production costs of fuels from various crops, not analysed further in this dissertation, based on literature. Technological and economic fuel production parameters for short term (400 MW$_{HHV}$ input), and production costs for short → long-term. Efficiencies are on whole used feedstock basis (see notes 3, 4 and 6).

Fuel	**Feedstock**[1]		**η_{HHV}**		**TCI**	**R**	**O&M**	**Production costs**[2]
		(€/GJ$_{HHV}$)	**Fuel**	**Electricity**	**(M€)**		**(% of TCI)**	**(€/GJ$_{HHV}$)**
Ethanol	Maize[3]	8.3	18 %	16 %	289	0.75	-8.3 %	25 → 19
	Wheat[3]	10	19 %	15 %	280	0.75	-8.3 %	29 → 24
	Sugar beet[4]	14	37 %	-2.2 %	149	0.75	-5.0 %	40 → 39
	Sugar cane[5] now	2.4	43 %	0%	48	0.8	13 %	11 ↘
	Sugar cane future	2.4	89 %	-1.2 %	153	0.8	10 %	8
Biodiesel (RME)	Rapeseed[6]	6.6	31 %	12 %	397	0.95	-0.9 %	25 → 23
DME[7]	Lignocellulose	3	58 %	-3.6 %	306	0.7	7 %	15 → 11
DMM[8]	Lignocellulose	3	46.4 %	0 %	242	0.7	4 %	15 → 11
SNG[8]	Lignocellulose	3	60 %	0 %	141	0.7	4 %	9 → 7

[1] Costs for maize, wheat and rape are for grain + straw (straw is free at field) [59]. All include 100 km truck transport of the raw material (0.5 €/GJ$_{HHV}$). Sugar cane feedstock price at conversion installation in Latin America, leaves are left in the field [32].

[2] The production costs are recalculated from total capital requirement and efficiency, assumptions: 8000 h load (3840 h for sugar cane), 10 % interest, 15 yrs lifetime, the relation between TCR and TCI was explained in Table 4, note 2. Electricity buy/sell 0.03 €/kWh$_e$, short term 400 MW$_{HHV}$ and long term 2000 MW$_{HHV}$ input. Long term includes cost reductions by learning (see respective notes).

[3] Ethanol from maize and wheat by the same wet milling-fermentation process. Conversion grain to ethanol is 37 → 40 % by HHV, conversion straw to electricity is 37 → 45 % by HHV (BIG-CC). The milling-fermentation process requires electricity 0.06 → 0.05 GJ$_e$/GJ$_{HHVfuel}$, and heat 0.24 → 0.20 GJ$_{th}$/GJ$_{HHVfuel}$. Efficiencies and costs reported in this table are for the total process of grain and straw to ethanol and electricity. Investment for the milling-fermenting part is 72 → 62 M€ for 104 MW$_{HHV}$ grain input , for the BIG/CC 2986 → 2398 €/kWh$_e$. The co-produced fodder has a value, which is settled with the O&M [29; 59; 69]

[4] Ethanol from sugarbeet conversion is 43 %, but requires heat 0.35 GJ$_{th}$/GJ$_{HHVfuel}$ and electricity 0.06 GJ$_e$/GJ$_{HHVfuel}$. The heat demand is deducted from the feedstock, the electricity supplied by the grid. 139 MW$_{HHV}$ input facility has TCR of 72 M€ now, and 10 % reduction towards future. Co-produced fodder has value, which is deducted from the O&M [29; 59].

[5] Conversion efficiency for an average Brazilian sugar/ethanol mill is 85 litre/tonne$_{wet}$ (mc 73 %) or 7.4 MJ$_{HHV}$/tonne$_{dry}$ or 42.8 % by HHV (sugarcane 17.35 GJ$_{HHV}$/tonne$_{dry}$) but could increase to 177 litre/tonne$_{wet}$ (mc 73 %) or 89 % HHV. Sugar mills assumed to produce or consume no extra energy on the short term, but on the long term produce electricity 55.4 MJ$_e$/tonne$_{wet}$ (electric efficiency 1.2 % by HHV). TCR for a 523 MW$_{HHV}$ input facility now is 63.2 M€, for a future 1951 MW$_{HHV}$ input 581 M€ [32].

[6] Efficiencies and costs reported in this table are for the total process of seed and straw to RME and electricity. Conversion of rapeseed to RME is 49 % by HHV. The extraction-esterification requires electricity 0.05 GJe/GJ$_{HHVfuel}$. A 1.875 MW$_{HHV}$ input esterification facility has a TCR of 2.1 → 1.9 M€ (10 % cost reduction by learning), costs and efficiencies for the BIG/CC as in note 3 [59].

[7] [70].

[8] [42].

obviously has a larger uncertainty than the method applied for the four selected biofuels.

The fuels could also be produced in the biomass supplying countries, provided that the amount of cheaply available biomass locally suffices, after which the biofuel is internationally shipped. This has two advantages: the elimination of the costly densification step otherwise necessary for producing pellets, and the higher energy density of the transported commodity. In Paper 1 it is shown that the joint cost reduction is about 9 % for methanol. Although ethanol has a higher energy density than methanol, the cost advantage will be practically the same. It can be concluded that this route is very attractive for sugarcane ethanol from Brazil.

The fuels' physical properties have a strong influence on costs and energy use of their distribution (Table 6). The energy density of ethanol and methanol is lower than that of gasoline and diesel, which results in proportionally higher costs. In this respect hydrogen is by far the most troublesome commodity. Pipeline transport in gaseous form is the cheapest and most energy efficient approach, but only suitable for large-scale, e.g. at national to regional level. Supplying individual gas stations requires the transport of liquid hydrogen by road tankers, which increases the costs [28]. Chemical properties of individual fuels require special materials and safety precautions (as was shown in Table 2). Alcohols and RME can best be blended with gasoline and diesel right at the refineries, or at intermediate depots between refineries and gas stations. The choice depends mainly on the biofuel production facility's location(s), on the blend's stability [37], and, in the case of alcohols, on whether or not they are also dispensed as neat fuels.

Table 6. Costs assumed for distribution and dispensing different fuels from a central production facility to gas stations [28; 59; 72].

Fuel	Costs (€/GJ$_{HHV}$)
Gasoline	1.41
Diesel	1.33
Methanol	2.12
Ethanol	1.84
Compressed hydrogen	4.50
LPG, DME	2.40

The resulting delivered costs are shown in Figure 7, left. On the short term, lignocellulosic methanol and sugarcane ethanol are the cheapest biofuels. They may be delivered at 13 – 14 €/GJ$_{HHV}$. RME from rapeseed is very expensive, but as has been reported by others [37], it has a very broad uncertainty range. Hydrogen has quite high distribution costs [59; 72], so end consumers costs are about 20 €/GJ$_{HHV}$, which is comparable with FT diesel. In the long term the delivered costs range from 10 – 15 €/GJ$_{HHV}$ for most biofuels, but the traditional biofuels lag behind. For comparison: gasoline over the last decennium costed 2.5 – 7.2 €/GJ$_{HHV}$ at Rotterdam port and diesel 2.4 – 6.6 €/GJ$_{HHV}$ [73], while

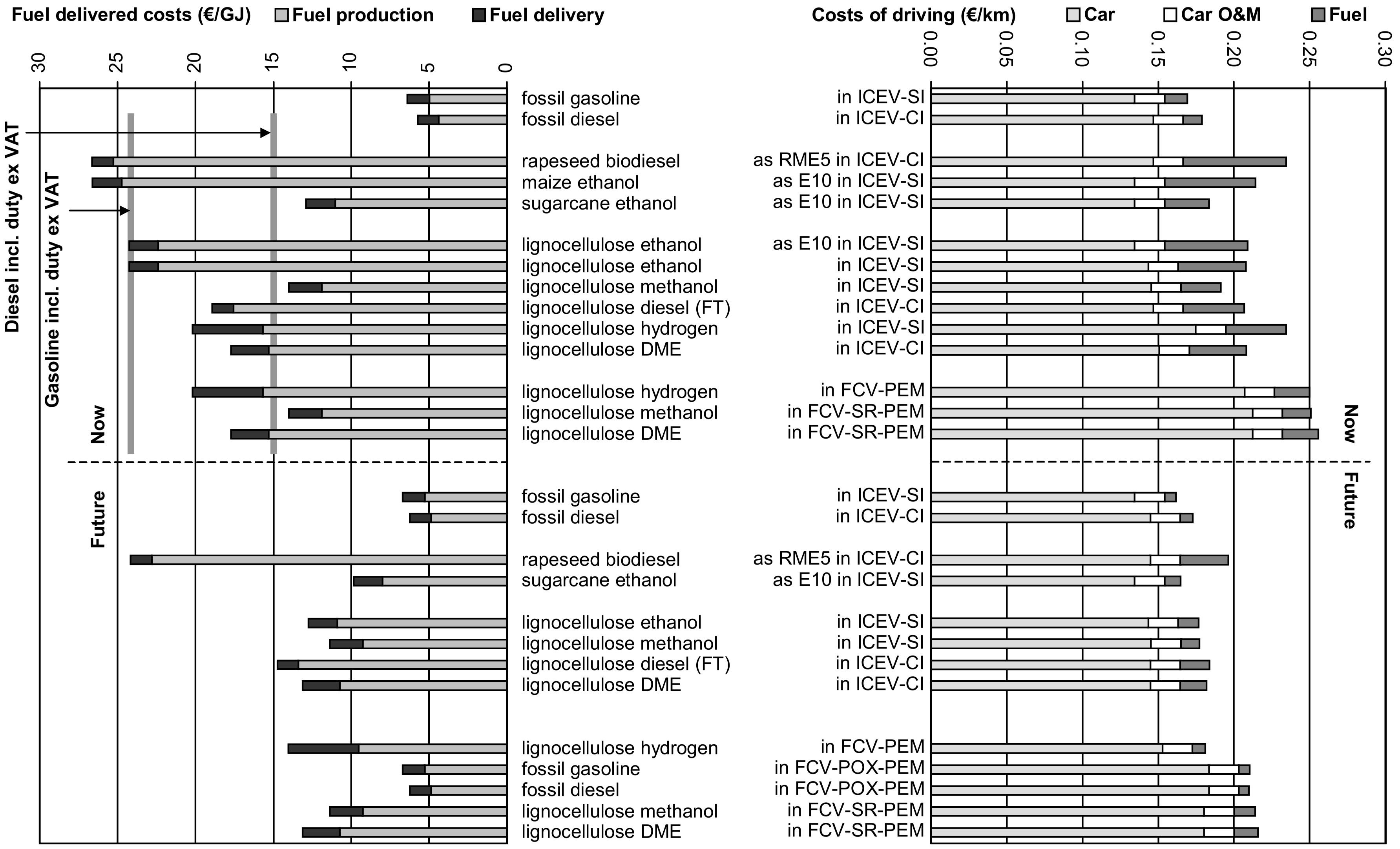

Figure 7. Delivered biofuels costs (left), and marginal costs of driving (right) for different fuel-car combinations for now → future. Costs exclude excise duty, fuel VAT and road tax. Both fuel and driving costs are on pure biofuels basis: all extra or avoided costs of blends, relative to pure gasoline or diesel, are allocated to the biofuel. Mineral oil derived gasoline costs 5 → 5.3 €/GJ_{HHV} and diesel 4.4 → 4.9 €/GJ_{HHV}. For all vehicles (100 kW wheel power passenger cars), annual distance is 20,000 km, lifetime is 10 yr, annual O&M is 400 €. RME5 means 5 % biodiesel in diesel, E10 means 10 % ethanol in gasoline.

distribution to gas stations adds about 1.4 €/GJ_{HHV} [28], so that the high ends of the delivered costs[**] were about 9 €/GJ_{HHV}. As projections for future fossil fuels' costs are highly uncertain[††], they may well come in the range of biofuels costs.

4.6 Energy requirement for energy

All biofuels require energy for growth, harvest and transport of the feedstock. Some require additional energy (electricity, heat, materials) in the conversion step, or co-produce electricity or materials. Finally, energy is required for distribution and fuelling. In general little energy is required for distribution and fuelling, the key exception being the liquefaction of hydrogen that may consume up to 20 % of its energy content.

The sum of all net energy inputs that can be allocated to the fuel output, determines whether the fuel's net energy yield is positive. Co-products are also charged with part of the inputs: the allocation is done by energy content; electricity is expressed in primary energy. In the Energy Requirement for Energy (ERE) in Table 5, biomass itself is not accounted for. The ERE thus becomes a measure for fossil input only, although part of this input could eventually be substituted by biofuels / bioelectricity from other sources.

The future lignocellulose feedstock yield may eventually be 20 – 30 $tonne_{dry}$/ha.yr for genetically improved eucalyptus in tropical plantation. Such high yields would decrease the costs for production and first truck transport. This effect is not analysed in this dissertation.

International bioenergy transport requires additional energy inputs, and part of the bioenergy is lost in conversion efficiencies, decay and spills. Pellets, delivered from Latin America or Eastern Europe, have a primary energy use (ex biomass energy itself) of respectively 9 and 6 % of the delivered biomass. This includes the energy for growth, harvest and local transport in the country of origin. Accounting for material losses, the energy requirement for biomass delivered is 1.26 and 1.32 MJ/MJ respectively. Methanol produced from the Latin American pellets will have a ERE only about 8 – 10 % points higher than reported in Table 7. It is concluded that more advantageous to produce the biofuels close to the biomass sources, and to transport the fuels instead of the raw biomass. Transport of sugarcane ethanol from Latin America to Western Europe consumes 2 % of the delivered ethanol energy as primary energy in the intercontinental shipping step.

[**] The sale price at the gas station further usually includes excise duty, in the Netherlands this is currently 18 €/GJ_{HHV} (0.64 €/l) for gasoline and 9 €/GJ_{HHV} (0.34 €/l) for diesel [37], and value added tax (VAT, 20 %). (Partial) duty exemption for biofuels could make their price competitive with gasoline and diesel. VAT is compulsory and the same percentage for all vehicle fuels. The current sale prices in the Netherlands are about 33 €/GJ_{HHV} (1.15 €/l) for gasoline and 21 €/GJ_{HHV} (0.80 €/l) for diesel.

[††] the world's oil supplies are not unlimited [74], and although the insights differ from increasing oil production until at least 2025 [27], down to declining oil production within the next 20 [75] or 5 to 10 years [76; 77]. The differences are partly explained by the definition of 'supplies'. This may or may not include unconventional oil that – although it cannot be exploited economically at present – could eventually be exploited economically depending on both technology improvement and the market's demand [8]. After a production peak, the world does not run out of oil, but the oil prices likely rise [27]. For comparison, over the last hundred years the crude oil price has usually been between 1.6 and 4.9 €/GJ [73], and up to 13 €/GJ during the oil crisis.

Table 7. Energy Requirement for Energy (ERE) up to fuelling, and fuel yield per area up to fuelling, for different fuel-crop combinations for short → long-term. Excludes the international transport (see text).

	Feedstock	**ERE_{fuel} ex biomass**[1] **(GJ_{prim}/GJ_{HHV})**	**Feedstock yield**[2] **$tonne_{dry}$/ha.yr**	**Area net fuel yield (10^1 GJ_{HHV}/ha.yr)**
Methanol	Lignocellulose	28 % → 10 %	11 → 30	9 → 30
Ethanol	Maize	40 % → 22 %	7(+6) → 9(+6)	-1 → 2
	Wheat	39 % → 21 %	7(+6) → 9(+6)	0 → 3
	Sugar beet	71 % → 60 %	13 → 19	3 → 6
	Sugar cane	12 % → 7 %	20 → 27	13 → 39
	Lignocellulose	16 % → 9 %	11 → 30	6 → 25
Hydrogen	Lignocellulose	14 % → 11 %	11 → 30	5 → 19
Synthetic diesel	Lignocellulose	14 % → 10 %	11 → 30	8 → 22
Biodiesel (RME)	Rapeseed	36% → 21 %	3(+2.5) → 4(+4.5)	1 → 2
DME	Lignocellulose	26 % → 23 %	11 → 30	9 → 26
DMM	Lignocellulose	15 % → 11 %	11 → 30	9 → 24
SNG	Lignocellulose	14 % → 11 %	11 → 30	11 → 31

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[1] Energy Requirement for Energy (ERE) is defined (here) as the Gross Energy Requirement (GER) per unit energy (HHV) of delivered fuel. The GER on its term is defined as the amount of energy required for producing the fuel. Here, the GER consists of the HHV energy in the raw material, the first order primary energy required for growth and harvest of this raw material, for transportation of the raw material to a central point, for conversion, and for delivery of the fuel, allocated to the fraction desired product. Energy values of feedstock: wood 19.5 $GJ_{HHV}/tonne_{dry}$, rape (seed only) 27.8 – 28.8 $GJ_{HHV}/tonne_{dry}$, sugar cane 17.4 $GJ_{HHV}/tonne_{dry}$, sugar beet 17.4 – 19.7 $GJ_{HHV}/tonne_{dry}$, wheat (grain only) 18.2 – 18.5 $GJ_{HHV}/tonne_{dry}$, maize (grain only) 15.8 $GJ_{HHV}/tonne_{dry}$, straw 19.5 $GJ_{HHV}/tonne_{dry}$ [32; 59; 78]. Energy use for growing and harvesting the feedstock (as percentage of the feedstock GJ_{HHV}) is: wood 4.0 → 2.1 %, rape (seed basis) 22.2 → 13.8 %, sugar cane stalks 2.2 %, sugar beet 12.6 → 8.3 %, wheat and maize (grain basis) 18.7 → 10.2 % [32; 40; 78]. Energy use for raw material transportation (100 km): wood 2.5 % of the raw material HHV, wheat/maize 2.1 % (grain and straw), sugar beet 8.3 %, rapeseed 0.7 % (seed and straw) [40]. Energy used for transportation and distribution of the fuel produced is 0.5 – 1.5 % of the HHV energy content for most fuels, but 11 % for hydrogen [28]. Electricity consumed accounts for 2.3 GJ_{prim}/GJ_e (44 % efficiency averaged on coal, oil, gas).

[2] Lignocellulose: now willow and poplar in USA and Europe → future eucalyptus in tropical plantation. Yield for rape, maize and wheat is for seed and (within brackets) for straw (used for heat and electricity generation) [8; 32; 59; 79].

Combination of the ERE and the biomass yield per hectare gives the Area net fuel yield, which is also shown in Table 7. According to literature, the yields of the fuels from traditional crops could be about 3 – 5 GJ_{fuel}/ha.yr higher than found here [29].

A large number of uncertain parameters is involved in the calculation of ERE and net fuel yield. When the energy inputs for production and delivery are assumed to have a summed uncertainty of 40 % and the conversion efficiency may be 5 % wrong, than the ERE may have 50 % uncertainty. This seems very large, but for most biofuels this still means a considerable bioenergy yield. Also, it can be concluded, that the energy yield for lignocellulosic crops is much higher than for annual crops [8; 78].

4.7 Biofuels' use in cars

Different fuels perform different in various drive chains. Therefore, the various biofuels should be compared on an equal basis, such as driving costs, their energy saving potential, or specific costs as CO_2 reducing option. Moreover, the prospects for biofuels cannot be seen apart from (the future of) vehicle propulsion. A selection of fuels, shown in Figure 7 (left), is used for further comparison. Table 8 overviews the performance of (bio)fuels in different engines and car costs. The fuel cell vehicle has been included both as an (expensive) option for the near-term and is assumed to be available at projected costs in the future.

The engine efficiency (measured in km/GJ fuel) of ICEVs can be improved for both spark and compression ignition engines. Alcohols clearly have a higher efficiency in spark-ignition ICEVs than gasoline, because of the higher octane[‡‡] number. Efficiency differences found for different fuels in compression-ignition ICEVs are so small that they will not be reckoned with [28]. Vehicle efficiency improvements in transmission and aerodynamics are generally independent of the fuel type [80] and are not anticipated here. Because of differences in energy content between the fuels, equal driving ranges require different fuel and tank loads (volume or mass), which indirectly influences the performance [29; 72]. It is not known to what extent this is dealt with in literature.

All biofuels require adaptations in ICEV cars, leading to extra costs. Especially the costs of individual cars depend on many choices that have little to do with fuel economy, such as luxury and status. Therefore, individual quotes, on how much a car would cost, are useless for the analysis. Only relative

Table 8. Car costs and fuel efficiency1) ($\text{km/GJ}_{\text{HHV}}$) on short → long term, for passenger cars (about 100 kW wheel power), for various fuels in various engine systems.

Engine system	Fuel		Car costs1) (€)		Efficiency2) ($\text{km/GJ}_{\text{fuel}}$)	
ICEV-SI1)	Gasoline		16500		430	→ 950
	LPG		18500		480	→ 1000
	SNG / CNG		17900		440	→ 950
	Methanol	M85 (FFV)	18500		490	→ 950
		M100	17800		540	→ 950
	Ethanol	E10	16500		440	→ 950
		E85 (FFV)	18500		520	
		E100	17600		540	→ 950
	MTBE/ETBE		16500		490	
	H2		21450		510	
ICEV-CI	Diesel (fossil, synthetic, RME)		18000	→ 17750	470	→ 760
	M100		18000		470	
	DME		18500	→17750	470	→ 760
	DMM		18000		470	
FCV-PEM	H2		25400	→ 18750	880	→ 1690
FCV- SR-PEM	M100		26050	→ 22100	760	→ 840
FCV-POX-PEM	gasoline / diesel / E100		26500	→ 22500	630	→ 960

1) Costs for the gasoline ICEV SI and diesel ICEV CI are set; the others are derived from absolute costs (extra costs for drive train) and relative costs in literature [28; 42; 59; 72].

2) Besides on the fuel and the engine, the fuel efficiency depends on other car features (weight, form, energy regeneration options) and the drive cycle [72; 81]. Efficiency is found expressed in many different ways: mile per gallon (mpg), 100 km per certain litres of gasoline equivalent, %, MJ/km. To calculate from km per litre gasoline equivalent to km per GJ, multiply by 28.4 litre/GJ (1 litre for 15 km equals 430 km/GJ). Efficiencies estimated from average driving cycles in literature [28; 42; 46; 59; 80; 82]. Efficiencies for blends are allocated to the biofuel only.

[‡‡] The octane number expresses a fuels quality rating, indicating the ability of the fuel to resist premature detonation and to burn evenly when exposed to heat and pressure in an (spark ignited) internal combustion engine. Normal gasoline has an octane number of 87–89. The cetane number is a primary measure of fuel suitability for diesel engines (compression ignited), but not related to fuel efficiency. It essentially expresses the delay before ignition. The shorter the delay the better—and the higher the cetane number.

costs from comparisons of different fuel-car combinations in literature are used.

Many advantages are attributed to future fuel cell vehicles[§§] over ICEVs. Potentially, they have a high energy efficiency (not limited to the Carnot efficiency of thermal energy processes), hardly produce emissions or make noise during operation, require little maintenance (no moving parts), and would be cheap in owning and operation [28; 46; 58; 72; 83]. However, fuel cells are (at present) expensive, large and heavy per kW output. Also, flexible car operation (changing load) requires the use of very efficient batteries. Whether and when FCVs will really perform better than future ICEVs is not clear [82].

To use a fuel other than hydrogen in PEM Fuel cells, it must be converted into H_2 (and CO_2) onboard the vehicle. The most simple fuels methanol and DME can be steam reformed (SR) at relatively low temperatures [28], while others require (harsher) partial oxidation (POX) or autothermal reforming. On board reforming implies an energy (direct and via vehicle weight) and cost penalty. The total fuel efficiency of a fuel cell system must then take into account the efficiency of the reforming process as well as the efficiency of the fuel cell. The heat generated as a by-product of the fuel cell can be used to increase the system's energy efficiency. Onboard reforming is only an option if the reformer is flexible in providing hydrogen to the fuel cell, as fast or slow as it is being consumed by the fuel cell. If additional hydrogen storage would be necessary, the onboard reformer looses its advantage [84]. Direct methanol fuel cells (DMFC) circumvent reforming, thereby offering lower system complexity. Although the methanol to electricity efficiency is theoretically high, results reported so far are not optimistic. Moreover, the catalyst is very expensive, and methanol still crosses the membrane [85].

For a 100 kW wheel power passenger car with annual 20,000 km driving cycle, the resulting costs per kilometre driven are shown in Figure 7. Biomass alternatives to diesel are all slightly more expensive than fossil diesel on the short term, but synthetic diesel and DME become competitive on the longer term. Most fuel cell options are significantly more expensive than gasoline/diesel ICEVs; only hydrogen in a future FCV PEM may become competitive.

In this Figure, excise duty and VAT are excluded, and the resulting fuel contribution may become strikingly small: only 9 % of the total driving costs for hydrogen in FCVs. This agrees well with values reported by others [42; 72]. Considering these findings with the fact that car costs are dictated by other factors than engine and fuel system costs, and with the observed uncertainty in future fossil fuel derived gasoline and diesel costs, there are no a priori economic barriers to develop and introduce cars that run on advanced fuels.

[§§] Different types of fuel cells are considered for vehicles: Alkaline Fuel Cell (AFC) and Proton Exchange Membrane (aka Polymer Electrolyte Membrane PEM) fuel cell that process hydrogen, and the Direct Methanol Fuel Cell (DMFC) that processes methanol. All have low operation temperatures (20 – 120 °C). Most literature and demonstration programs focus on PEM FCVs. The AFC is extremely expensive and requires very pure H_2.

4.8 CO_2 emission avoided, and costs

Since energy inputs (typically fossil fuels energy) are needed in the growing, harvesting and transport of the biomass, and in the delivery of the fuel to the end-user, the total lifecycle emissions, though small compared to fossil fuel systems, are not zero. The net CO_2 emission of fuel use can be calculated from the net primary energy inputs allocated to the fuel. The CO_2 emission (avoided) from electricity is set 0.068 tonne CO_2/GJ_P (or 560 g CO_2/kWh_e). This value holds for a typical Western European electricity mix of 37 % coal, 5 % oil 53 % gas (Paper 1). All other energy inputs and outputs are assumed to stem from oil (0.068 tonne CO_2/GJ_P). It would be more exact to judge all energy entries individually by their origin, but not enough material was available to do this. Greenhouse gasses other than CO_2, or other lifecycle emissions, have not been accounted for in this dissertation.

Results are shown in Figure 8 (bars). The net CO_2 emission of gasoline in a current modern ICEV-SI is indicated as reference level. Using the efficiency value from Table 8 and a ERE of 110 % this level is calculated to be 172 g CO_2/km. This is much lower than what others reported for the present average Dutch vehicle[***] (218 g CO_2/km [42]), or gasoline vehicles in general (230 g CO_2 equivalent/km [37]). The difference can be explained from the assumed gasoline fuel economy (1 litre gasoline for 15 km). Note that all the future fuel-car combinations may emit less than 100 g CO_2/km. This fits the agreement between the car industry and the European Union, that cars sold in Europe by 2008 should not emit more than 140 g of fossil carbon dioxide per km, and less than 120 g/km in 2012.

Any uncertainties in the ERE calculation (± 50 %) and uncertainties in vehicle efficiency (estimated 5 %) are translated into uncertainties in the net CO_2 emission. Therefore, values in Figure 8 may well be comparable with emissions previously reported by others [37; 46; 86]. With the uncertainties in the present results ranking the various fuel-car combinations is difficult. It can, nevertheless, be concluded that fuels from lignocellulosic crops have a 2 to 3 times better CO_2 reduction potential than maize ethanol or RME on the short and longer term. Although the absolute emissions have a high uncertainty, the absolute CO_2 emission *reduction* is much more certain.

Dividing the relative costs per kilometre of a fuel-car combination compared to *present* gasoline-ICEV by the relative CO_2 emissions per km provides a measure for CO_2 emission reduction costs (Figure 8, circles). Uncertainty is estimated about 35 % for lignocellulose pure methanol in current ICEVs, and worse for fuels that reduce less: up to 50 % for RME in current ICEVs. Emission reduction costs 100 – 300 €/tonne CO_2 avoided for ICEV applications on the short term. It is concluded that at present biofuels use in FCVs would be an expensive CO_2 emission reduction option.

As future gasoline and sugarcane E10 ICEVs were projected to have lower costs per km (refer to Figure 7, right), the CO_2 avoidance costs are negative. This is very interesting for sugarcane ethanol because it also has a large reduction potential (albeit not in its E10 form). Most of the future lignocellulosic fuel-car combinations do have both an attractive CO_2 emission reduction potential and possibly attractive associated costs (about 50 – 100 €/tonne CO_2 avoided), which agrees with literature.

[***] Average Dutch vehicle consumes 47 % gasoline, 46 % diesel and 7 % LPG (energy basis) [42].

It seems that ethanol and methanol in ICEVs have lower associated costs than FT diesel and DME. Also it seems (again) that the use of methanol or DME in FCVs is a costly option (about 250 €/tonne CO_2 avoided). However, these high costs are entirely caused by the high but uncertain FCV costs.

By others, cellulosic ethanol in ICEVs and methanol in FCVs have been ranked to be the most cost-effective options (25 – 60 €/tonne CO_2 avoided), where DME, methanol and FT diesel in ICEVs are ranked among the more expensive options (120 – 170 €/tonne CO_2 avoided) [21; 42; 46].

The CO_2 emission reduction costs are high when compared to the current CO_2 emission market price (about 13 €/tonne CO_2 avoided). On the other hand, a large share (65 %) of the eventual required CO_2 emission reduction in the Netherlands is assumed to be realised at costs below 100 €/tonne CO_2 avoided, by energy savings, renewable energy (wind, hydro, biomass), and clean use of fossil fuels [42; 87-90]. When compared to this, some biofuels can cost-effectively contribute to CO_2 emission reduction.

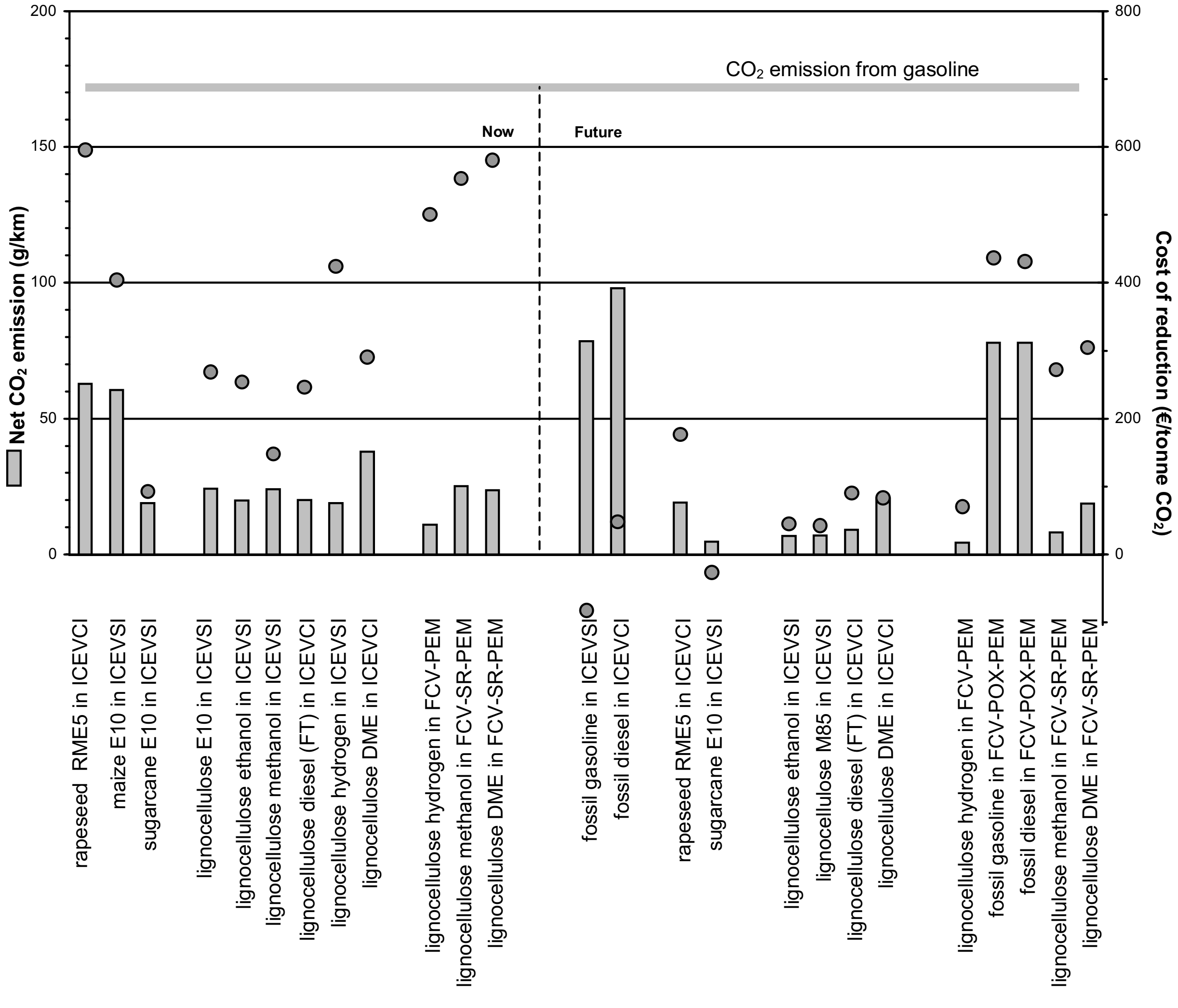

Figure 8. CO_2 emission and costs of emission reduction for different fuel-car combinations on now → future, compared to gasoline ICEV. Same assumptions as for Figure 7. RME5 is a blend of 5 vol % biodiesel in diesel, E10 is a blend of 10 vol % ethanol in gasoline, M85 is a blend of 85 vol % methanol in gasoline.

4.9 Transition

When concerning the costs of driving, as well as the potential and costs of CO_2 reduction, fuels from lignocellulosic crops and sugarcane are found to have the best prospects. With the present knowledge and data quality it is, however, difficult to point out the absolute best fuel. Eventually, gasification based processes give a broader flexibility in fuel choice than hydrolysis fermentation. Further development of gasification leaves many fuel possibilities open. Of these, methanol, DME and Fischer-Tropsch diesel are fuels that could gradually be introduced as blends with gasoline and diesel, hardly requiring infrastructure adaptations (though DME requires a separate LPG like infrastructure).

The production of FT diesel production via gasification has been demonstrated [91], and a 1 MW input test facility has successfully produced both methanol and FT diesel [92]. However, large-scale introduction of any of these fuels takes time, not in the least because large conversion plants have to be realised. When reckoning with developing, engineering, legal procedures and building, the realisation of a new facility takes at least 5 – 6 years. Furthermore, these facilities require a constant large biomass supply. During the last decade especially Sweden and Finland gained experience with trading biomass for energy and heat, and a international biomass market is currently emerging. Enlargement of this market directly interacts with the development of new and larger conversion technology.

Sugar/starch derived ethanol blended with gasoline, and rapeseed derived biodiesel with diesel may remain the most popular (and important) biofuels until 2010 [39], despite their high costs and unfavourable land-use. Ethanol-gasoline blends may facilitate development of better ethanol processes, and facilitate the introduction of neat ethanol vehicles later [39], biodiesel does not have such advantages. Nevertheless, almost all existing vehicles (and consumers) can directly be addressed with these fuels [37].

On longer term, hydrogen, methanol and DME are the most suitable in a transition to fuel cell vehicles, if that would be desirable. The eventual shift to hydrogen then requires concurrent development of new infrastructure and FCV introduction. Most proposed implementation strategies come down to demonstration of fuels in taxi or bus fleets, being expanded to larger regions [84]. The other route applies flexible fuel vehicles that run two fuels to facilitate the transition from one to another. The competition or collaboration between these two options deserves more attention.

5 Conclusion

Biomass could play a large and important role in a future sustainable energy supply as source for modern energy carriers as heat, electricity and transportation fuels. Introduction of biofuels is especially attractive because it can both strongly reduce CO_2 emissions against (eventually) reasonable costs, and because it decreases or spreads fuel dependency. Of the many conceivable biofuels, on the long-term fuels from lignocellulosic biomass are probably the most attractive especially because they may allow for a higher fuel yield per hectare, require less additional energy for growth and harvest, and have better projected economics.

5.1 Results

In this dissertation, the production of four promising biofuels – methanol, ethanol, hydrogen, and synthetic diesel – is systematically analyzed. Production costs of these fuels range 16 – 22 €/GJ_{HHV} now, down to 9 – 13 €/GJ_{HHV} in future (2030). This performance assumes both certain technological developments as well as the availability of biomass at 3 €/GJ_{HHV}. The feedstock costs strongly influence the resulting biofuel costs by 2 – 3 €/GJ_{fuel} for each €/GJ_{HHV} feedstock difference. In biomass producing regions such as Latin America or the former USSR, the four fuels could be produced against 7 – 11 €/GJ_{HHV}. The uncertainties in the biofuels production costs of the four selected biofuels are 15 – 30 %, which is small when considering the large uncertainty in future (2030) gasoline and diesel prices.

The gasification-derived fuels require the development of large (about 400 MW_{HHV} input) pressurised gasifiers, a gas cleaning section that matches the catalyst's specification, increased catalyst selectivity (for FT diesel production), and ceramic membranes (hydrogen). Hot gas cleaning and CO_2 removal positively affect the total plant efficiency, but the economic effect is ambivalent. The production of ethanol from lignocellulosic biomass requires the development of more efficient pretreatment technology, and of micro-organisms that yield higher conversions, as well as the integration of several conversions into fewer reactors.

The different fuels are analysed at different levels of detail. This yields a different quality of results and insights. In the production of ethanol the size of material flows is directly translated into costs of reactor trains. Closer modelling of the separate reactors, and better understanding of microbiological reactor engineering would increase the certainty of the results. The matching of heat supplies and demands within plants is done more conscious for the methanol and hydrogen concepts than for the other fuels. However, the advantage gained is small, and it appears that for the estimation method used it is good enough to match the largest streams only.

Not all options to improve plant performance are researched. For example, the maximum size of pressurised gasification may well be beyond the 400 MW_{HHV} assumed. The intermediate energy carriers delivered by the international transport chains in Paper 1 are of higher quality (drier and in compressed form), than the feedstock assumed in Papers 2 – 5 (wet chips). This means that the projected plant efficiencies will in fact be higher and the TCI slightly lower.

The same assumptions on feedstock costs (if possible), scale, load, and capital depreciation were used for calculating the production costs of other biofuels. This learnt that also DMM, DME and SNG might fall in the same cost range. RME from rapeseed and ethanol from sugar beet or starch crops are already available, but their production is expensive and inefficient.

When applied in cars, biofuels have driving costs in ICEVs of about 0.18 – 0.24 €/km now and maybe about 0.18 in future (fuel excise duty and VAT excluded). This is only slightly higher than the driving costs of fossil fuels. Moreover, the cars' contribution to these costs is much larger than the fuels' contribution, and the differences in the fuels' contributions are only about 1 cent/km. The driving costs for fuel cell vehicles are larger because of the projected larger vehicle costs. At the same time it is known that the car sell prices do not always correlate with the engine type and fuel system. From this perspective one can conclude that cars that run on advanced fuels can be brought on the market at competitive prices, and that the biofuels' driving costs can compete with those of fossil derived fuels.

As a CO_2 emission reduction option, future lignocellulosic biofuels may cost about 50 - 100 €/tonne CO_2, which is acceptable when considering many other options. It must be realised that these costs are more influenced by the cars' costs, than by the fuels' costs.

5.2 Recommendations for research and development

Our knowledge on the performance of biofuels can further be improved by closer assessment of the conversion systems and especially of the whole chain from crop to motion:

Biomass supply costs. Eventually, the competitiveness of future large-scale conversion facilities depends on the evolvement of an international biomass market, with large quantities of biomass against low costs. It is unclear to what extent trade barriers increase these costs. A significant part of the delivery costs is in the local transport from field to a central gathering point. Here, smarter logistics and the use of larger trucks could further decrease the delivered costs.

Sustainability. If biofuels are to contribute to sustainable development, then it must be guaranteed that the biomass is sustainably grown: this supposes knowledge on the net CO_2 balance depending on the cultivation method, the emission of other greenhouse gases than CO_2, the disadvantages of monocultures (for biodiversity), the use of pesticides and fertilizer, ground water level, and local socio-economic (dis)advantages [4; 93]. Note that the cropping of sugar cane requires a high quality soil that would otherwise be used for agriculture.

Gasification. All biofuels can be produced by using today's technologies and demonstration of especially thermal routes should start swiftly. (Oxygen blown) pressurised biomass gasification must be further developed to enable the large-scales assumed in this dissertation (400 MW_{HHV} gasification). More insight is required about the technical maximum of such gasifiers and the accompanying costs.

Gas cleaning. This section still requires special attention to guarantee sufficient cleaning. In most BIG/CC failure instances, the cause was insufficient gas cleaning.

Economic analysis. The factored estimation method yields investment costs with an uncertainty range of ± 30 % for methanol, hydrogen and Fischer-Tropsch diesel. To decrease these uncertainties, detailed engineering would be needed. The ± 50 % uncertainty for lignocellulosic ethanol can be decreased to about ± 30 % by closer assessment of the amount and size of required reactor vessels, solids loading, and residence time. The poor availability of cost figures in general, often leads to repeatedly quoting old quotes, and losing the meaning of the economic numbers.

End use. Relative numbers have been used for fuel distribution costs, car costs and fuel economies. This part should be better underpinned to decrease uncertainties in driving costs and eventually enable a more specific biofuels ranking. The vehicle's performance and especially its cost are the crucial factors for the costs of each biofuel chain. This requires substantial research in development of required infra-and dispensing structure, in bare engine performance and whole-car improvements, and in absolute costs of the bare engines with their fuel systems. Although in relative sense biofuels' costs are only a small part of the driving costs, they may play an important role in consumers' perception.

Transition. Transition is a very complex issue that cannot be understood by techno-economic insights only. It also has practical, social and economic dimensions, the interaction of which justifies another dissertation. Choices for one fuel could block or slow down the development of others. Interacting effects between biofuels and technology development should be mapped. Also, the effect of different governmental stimulation measures on the different fuels is of interest.

Overall. The combination of fossil fuels and biomass could facilitate scale enlargement and cost reduction in both the international transport chain and the biofuels production facilities.

5.3 Closing remarks

For advanced biofuels, this is just the beginning. Biofuels are, at the very present, a hot issue in the European Union. National governments can give biofuels momentum by both stimulating technology developments and by stimulating biofuels use by e.g tax exemption, or by imposing a biofuel fraction on the market. Biofuels will certainly play a role in the future energy supply system. The weight of this role depends on the importance attached to fuel security, and on the willingness to address the greenhouse problem and reduce CO_2 emissions.

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Paper 1

International bioenergy transport costs and energy balance

Contents

Abstract..........55

1 Introduction..........56

2 Conceptual approach..........58

2.1 System outline 58

2.2 Modelled chains 59

2.3 Model set-up 61

3 Component data..........62

3.1 Biomass sources 62

3.2 Storage and treatments 65

3.3 Conversion to motor fuels or electricity 67

3.4 Transport 68

4 Results..........70

4.1 Production areas and transport distances 70

4.2 Mass flows 71

4.3 Economy 72

4.4 Energy and CO_2 75

5 Summary and Conclusion..........77

6 Acknowledgments..........78

References..........79

International bioenergy transport costs and energy balance*

Abstract

To supply biomass from production areas to energy importing regions, long distance international transport is necessary, implying additional logistics, costs, energy consumption and material losses compared to local utilisation. A broad variety of bioenergy chains can be envisioned, comprising different biomass feedstock production systems, pre-treatment and conversion operations, and transport of raw and refined solid biomass and liquid bio-derived fuels. A tool was developed to consistently compare the possible bioenergy chains and assess the influence of key parameters such as distance, timing and scale on performance. Chains of European and Latin American bioenergy carriers delivered to Western Europe were analysed using generic data. European biomass residues and crops can be delivered at 90 and 70 €/tonne$_{dry}$ (4.7 and 3.6 €/GJ$_{HHV}$) when shipped as pellets. South American crops are produced against much lower costs, and despite the long shipping distance, the costs in the receiving harbour can be as low as 60 €/tonne$_{dry}$ or 3.1 €/GJ$_{HHV}$; the crop's costs account for 25 – 40 % of the delivered costs. The relatively expensive truck transport from production site to gathering point restricts the size of the production area, therefore a high biomass yield per hectare is vital to enable large scale systems. 300 MW$_{HHV}$ Latin American biomass in BIG/CC plants may result in cost of electricity as little as 3.5 €cent/kWh, competitive with fossil electricity. Methanol produced in Latin America and delivered to Europe may cost 8 – 10 €/GJ$_{HHV}$, when the pellets to methanol conversion is done in Europe the delivered methanol costs are higher. The energy requirement to deliver solid biomass from both crops and residues from the different production countries is 1.2 – 1.3 MJ$_{primary}$/MJ$_{delivered}$ (coal ~ 1.1 MJ/MJ). International bioenergy trade is possible against low costs and modest energy loss; it is not blocking the development of large bioenergy facilities.

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* Manuscript. Co-authors are Roald AA Suurs and André PC Faaij.

1 Introduction

The continuous growth of global energy consumption raises urgent problems. The larger part of mineral oil and gas reserves ($^1/_3$ of the world's energy supply) is located within a small group of countries, forming a vulnerable energy supply [1]. Moreover this supply is expected to reach its limits someday in the future. On the other side the use of fossil fuels causes numerous environmental problems, such as local air pollution and greenhouse gases emission [1].

A possible way to deal with these problems is the development of cleaner and renewable energy sources. Modern use of biomass is an interesting option, because biomass is worldwide availability, it can be applied for power and biofuels, and it may be produced and consumed on a CO_2-neutral basis [2-4]. Disadvantages are the current high costs when substantial amounts of energy are required, and the involved large areas of land. In theory, energy farming, especially on surplus agricultural land, can contribute between 33 and 1130 EJ/yr in 2050, without jeopardising the world's food supply [5]. By assessing the land use and productivity on grid cell (0.5° by 0.5°) level, and application of IPCC SRES scenarios, this range may further be narrowed to 310 – 660 EJ [6]. For comparison, the current global energy use is 410 EJ [5].

Key drivers behind large-scale production and trade of biofuels are currently the climate policies of various western countries. In the Netherlands and Sweden, several energy producers already import biomass in order to produce 'green' energy and started with long distance bioenergy transport operations. Cheap organic industrial by products and refined biofuels are imported from the world, to comply with the 'green' energy demand.

Some regions (like for example Latin America and Eastern Europe) have a much larger bio energy production potential than others, due to a combination of large land areas with good crop production potential, low population density and extensive agricultural practices. Consequently, various countries may become net suppliers of renewable bioenergy to countries that are net importers of energy [2; 5; 7; 8]. In order for bioenergy to be available to importing regions, transport of biofuels over relatively long distances is necessary. This, however, implies additional logistics, costs, energy use and material losses.

When envisioning a fully developed bioenergy market in the long run, in which some regions are suppliers to other regions, the question emerges whether large-scale long distance transport is economically and energetically feasible, as well as attractive in terms of GHG emission reductions.

A few earlier studies [8; 9] have given indications that intercontinental trade of biofuels or even bulk transport of wood could be economically feasible and does certainly not lead to dramatic energy losses: Electricity delivered to the Dutch grid, from Scandinavian, Eastern European and Latin American energy crops transported as bales, pellets, chips or electricity, may cost 0.08 – 0.09 €†/kWh [8] or 0.06 – 0.12 €/kWh [9]. Imported bio-methanol would cost 12 - 14 €/GJ_{HHV}. External energy input is 4 – 20 % of the solid biomass delivered.

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† All costs are in $€_{2003}$.

The costs and energy efficiency of international bioenergy trade are influenced by many factors. The biomass may origin from plantations, forestry residues, or industrial processing (e.g. waste from pulp and paper production, sawmill residues or bagasse from sugar production). The bioenergy can be transported many forms: 'raw' biomass (chips, logs, bales), intermediate energy carriers (such as bio-oil or charcoal) or high quality energy carriers such as ethanol, methanol, Fischer-Tropsch liquids and hydrogen or even electricity. Other parameters in the design of the bioenergy chain are the order and choice of pre-treatment operations, equipment performance, transport distance, dry matter losses, supply pattern, fuel prices, and scale. Theoretically, a large number of bioenergy chains can be envisioned. It is important to gain insight in the effects of all these variables on the total costs and energy consumption of bioenergy chains. Doing so allows for identification of best configurations for bio-energy supply systems, as well as improvement options. The influence of most of these parameters on the performance of complete transport chains was not previously addressed.

Therefore, the present study [10; 11] considers both overland and sea transport, more detailed analysis of costs and energy use of components (storage, pre-treatment, transfer and transport), and logistic organisation of the chains. Parameters such as distance, biomass characteristics (density, moisture content, heating value, particle size), matter losses (throughput changes along the chain), scale of the whole system, and timing are expected to influence the performance of the separate steps considerably. Therefore, their influence is explicitly assessed and modelled in this study. The data, used for conversion and transport operation and the changing characteristics of the biomass, therefore have a rather detailed level. The study focuses on long-distance-large-scale bio energy trade, and tries – by analysing many possible chain variations – to identify optimal chain configurations for the on long-term.

For a fast analysis of a wide variety of chains, a flexible model has been created with a modular structure. The different processing and transportation steps can easily be stringed together in any configuration. This allows for analysis of many chains. In each step the biomass characteristics are altered and costs and energy use are summarised. In principle generic data is used, derived from literature, close analysis of key processes, and interviews. All relevant parameters can easily be adjusted and their impact analysed, for sensitivity analysis and case studies.

Section 2 discusses how the problem is approached, which chains are analysed, and – in general terms – the developed spreadsheet. In Section 3 harvesting, conversion and transportation operations are assessed to yield necessary data input for the model. Results of cost, energy use and CO_2 balance calculations are evaluated in Section 4. Finally Section 5 discusses and concludes the study and our findings.

2 Conceptual approach

2.1 System outline

For structuring international supply systems, four general system components or operation types are distinguished: biomass production, pre-treatment, transport and energy conversion. For each operation type various parameters determine its performance (Table 1.) and individual cost and energy analyses can be made. A more elaborate description of the separate system components will be given in Section 3.

Table 1. Operation types, with possible options and key model variables.

Operation type	Options	Key variables	
Biomass production	Forestry residues · Energy crops · Felling · Industrial rest stream · Chipping · Baling	· Harvesting window1) · Production costs (location dependent)	
Pre-treatment	· Storage · Chipping · Drying · Pelletising	· Equipment capacity · Capital and O&M · Energy consumption (power, fuel, heat)	· Load factor2) · Dry matter loss · Moisture loss
Transport	· Truck · Train · Ship	· Transport distance · Speed · Capacity · Product weight · Product volume	· Capital and O&M · Fuel consumption · Load factor · Transfer time & costs
Energy conversion	· Power · Methanol · Pyrolysis oil	· Conversion efficiency · Capital and O&M · Load factor	

1) Section of the year in which the biomass comes available, e.g. October – March. A window could also apply to apparatus that is only available during part of the year.

2) Or: operation time, part of the time that a facility or transport means is available within its operation window.

Chain components can be selected and organised in many ways, but many aspects are interdependent, so there is a limited degree of freedom in choosing alternatives (e.g. pelletising is only possible with sized biomass). In addition, some arrangements are unrealistic because of obvious disadvantages (e.g. pelletising of already transported biofuels).

Several generic logistic situations are shown in Figure 1, a situation with five possible transfer points is assumed: the production site, a central gathering point (CGP), two transport terminals (export and import) and the energy plant. In general, biomass is collected locally at small scale production sites and transported to a CGP. This allows for larger capacity treatment facilities, which makes the use of costs-intensive pre-treatment and conversion technologies economically more attractive. Local transport to CGP, harbour or conversion facility takes place by truck (a). Long distance transport is done by train or ship (b & c).

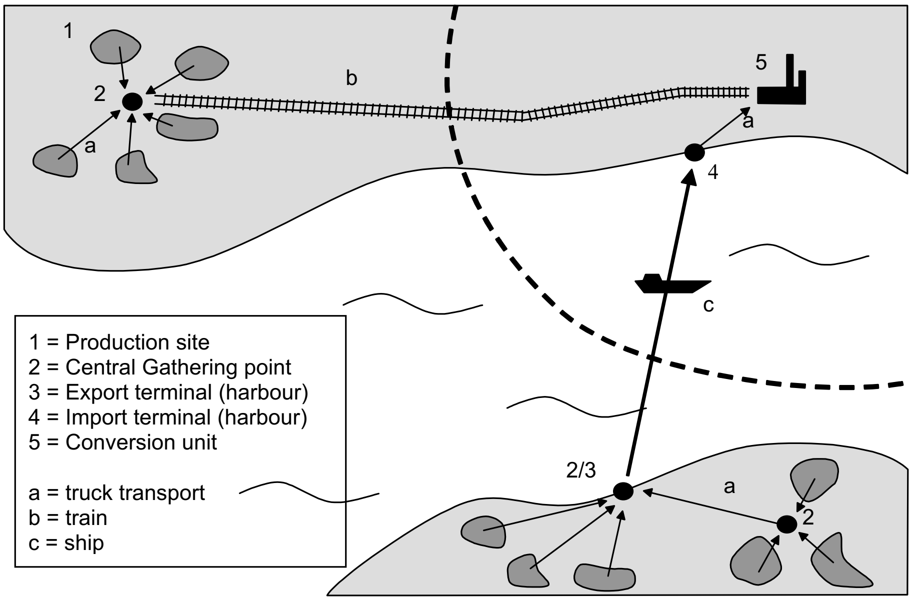


Figure 1. Geographical system outline.

The efficiency of bioenergy transport chains may be improved when drying and/or densification are applied early in the chain. At the production site, baling or chipping can be integrated in the harvest method. At the CGP and the energy plant, the biofuels can be subjected to pre-treatment and conversion operations, e.g. sizing, drying, densification but also the conversion of woody biomass to liquid fuels like methanol or ethanol. The biomass could also be converted to intermediate products such as pyrolysis oil, pellets or charcoal. Various techniques for energy densification like drying, methanol synthesis or pelletising, are only cost efficient at larger scales (at the CGP).

Capacities or scales of operations (pre-treatment, transport, conversion) decrease along the chain, due to dry matter losses, drying, or conversion. As most processes have a volume-limited throughput, the influence of moisture content of the processed feedstock on operation's size is of minor importance.

For capital-intensive steps in the chains, an important aspect is the operation time, which is determined by Operation Window (OW) and the load factor. The OW is defined by the annual period that the equipment is available/used for processing, e.g. feedstock supply may limit the use of a harvester to several months. The load factor is a percentage of the time that the equipment can be used within the OW, it may depend on the working shift (8 hours/day, 5 days/week), the delivery of truck loads, equipment reliability, or equipment transfer from one harvesting site to another.

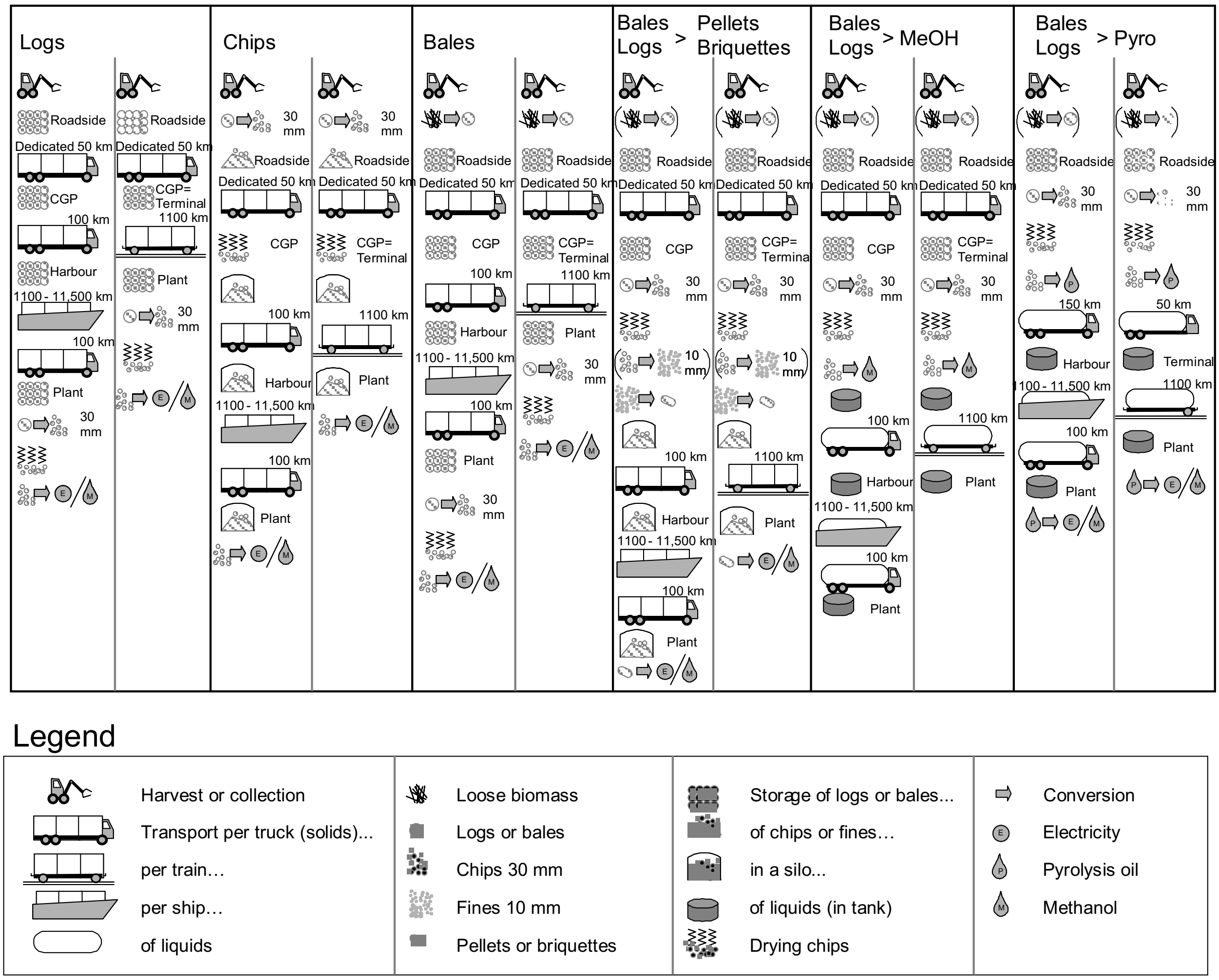
2.2 Modelled chains

Considering all the transportation means and bio-energy commodities, many chains are possible to move bio-energy from one country to another. Different chains and optimisation options will be analysed and compared using generic information to be discussed in Section 3.

Table 2 defines the chains to be modelled, using a limited number or steps. Each chain generally follows the route of collecting biomass – local transport – central gathering point (pre-treatment, conversion possible) – transport to a harbour (if applicable) – international transport – local transport (if applicable) – conversion to another energy carrier. The generic chains represent transport of (Eastern) European biomass on the one hand and of Latin American biomass on the other. The choice for a region influences the assumptions about productivity, biomass price, front-end amount, local energy composition and costs, local transport distances, and the international transport distance.

The chosen chains can be characterised by the commodity in their main (international) transportation step: logs, chips, bales, pellets or briquettes, methanol, or pyrolysis oil. The first three commodities are produced in the field or at the roadside; the other (denser) bio-energy commodities are produced from bales or logs at the CGP. Methanol is an end product that will not be processed further. All other commodities are converted at the end of the chain into electricity (in a pulverised fuel plant or a gasification combined cycle plant), or methanol (gasification), although also the delivering of solid biomass will be analysed. The chains are analysed for different scales: 300, 600, and 1200 MW_{HHV} input.

Table 2. Theoretical transport chains, from the different export regions to Western Europe, to be analysed. The chains are named after the commodity in their international transportation step, each with an overseas (left) and overland (right) variation.



2.3 Model set-up

A flexible modular spreadsheet has been developed to enable the technical-economic analysis of a large variety of chains. First a chain is defined by the user, then the chain is processed by the spreadsheet, to yield results on costs, energy use and CO_2 emissions for each step in the chain and cumulative for the end product.

In each step along the transport chain, a biomass processing operation (from Table 1.) can be chosen and characterised by user input parameters (dedication, transport distance…). The model (see Figure 2) processes all the steps in consecutive order. After each step the results for costs, energy use and emissions are retained for the overview and the new biomass characteristics are written to the next step.

Annual costs for a step consist of the annual capital costs, operating and maintenance, and auxiliary energy use (electricity, heat or fuel). Possible benefits from co-produced electricity, heat or products may decrease the costs. When biomass is lost or used for heat generation along the chain, this is implemented in terms of efficiency. The annual capital costs are the pay-down and interest of the Total

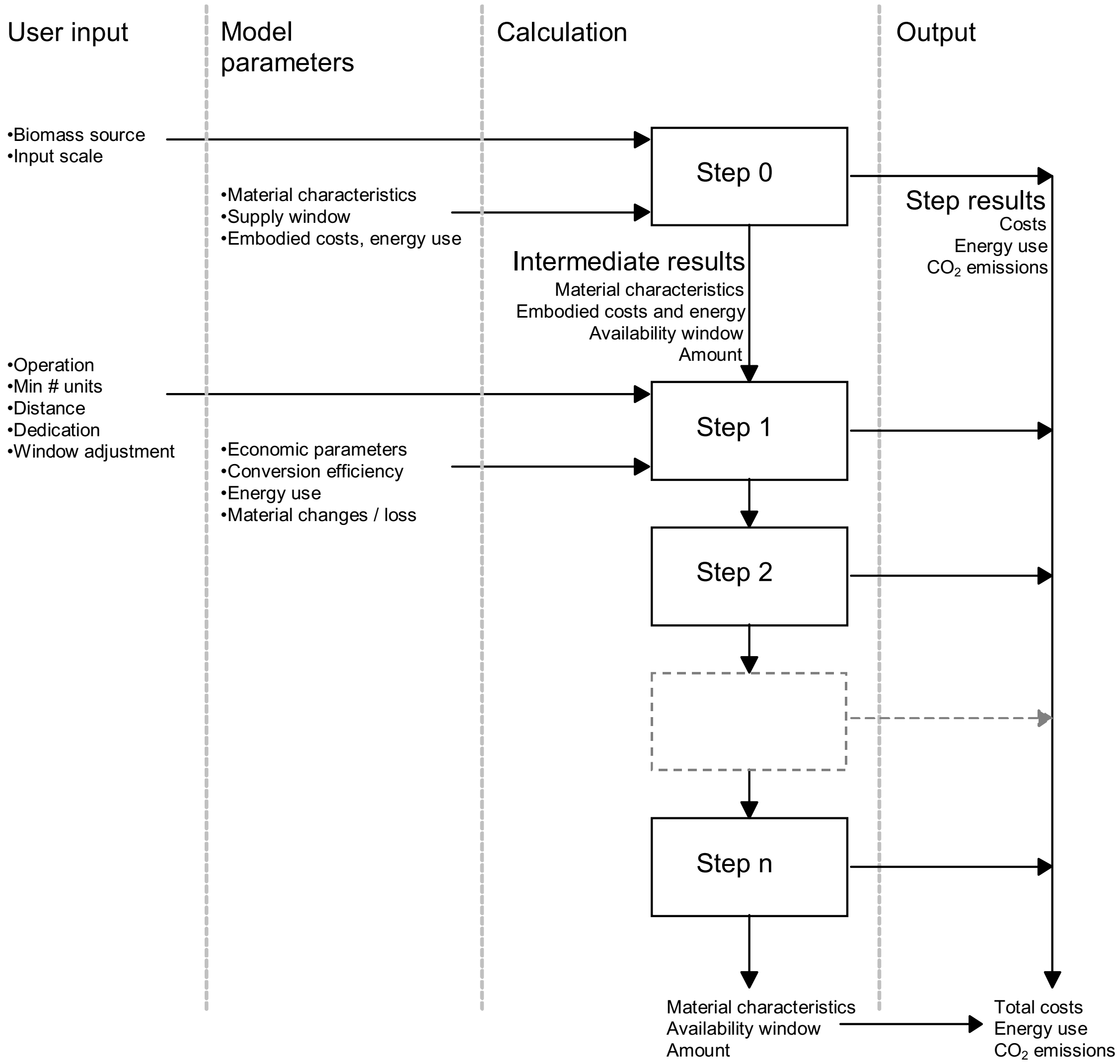


Figure 2. Information flow through the modelled chain.

Capital Requirement (TCR), and depend on economic lifetime and interest rate. The TCR is calculated from a base capital at a base scale, using an exponent factor [12; 13]. Costs for consumed energy vary per region. The assumed numbers (Table 3) are of a rather exemplary nature, taken from representative countries in the assessed regions.

The chain's energy balance and CO_2 emission balance are influenced by the local energy mix (Table 3). At some places biomass is used to generate power or heat used by the bioenergy chain. Although this gives a direct CO_2 emission of about 84 kg/GJ_{HHV} wood, the net (lifecycle) emission is set zero, assuming that all emitted CO_2 is captured by growing biomass.

Table 3. Local parameters assumed for different regions.

		North western Europe	**Scandinavia**	**Eastern Europe**	**Latin America**
Electricity mix[1)]	Coal	37 %	2 %	18 %	2 %
	Oil	5 %	3 %	9 %	3 %
	Gas	53 %	1 %	40 %	8 %
	Nuclear	5 %	44 %	12 %	3 %
	Hydro	0 %	44 %	1 %	82 %
	Other	0 %	6 %	20 %	2 %
Price (€)[2)]	Electricity (per MWh)	58	34	48	34
	Fuel Diesel (per GJ_{HHV})	16	16	13	11
	Fuel HFO (per GJ_{HHV})	3.1	3.1	3.1	3.1
CO_2 (tonne)[3)]	Electricity (per MWh)	0.64	0.046	0.41	0.072
	Fuel Diesel (per GJ_{HHV})	0.073	0.073	0.073	0.073
	Fuel HFO (per GJ_{HHV})	0.078	0.078	0.078	0.078
Prim (GJ_{HHV})[4)]	Electricity (per MWh)	8.9	7.2	7.7	4.4
	Fuel Diesel (per GJ_{HHV})	1.2	1.2	1.2	1.2
	Fuel HFO (per GJ_{HHV})	1.1	1.1	1.1	1.1

[1)] WEC [14] International Energy Data for 1995 are used from respectively the Netherlands, Sweden, the Russian Federation and Brazil/Argentina (average). These countries are assumed representative for the concerned regions. "Other" comprises solar, wind, and biomass.

[2)] Retail prices as presented by the IEA [15]. Electricity prices for industry in the Netherlands, Sweden, and Poland. HFO is especially used for international transport, and is assumed have a flat global price of 130 $US\$_{2001}$/tonne (42.1 GJ_{HHV}/tonne). Automotive diesel oil for these countries costs 606, 624 and 514 $US\$_{2001}/m^3$ (38.7 MJ_{HHV}/l), and 433 in Mexico. Average Brazilian price for industrial electricity is 34 € [16].

[3)] Specific CO_2 emission for electricity are calculated from the local electricity mix, efficiency and fuel combustion emissions from the Revised 1996 IPCC Guidelines. Specific CO_2 emission for fuel diesel and fuel HFO stem from EIA [17]. Specific emissions for fuels stem from Forsberg [18].

[4)] All fossil, sustainable and nuclear "fuels" require indirect embodied energy for winning, transport, storage, and refining, before they can be converted [19], the primary energy for electricity is calculated from the national electricity mix and the conversion efficiency.

3 Component data

3.1 Biomass sources

Although biomass plantations could eventually supply 310 – 660 EJ [6], for near term applications, biomass residues from forestry and industry will most probably be used first, because they are cheaper and available at locations closer to conversion facilities, albeit at smaller scale. The range of possible biomass sources from residues on the short term, towards short rotation coppice systems on a fully developed (future) market will be discussed and the most important generic data is given in Table 4.

Forest residues consist of treetops, branches, foliage, unmarketable bolts and undergrowth trees. They come available as bi-product from harvest operations (for timber or pulp and paper), or directly as forest thinnings. Residues form the most economically significant source of raw material in the production of wood fuels on the European continent. They are available in large amounts in e.g. Sweden, the Baltic States, Canada and Russia. Spruce trees make out the major part of the residues, and come available during the whole year, since they can be harvested anytime of the year. However, most harvest is done in winter: the frozen soil eases the forest operations. Leaves have fallen off in autumn and needles dry and fall off after harvest so that valuable nutrients remain in the forest ecosystem. The wood can than dry over summer [20]. Felling areas measure typically 0.5 – 1 ha and are spread throughout a large area of typically 100,000 ha. Forestry and harvesting operations are fully mechanised in Sweden and Finland. While cutting down the trees, the harvesters sort out the logs and the residues on different piles.

· If *chips* are the desired commodity, two approaches are distinguished: First residues may be left in piles on the cutover, to dry during the spring and early summer. Eventually, they are forwarded to the roadside, where the residue piles are covered with paperboard to keep the moisture content down. Gradually parts of the pile will be chipped locally, just before transportation [21]. Chips have a relatively low bulk density and are (due to moisture content and large specific surface) vulnerable to fungi, which make them less interesting for long distance transport. In the second approach a terrain chipper with a bin, chips the logging residues at the site, shortly after the harvest. Then, the chips are hauled into lorry trailers at the roadside, which transport the chips to a Central Gathering Point or power plant.

· Unchipped *logging residues* are – ever more – compacted to bales (Composite Residue Logs or CRLs). The CRLs have a diameter of roughly 0.75 m and a length of 3 m. They weigh between 400 and 600 kg and have an energy output in excess of 1 MWh. The compaction units are mounted on medium-duty forwarders. A major advantage of the CRL system is that CRLs can be handled as round wood throughout the handling chain: conventional forwarders and round wood haulage rigs can be used, and they also facilitate efficient haulage by rail. Several problems do remain to be solved. Carrying CRLs on round wood haulage rigs may require the use of netting or solid sideboards to prevent material falling off during transit. The influence of storing CRLs (large specific surface) for a longer time, on the biomass characteristics is unclear, and CRL chipping is as yet less efficient than chipping round wood [21].

At present, the lower part of the tree stem is used as timber, whereas the next three meters are normally destined for the pulp and paper industry. Depending on their respective markets and on the price paid for bio-energy, this upper part of the tree (and also logs from thinnings) could come available for energy production.

Energy crops. Sweden has also sites with dedicated energy crops, but current production levels are at present limited [10]. In Sweden, mainly Salix is grown as energy crop. Salix (willow) is an short rotation coppice that is harvested every 4 year, after about 25 year the cycle ends and the stand is renewed. Agricultural plots of Salix can be found scattered all over the country, including 18,000 ha in southern Sweden [20]. Cultivation and harvest are still rather inefficient and costly due to the small scale. However, in the future production and efficiency could improve and prices for dedicated energy crops are expected to evolve to the same range as forest residue wood [22]. Harvest is done either by direct in field chipping, or by cutting and bundling the stems. We assume that the bundles can be handled in the same way as CRLs and logs.

Compared to Sweden, Eastern European countries have much lower land and labour costs, and therefore a larger potential bio energy production against lower costs. Large agricultural areas may come available when entry into the European Union, which can lead to increased competition and improvements in agricultural productivity and efficiency. Although data is only scarcely available, projections are that, at present, poplar could be produced in Poland at about 2.2 €/GJ_{HHV} [23]. On longer term, bioenergy crops could be produced in e.g. Ukraine. Assuming much lower land costs and income (currently 6-fold lower) compared to Sweden, production costs may be three times lower.

Latin America offers great potential for large scale Eucalyptus cultivation, also a short rotation coppice.

Table 4. Input data for bioenergy sources used in this study. An arrow (→) indicates parametric changes from short to long term (now → 2015). Costs and energy use include any harvesting, haulage, in field compacting and in-field chipping necessary, to deliver the described form at the roadside.

	Forestry residues1)			**Energy crops2)**		
Region	**Scandinavia**			**Scandinavia**	**Eastern Europe**	**Latin America**
Form	**Logs**	**Chips**	**CRLs**	**Bundles**	**Bundles**	**Logs**
Costs (€/$tonne_{wet}$)	9.3	27.5	17.4	48.5	18.4	16.8→10.2
(€/GJ_{HHV})	0.9	2.6	1.5	3.9	1.5	1.1→0.7
Diesel use (MJ_{HHV}/tonne)	20	600	220	47→25	25	60→48
moisture content	50 %	50 %	45 %	37 %	37 %	20 %
ash (db)	0.2 %	0.2 %	0.2 %	1.6 %	1.6 %	0.5 %
C (daf)	47.4 %	47.4 %	47.4 %	49.5 %	49.5 %	49.8 %
H (daf)	6.3 %	6.3 %	6.3 %	5.8 %	5.8 %	5.8 %
O (daf)	46.2 %	46.2 %	46.2 %	43.4 %	43.4 %	44.2 %
N (daf)	0.1 %	0.1 %	0.1 %	0.4 %	0.4 %	0.1 %
avg ps (mm)	3000	30	3000	3000	3000	1000
Density (kg/m^3_{bulk})	462	219	251	160	160	280
HV ($GJ_{HHV}/tonne_{dry}$)	20.8	20.8	20.8	19.5	19.5	19.4
Average yield (tonne/km^2.yr)	0.6	0.43	0.43	370→675	675	467→583
Supply window	oct-mar	oct-mar	oct-mar	oct-mar	oct-mar	whole year

1) Averaged costs from [18; 20; 24]. Required energy derived from Forsberg [18]. Composition and heating value is for spruce [25]. The average yield is low because the production of residues after felling per area is low and the average rotation period of the stand is long (106 y) [18; 20; 26].

2) Energy use and costs may decrease over time, as the production per area increases, this is indicated by an arrow (→). Costs and energy data for Scandinavia (salix and poplar) are average from several literature sources [8; 25-27]. For Eastern Europe (long term), most parameters are taken the same as for long term Scandinavia, except for the production costs, which are assumed 2 – 3 times lower. For Latin America (eucalyptus) parameters where compiled from Brazil and Nicaragua [25-28].

Brazil already has 6 Mha of plantations, which especially produce wood for charcoal to be used in iron smelting. For the north-east part alone a potential of 8-13 EJ/yr of Eucalyptus energy exists. The tree stems can be harvested during the whole year, however drying conditions vary enormously with seasons and differences in biomass quality can be expected. Leaves and small branches are left in the field, since the amount is too small to make recovery for energy use feasible [28]. The harvesting site area is currently typically 1,000-10,000 ha, but large cultivation areas of over 100,000 ha are foreseen for the future.

3.2 Storage and treatments

Storage is needed at points in the chain where delivering and further transportation do not match. At the roadside at the beginning of the chain, storage is often applied to dry the biomass during the winter from 50 % moisture to about 30 % against no costs. However, biomass may decompose when kept for longer time, and consequently part of the biomass is lost. Decomposition speed increases with moisture content, and is largest for wet chips: outside storage causes up to 3 % dry mass loss per month [18]. Nevertheless, logs, chips and bales are generally stored in the open air (1.1 €/m^3.yr, [10]). Dried chips and pellets (more valuable) are stored in bunkers (87 €/m^3.yr) and liquids are stored in tanks (0.31 k€/m^3.yr) Roofed, bunker, silo and tank storage have less or no influence on the biomass characteristics.

Sizing (Table 5) can be integrated in the harvesting procedure, or done at a central location such as the CGP or the final conversion plant. Chips have a too low density for long distance transport, and will primarily be a short period intermediate, just before pelletising or thermal/chemical conversion. There are large differences in the scales at which chipping is applied, and different feedstock types and output particle sizes are possible, causing large differences in costs and energy use for the various chippers found in literature. For the present analysis, a roll crusher is applied to do coarse chipping from stems to about 30 mm. This size is sufficiently small for fluidised bed gasification [29]. Further grinding by a hammermill to 10 mm may be necessary for the production of pellets [30].

Drying is applied for several reasons. The efficiency of combustion or gasification improves with drier biomass feed. Dry feedstock is required for BIG/CC application, as it increases the biogas heating value, making it suitable for the gas turbine. Furthermore, drying reduces the biomass susceptibility to decomposition, associated matter losses, and fire and health hazards. And drying reduces the weight and may thus give a cost advantage further down the chain, especially in the long distance step. Drying doesn't seriously affect the volume of the material [30]. The optimum moisture content is 10 - 15 %, because further drying would require an increasingly larger installation, and the specific energy use increases. Many biomass dryer types have been developed. Here, the rotary drum is chosen; it applies a proven technique and is applicable at large scale. The choice for one dryer or another will in practice depend on the scale and on the availability of hot air, flue gas or steam. If no heat source is around, part of the biomass feedstock needs to be combusted. All dryers demand the feedstock material to be chip size, so a sizing step will always precede the drying operation. [18; 30].

Table 5. Generic performance data and characteristics for treatments used in the chain modelling. Data on possible variations can be found in Hamelinck *et al* [11].

	Sizing1)		**Drying2)**	**Densification3)**		**Pyrolysis4)**
	Roll Crusher	**Hammermill**	**Rotary Drum**	**Pellet press**	**Piston press (briquettes)**	
Base Scale5) (tonne/hr)	10	50	100	6	5	1 MW_{LHV} input
Base Capital6) (M€)	0.14	0.37	5	0.12	0.425	0.10
Scale factor R	0.7	0.7	0.7	0.61	1	0.62
Load factor	90 %	90 %	100 %	90 %	90 %	90 %
O&M	20 %	20 %	3 %	197 %	37 %	4 %
Lifetime	15	15	15	10	10	25
Energy-e (kWh/tonne)	8.22	3.5	20	28	34	37.2
Energy-h			2.5 GJ/twe			
Form	Chips	Chips	Chips	Pellets	Briquettes	Pyro oil
average particle size (mm)	3000→30	30→10	30	10	40-125	n/a
Bulk density (kg/m^3bulk)	240	240		650	600	1175
all matter loss/action	2 %	2 %	1 %			
moisture content			7 %	8 %	10 %	*f* (feed)

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1) The energy requirement will actually be calculated from the input and output particle size via the Bond work index [31]. The bulk density of fine ground particles increases with decreasing size. The average particle size indicates the change from input to output by an arrow (→). Costs and energy use from [30].

2) Heat for drying, expressed per tonne water evaporated, or twe [18; 30], will mostly be provided by combusting a part of the biomass, in that case this can be expressed by a matter loss of 0.125 tonne_{dry}/twe. Costs and energy use from [30].

3) Both pellet and briquette production require a low feedstock moisture content (8 – 12 %) [32]. The operational costs for pellet presses are rather high because the dies wear out fast [32]. The energy use (per throughput) for the piston press decreases with scale [30; 33].

4) Pyrolysis [34] has stringent demands for particle size (<2mm) and moisture content (~7%) of the feed material. The moisture content, hydrogen content and heating value of the product depend on the feedstock. A small amount of electricity (tonne feed basis) is required for processing. The heating value depends on the mc of the product, which in turn depends on the mc of the feed. The maximum scale is 10 MW_{LHV} input. Conversion efficiency is 67 % on LHV basis.

5) Unless indicated otherwise, the base scale is also the maximum scale, above which multiple units are necessary.

6) The base capital or Total Capital Requirement (TCR) mostly equals the Total Capital Investment (TCI), only for large plants with an investment path the TCR is larger.

Densification. Biomass may be compacted to briquettes (diameter greater than ~ 30 mm) or pellets (smaller) to save transport and handling costs, and to improve combustion efficiency. It essentially involves two parts; pressurised compaction of loose material to reduce its volume and binding the material so that the product remains in the compressed state. The binding agent is lignin from the biomass itself. It softens above 100 °C and glues the fibres when cooled. The production of briquettes requires not so small feedstock particles (< 50 mm) as the production of pellets (3 – 10 mm, achieved by hammermill) [30]. Pellets and briquettes can directly be applied in gasification [29].

The bulk or packing density of pellets is lower than its specific density, as the pellets will not pack perfectly, about a factor 2 depending on the size and shape. Bulk densities of 600-700 kg/m^3 are usual, which is 2-3 times the bulk density of the original chips or bundles [32].

Pyrolysis: Pyrolysis oil [34] is an intermediate product that can be combusted in a later stage to produce electricity (next Section). It can also be converted, via gasification, into motor fuels [35]. The liquid form eases transfer operations, and the very high bulk density decreases the amount of ship movements required. Pyrolysis has attractive economics at small scale, and could be used decentred at the beginning of bioenergy chains [34].

3.3 Conversion to motor fuels or electricity

Eventually, at the end of the transport chain, biomass will generally be used for the generation of electricity or for the production of motor fuels (see Table 6. These conversions can also be done earlier in the chain, e.g. to decrease the transported volumes and ease handling.

Many ways to generate electricity from biomass are possible. Here we consider BIG/CC [36]. Although this technology is not yet commercially available, it does use available technology. It has higher investments, but also a better efficiency, compared to combustion and co-combustion and it therefore more suitable for large-scale application. The data presented in Table 6 assumes a dry and chipped feedstock.

Methanol and Fischer Tropsch [12; 13; 38; 39] derived via gasification and ethanol [40; 41] via hydrolysis-fermentation are suitable fuels for the transportation sector. Both the conversion of solid biomass and of the intermediate pyrolysis oil to methanol are considered here. Performance data are taken from Hamelinck and Faaij [12], but with subtraction of the pre-treatment operations drying and

Table 6. General modelling assumptions for conversion to electricity or methanol, as used in the chain modelling. Arrow (→) indicates parameter change from short to long term. Data on possible variations can be found in Hamelinck *et al* [11].

	Electricity[1)]	**Methanol**	
	BIG/CC	**From solid biomass**[2)]	**From pyrolysis oil**[3)]
Base scale (MW_{LHV} input)	105	356 → 1780	845
Base capital[4)] (M€)	90	213 → 571	480
Max scale	408		
Scale factor R	0.61	0.86	0.86
Load factor	90 %	90 %	90 %
O&M	4 %	4%	4 %
Lifetime	25	25	25
electricity use (kWh/tonne)		-15.3	225
efficiency or yield (HHV basis)	57 %	58 → 60 %	63 %
Product heating value ($GJ_{HHV}/tonne_{dry}$)		25.6	25.6
Product density (kg/m^3_{bulk})		790	790

[1)] Costs for the BIG/CC are derived from Faaij *et al.* [36]; above 408 MW_{LHV} input, multiple units are needed. The solid feedstock requires a moisture content of maximally 15 % and a particle size of maximally 30 mm.

[2)] Hamelinck [12] assumed a feedstock with HHV_{dry} 19.3 GJ/tonne, mc = 30 %. Here it is assumed that the biomass is dried to mc 10 % in a preceding operation. Also the feedstock is assumed to be grinded (< 30 mm) in a previous step. In methanol production, a small amount of electricity is co-produced. Ranges (→) indicate the short and long term performance; the long term assumes larger scale, advanced conversion technologies and general cost reductions through learning.

[3)] Methanol from heavy residue gasification [37], has an investment of 380 $MUS\$_{1995}$ or about 480 $M€_{2003}$ for an installation of 660000 tonne/yr methanol, or 16.9 PJ_{HHV}. The efficiency is 63 % LHV (~ HHV). Accounting for the 90 % load, the input is thus 945 MW_{HHV} pyrolysis oil or 845 MW_{LHV}. Energy consumption is 225 kWh/tonne feed.

[4)] This is the total capital requirement. Most of the sources give Total Plant Costs (TPC) or total capital investment (TCI), but neglect the investment path during construction. Assuming a three year construction period (20 % of investment in first year, 30 % in second and 50 % in last year), the TCR is 118 % of the TCI. In the fourth year, the plant starts production at the indicated load.

sizing, so that pre-treatment operations can be decoupled from final conversions and be performed at an earlier stage in the bioenergy chain.

3.4 Transport

Truck transport (Table 7) is generally applied for relatively short distances (< 100 km), when flexibility is required because multiple (small) production sites have to be accessed, or when train and ship infrastructure is absent. The costs of truck transport consists of the total driving and loading/unloading costs, these in turn depends on the number of trips and the trip's distance. The driving costs consist of flat km costs (charter) and fuel costs. Both will differ per region, but only differences in regional fuel costs are incorporated (refer to Table 3). Because overloading has an immediate effect on forest roads condition, the gross vehicle weight between roadside and terminal is restricted (e.g. 38 tonne in the UK, [42]). Both transport from the forest to a terminal and from a terminal onwards to a harbour are likely to be dedicated: the trucks are only used for biomass transport and are thus empty on their return.

Train transport is applied for the longer overland distances (>100 km). It may be a serious competitor to ship transport for the middle distance international transports, because transfer points in harbours are not needed (refer to Table 2). It is difficult to gain insight in the actual costs of rail transport,

Table 7. Selection of data for transportation means, for both solid biomass and liquid energy carriers.

	Truck[1]	**Train**[2]	**Ship**[3]	
	(Solids … liquids)	**(Solids … liquids)**	**Small**	**Large**
Cargo capacity (tonne)	40 … 25	1,000	4,000	63,000
Cargo capacity (m^3)	130 … 33	2,500	6,700	105,000
Investment (M€)			11.4	23.8
O&M			10 %	8 %
Lifetime			25	25
charter costs (€/occasion)			4.9	10.1
charter costs (€/km)	0.85 … 1.24	8450/d + 4 … 10560/d + 5		
Other charges (€/trip)			4.8	75.6
Speed average (km/h)	65	75	27.8	27.8
Energy-e (kWh/km)		163		
Energy-f (MJ_{HHV}/km)	18.1		647	2517
Load/unload speed (m^3/hr)	260 … 500	240 – 500	60	300
Load/unload costs (€/m^3)	0.5	0.23		
Load/unload costs (€/tonne)			7.4	2.0

[1] Parameters shown hold for the transport of logs, bales, chips and pellets on one hand and liquids on the other [10; 18; 21; 24; 34]. Transport of pyrolysis oil is done in carbon steel tanks with lining, these cost 14 % more than normal carbon steel tanks [40].

[2] Transportation by train gets relatively cheaper on longer distances (d in km) [24]. Transportation of conventional liquid fuels (per tonne) is assumed to be 25 % higher than for solid fuels [24], and the transport of pyrolysis oil costs another 14 % more (see note 1). Electrical energy use is based on the transport of wood, which is known to consume 0.08 MJ/km per GJ transported, similar numbers were found in different literature sources. In some countries train transport will be fuelled with diesel.

[3] Presented values hold for solid biomass. Ships can be owned, hired on trip basis or hired on time basis, but the costs presented here are valid for ownership. The price of ships is very dependent on the market, but some generalisations can be made and the price is found to be quite proportional to the cargo capacity [11]. Tankers are more expensive than bulk carriers on cargo tonne basis (but much cheaper on transported energy basis). For transporting pyrolysis oil, lining is necessary and adds 10 % to the costs of a crude tanker. Fuel HFO use per km is a function of both size and speed (see Figure 3).

because of differences in logistic conditions between countries. Costs often depend on the availability of return-freights, the total volume of transport in the same direction, transfer terminal policies and the route. For example: a container transport from Rotterdam to Paris may cost four times as much as from Rotterdam to Barcelona [43]. For the present analysis generic km-costs and electricity use [24] are assumed for all concerned regions; only differences in the electricity costs yield regional differences.

Sea transport is applied for the long distances. It has low variable costs and a low energy use per tonne·km compared to other transport means. Ocean ships exist in a wide capacity range, from less than one to several hundred thousand tonnes deadweight‡ (dwt). The bigger the cargo capacity of a vessel, the more efficient a transport can take place. The best suitable size of a vessel follows from a trade-off between e.g. port charges, tugging, velocity, and cargo capacity. For the large logistic scales and logistic routes considered here, large vessels are employed. For transport from Eastern European countries, this may be a Handymax bulk carrier (35-50 thousand tonnes dwt), for South America a Panamax (50-80 thousand) is the largest vessel that can reach the regional harbours [45]. There are generally two vessel types: dry bulk carriers (for steel, iron ore, coal, grain) and tankers (for crude, petroleum products, etc.). Oil/bulk/ore (OBO) carriers are designed for a variety of cargos. Ship prices – especially for tankers – change heavily over time, being very dependent on the oil market and recently

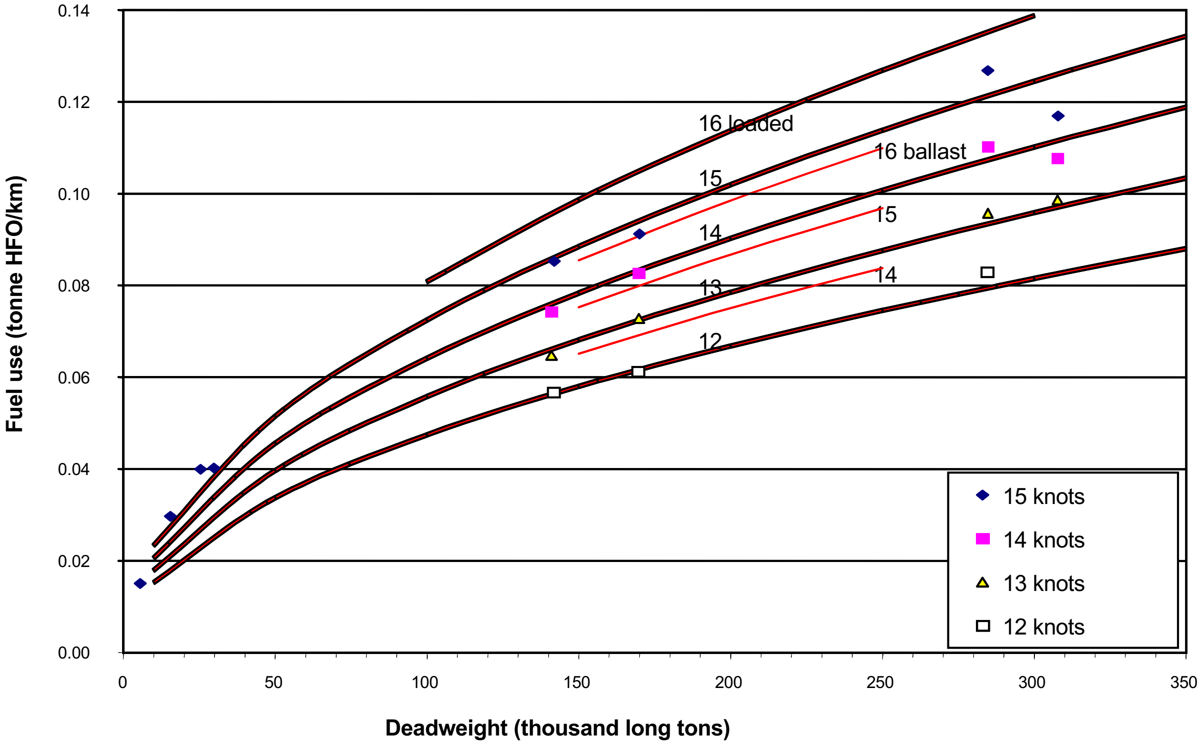

Figure 3. Fuel use in tonne HFO/km, at full draft and ballasted (~ 1/3 dwt), as a function of the ship size. Data stems from Suurs (5-25 kton dwt vessels [10]), and from the vessels "Front Granite" (Suezmax), "Front Breaker" (Suezmax OBO), "Front Vanadis" (VLCC) and the "Front Eagle" (VLCC) [44].

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‡ Deadweight is the lifting or carrying capacity of a ship when fully loaded, expressed in long tons or metric tonnes. It thus includes cargo, bunkers, water, (potable, boiler, ballast), stores, passengers and crew. The deadweight of a ship varies with season and water sort. In this study we use the summer deadweight: the ship is in salt water and loaded to the summer load line mark. The cargo capacity is about 85 % of the dwt for bulk carriers and 95 % for tankers.

facing stricter safety regulations.

In practice, ships are often hired on charter basis for a specific voyage (including bunker, canal tolls and port charges) or defined time. However, the charter business is an opaque field where prices change with routes, time, and market; generic prices cannot be given. Therefore, for an established biomass market, cost for a longer time period can best be derived from ship prices. Costs for various sizes of new and second hand bulk carriers and crude carriers [10; 46-48] have been fitted to yield ship prices quite proportional to the cargo capacity.

Operating costs are made when the vessel is in service. At sea, these vary with distance, in port with loading/discharging time. The consumption of liquid fuel§ is found to be a function of the ship size and its speed (see Table 7 and Figure 3). Port charges are assumed to amount 0.6 - 1 €/tonne dwt [45; 49] for each port on the voyage. Costs for loading and unloading cargo depend on the type (piece or bulk), form, density and amount of cargo, varying from 1.6 €/tonne for liquids to 10.1 €/tonne for bales [10]. Other O&M costs (crew salaries, insurance) are taken as a percentage of the investment, from 5 % for 200 thousand tonne dwt bulk carriers to 10 % for small vessels [10; 50].

Transport of gases/liquids through pipelines or electrical energy through (high voltage) power lines is not considered in this study. These options could be analysed in similar ways, provided that sufficient information is available.

4 Results

4.1 Production areas and transport distances

Arbitrarily, the first truck distance (surface average) from the roadside to a (central) gathering point is taken limited to 50 km (Table 8), which limits the accessible area to $16 \cdot 10^3$ km^2 (half of the area is closer by than 50 km). In the case of forest residues, which have a low spatial distribution (about 0.6 tonne$_{\text{fresh}}$/km^2, see Table 4), this limits the chain input from one production area to 3.1 MW$_{\text{HHV}}$. The second transport to the CGP or an inland harbour is limited to 100 km (average), which means that the total exploited area is $62 \cdot 10^3$ km^2 (logs: 37.7 ktonne$_{\text{fresh}}$/yr) so that four adjacent smaller areas supply the CGP, and the total front-end capacity is 12.5 MW$_{\text{HHV}}$. The GP can also be a transfer points to rail transport. In that case the production areas are not necessarily close to each other, and their number is not limited by the distance of each GP to a CGP.

For bio energy crop feedstock, first truck-transport distances are calculated for 300 MW$_{\text{HHV}}$ (9.5 PJ$_{\text{HHV}}$) 600 and 1200 MW$_{\text{HHV}}$. For Scandinavia and Eastern Europe this is 0.77 Mtonne$_{\text{fresh}}$/yr and up. The increasing energy crop yield per hectare (see Table 4) yields short distances, even for the largest systems. In calculating these distances, closed planting is assumed (100 % of the area is used for energy cropping), but with the 100 km CGP to a harbour distance, the production can as well take place on multiple less dense planted patches.

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§ Also known as residual fuel oil, bunker C, heavy fuel oil, or HFO.

The small scale and the use of inland harbours in residue chains, limit the ship size to 4000 tonne cargo. The ship size for larger systems is assumed to be Panamax (63,000 tonne). The international transport distance from Scandinavia and the Baltic countries to the Netherlands is taken 1100 km [8]. The international transport distance is 800 km by train from Poland, 1600 km by train from Ukraine, and 2000 km by ship from Ukraine. The international transport distance from Latin America is set at 11,000 km.

Table 8. First truck-transport distances[1)] for inland production areas, for various biomass sources and input scales, calculated from the data in Table 4. The increase in land productivity, from short to long term, decreases the surface required to grow the same amount of biomass, this is indicated by arrows (→).

	12.5 MW_{HHV}	300 MW_{HHV}	600 MW_{HHV}	1200 MW_{HHV}
Scandinavian forest residues[2)]	50 km			
Scandinavian energy crops		18 → 14 km	26 → 19 km	36 → 27 km
Eastern European energy crops		14 km	19 km	27 km
Latin American energy crops		14 → 13 km	20 → 18 km	29 → 26 km

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[1)] The shortest average bird flight distance from all points on the surface to a central point, is assumed to be expressed by $\frac{1}{2}\sqrt{2}$ times the radius of a circle with the same surface A, or $\sqrt{(A/2\pi)}$ [36]. If the “central” point lays at the border of an area (coastal CGP, expressed as the center of a circle of which only half of the surface is used), than the shortest average distance is expressed by $\sqrt{(A/\pi)}$.

[2)] Four areas smaller areas have a 50 km (surface average distance) truck transport to their gathering point, from where the biomass is transported 100 km to a central gathering point or inland harbor.

4.2 Mass flows

Moisture content and dry matter losses depend on the applied technology and the order of treatments within the chain. As an example, for European residues, transported as pellets by ship, and eventually converted to methanol, the chain mass balance is given in Table 9. The amount of pellets that can be transported by ship is restricted by the weight in this example, because the density is larger than the capacity mass/volume ratio of the ship. If, however, the ship transport takes place before densification of the biomass, then the volume would be restrictive and about 2½ - 3½ times more ship movements

Table 9. Chain mass balance for pellets transported by ship (from residue CRLs, Europe, available at roadside) with an annual front-end scale of 1 $Mtonne_{fresh}$ (~460 MW_{HHV}).

Chain structure	Step dry matter loss	Moisture content	Bulk density (kg/m^3_{bulk})		Logistic capacity	
				(Mtonne)	($Mtonne_{dry}$)	10^6 m^3
Residue bales at roadside		30 %	251	1	0.70	3.98
Storage pile at roadside	3.5 %	30 %	251	0.97	0.68	3.86
Truck transport 50 km		30 %	251	0.97	0.68	
Storage CGP	2.0 %	30 %	251	0.95	0.66	3.78
Chipping	2.0 %	30 %	240	0.93	0.65	3.88
Drying	1.0 %	7 %	192	0.69	0.64	3.59
Densification		7 %	650	0.69	0.64	1.06
Storage bunker at CGP	2.0 %	7 %	650	0.68	0.63	1.05
Truck transport 100 km		7 %	650	0.68	0.63	
Storage bunker in harbour	2.0 %	7 %	650	0.66	0.62	1.02
International shipping	0.3 %	7 %	650	0.66	0.62	
Truck transport 100 km		7 %	650	0.66	0.62	
Storage pile at conversion plant	2.0 %	7 %	650	0.65	0.60	1
Conversion to methanol	53 %	0 %	790	0.28	0.28	0.35

would be necessary. At the other hand, early conversion to methanol would decrease the number of ship movements needed by 40 - 50 %.

4.3 Economy

For the short distance the decrease in logistic capacity influences the costs of transport steps, but is not decisive for the costs of the whole chain. This can be seen in Figure 4: the transportation costs (truck + ship) of the 'pellets by ship' scenario are only half of that of 'bales by ship'. However, the costs of the original biomass contribute strongly to the delivered costs and the costs for densification cancel out half of the avoided transportation costs. For logs and pellets, rail and sea transport perform the same: the shorter truck distances are outweighed by the higher costs for using trains. Materials with densities below 300 kg/m^3_{bulk} (especially chips) are cheaper transported by ship. Storage and sizing have a very limited effect on the total chain costs.

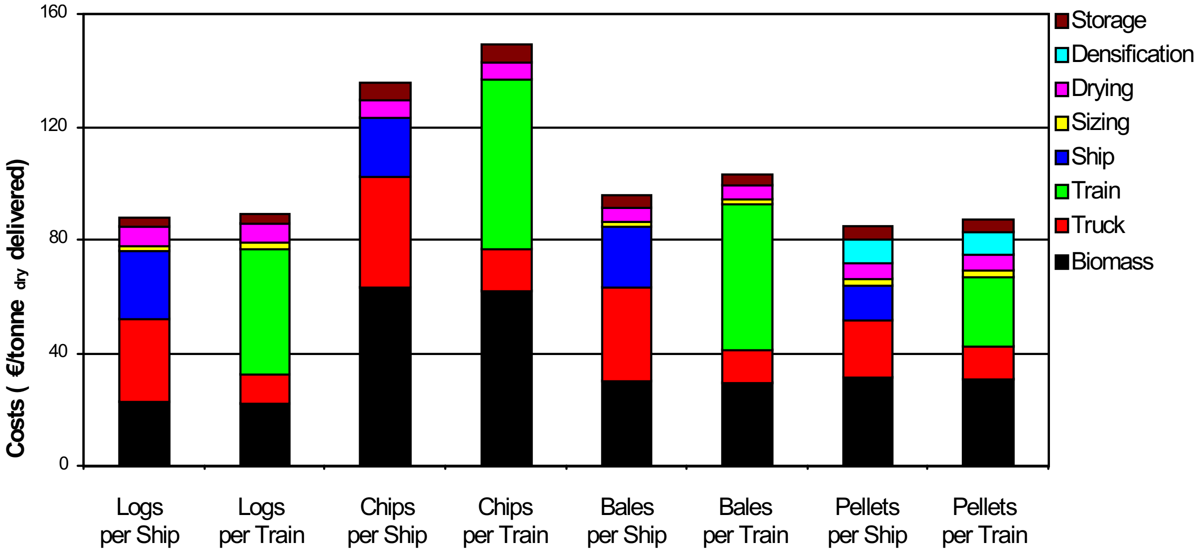

Figure 4. Breakdown of costs for Scandinavian forest residues delivered as solid biomass to a power plant in the Netherlands, transported as different commodities.

In Figure 5, it is shown that biomass from present Western European energy plantations (Scandinavia) suffers from high production prices, which cannot be compensated by the lower logistic costs. Eastern European (Ukrainian) bio energy crops are available at lower prices, and with modest transport costs they can be delivered cheaper than residues. Latin American biomass, assumed available at a very low price, has low transportation and handling costs at the scale considered. Especially the longest transportation step, Atlantic shipping, profits from the large scale. Truck transport gets slightly more expensive, naturally because of the larger supply area. Latin American biomass delivered at a Dutch power plant is calculated to cost about 60 €/$tonne_{dry}$, or 3.1 €/GJ_{HHV}, two thirds of the price for European residues and one half of Scandinavian Energy Crops. Logistic costs and losses (13 % of dry matter) for large-scale long distance transport adds 2.0 €/GJ_{HHV} to the roadside biomass price ($^1/_3$ to $^2/_3$ of this 2.0 €/GJ_{HHV} is in road truck transport). Future Latin American production cost reductions are only weakly visible in the delivered costs. Eastern European energy crops may cost about 3.6 €/GJ_{HHV}

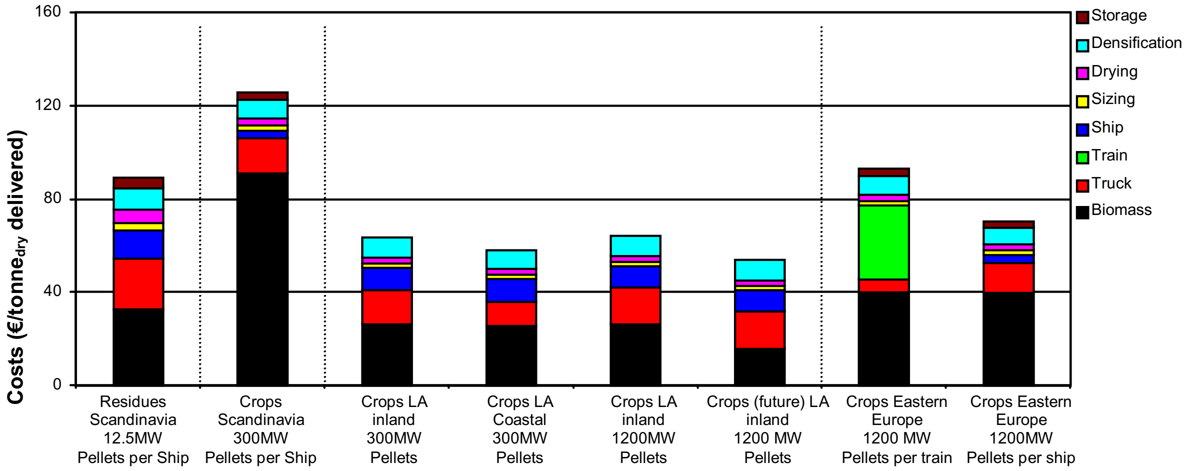

Figure 5. Cost breakdown of solid biomass delivered to a power plant in the Netherlands (Scandinavian forestry residues, Scandinavian, Latin American (LA) and Eastern European energy crops). The main transportation step is by ship.

delivered. Despite the longer sea than overland distances, ship transport is preferred for distances over 1100 km.

Results for electricity delivering chains are shown in Figure 6. For electricity produced from Scandinavian residues, the cost differences between different transport chains are less pronounced, 0.09 – 0.12 €/kWh, than for the delivered biomass itself, because at this small scale the cost for final conversion contributes 50 – 70 % of the power costs. The 0.09 – 0.12 €/kWh resembles the range found be Agterberg and Faaij [8]. However, conversion costs prevail in the present cost breakdown (smaller scale), where in Agterberg and Faaij the biomass feedstock and international shipment are more costly.

Conversion to pyrolysis oil and from pyrolysis to power (co firing) is so much cheaper than the BIG/CC option at this small scale that it fully compensates the low chain efficiency. This can be seen

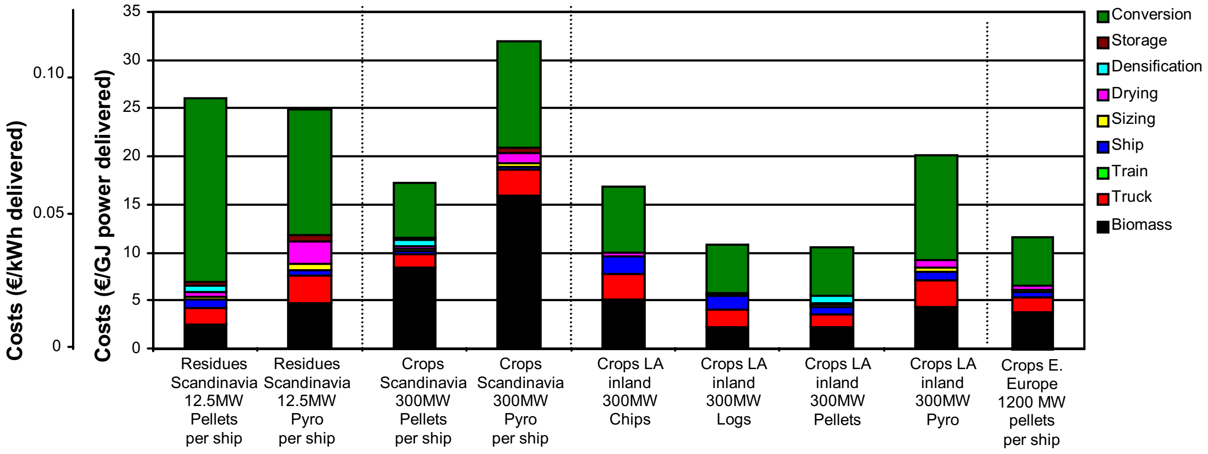

Figure 6. Cost breakdown of electricity delivered to the Dutch grid, generated from Scandinavian forestry residues, Scandinavian, Latin American (LA) and Eastern European energy crops.

from the doubled biomass and truck transport costs in the residue pyrolysis chain, compared to residue pellets. In either case the eventual electricity delivered costs about 0.09 €/kWh. The low efficiency of pyrolysis chains is a serious obstacle at larger scales, where the two conversion steps (pyrolysis and power generation) barely profit from scale. Electricity produced by BIG/CC from Latin American biomass may cost as little as 0.04 €/kWh. Agterberg and Faaij find much higher costs (0.09 €/kWh for logs from Ecuador), but that can be explained from their higher biomass feedstock costs (2.0 instead of 1.1 €/GJ_{HHV}), much smaller logistic scale (30 instead of 300 MW_{HHV} input), and much lower biomass to electricity conversion efficiency (40 instead of 55 %).

Results for chains delivering methanol are shown in Figure 7. When producing methanol from Scandinavian biomass, the cost advantage of the larger scale possible with energy crops, compared to forestry residues, cannot compensate the higher price of these crops. Both methanol from forestry residues and energy crops are calculated to cost about 14-15 €/GJ_{HHV}. Eastern European biomass may deliver methanol at about 10 €/GJ_{HHV}. The costs may further be reduced to 8 – 9 €/GJ_{HHV} when using Latin American bioenergy crops at an input scale of 300 MW_{HHV} and up. Larger scales do not logically decrease the system costs further, as most components have reached their maximum capacity. A very large methanol plant in the Netherlands (1.1 $GW_{HHV,input}$) is found to deliver at the same costs as four times smaller plants in Latin America, because of the higher costs of pellets shipping. However, if one methanol plant could be build in the exporting region, at the very large scale of 1200 MW_{HHV} front-end, the delivery costs may become below 8 €/GJ_{HHV}, which is a very attractive cost level compared to current gasoline and diesel production costs [51]. The methanol delivering cost found for Scandinavian and Eastern European biomass are similar to those found by Agterberg and Faaij for Sweden and Estonia. For Estonia, they found a larger share in conversion (small scale).

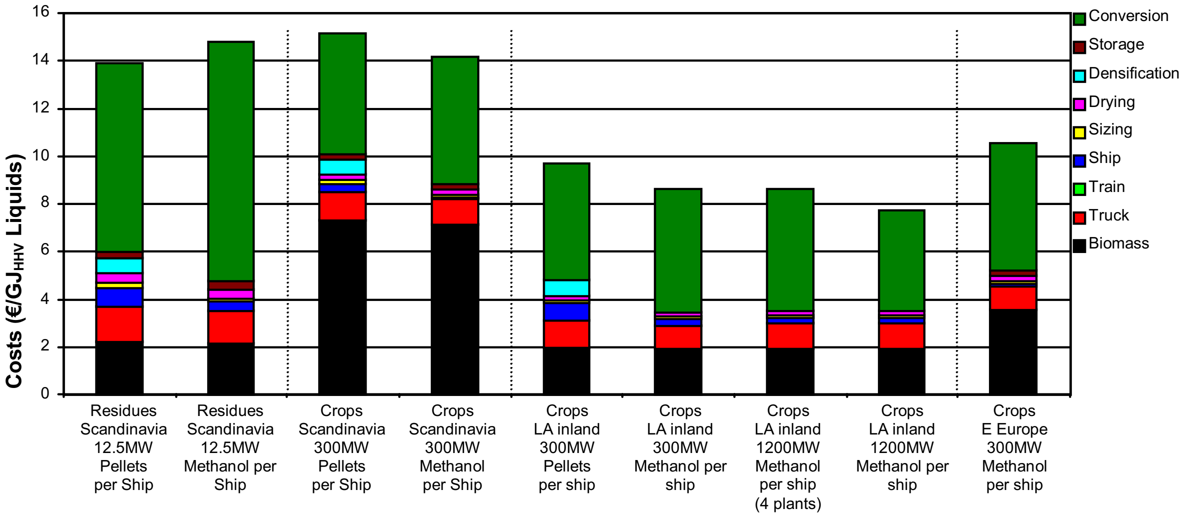

Figure 7. Cost breakdown of methanol delivered to a central distribution point in the Netherlands, produced from Scandinavian forestry residues, Scandinavian, Latin American (LA) and Eastern European energy crops.

4.4 Energy and CO_2

Energy use. Figure 8 shows the primary energy use for a crosscut of the discussed concepts. Residues start with substantial energy consumption in the collection step. The small ships in the main transport step are less efficient than the Panamax used for transporting energy crops, and the low density of chips decreases efficiency further. Trains have a high electrical energy use (without scale profit), and a low efficiency from primary energy to electricity. Still the primary energy use for residues chains can be as low as 8.5 % of the delivered HHV. Energy crops, due to their denser distribution and the high efficiency of cultivation and harvest, can be delivered at similar (Latin America) or even lower (Europe) primary energy consumption figures. The energy requirement for energy is higher, because part of the bioenergy is lost along the chain. Pellets chains loose 15 % of the initial energy, consequently the energy use is about 1.26 $MJ_{primary}/MJ_{delivered}$ for European residue pellets, and 1.23 $MJ_{primary}/MJ_{delivered}$ for energy crop pellets from Eastern Europe. For comparison, the energy requirement of coal is 1.05-1.10 $MJ_{primary}/MJ_{delivered}$ [19]. When methanol is the desired product (and transported form), the primary energy use can be as low as 5 – 14 % for European respectively Latin American situations. The total energy requirement is 1.87 –1.93 $MJ_{primary}/MJ_{delivered}$. If the biomass is transported as pellets both the primary energy use and the total energy requirement increase, because of decreased density in the transport step, energy use in the (additional) pelletising process, and extra material losses. The energy requirement for fossil oil products is much lower: 1.05 – 1.19 $MJ_{primary}/MJ_{delivered}$ (1 % in winning, 3 % in transport, 2 % in storage, and 5 – 10 % in refineries). The difference is mainly in the low conversion efficiency of biomass to methanol (58 %), and only for a small part in the transport.

CO_2 balance. The CO_2 emission of transportation chains is assumed a direct function of the (secondary) energy use and local efficiency parameters (see § 2.3). In that case the CO_2 amount released in conversion of the biomass and loss is considered to be compensated by the CO_2 caught in the harvested biomass at the beginning of the chain. Cultivated biomass crops transported in a dense form logically have the lowest emission (Figure 9). Larger scales may further improve this performance, while

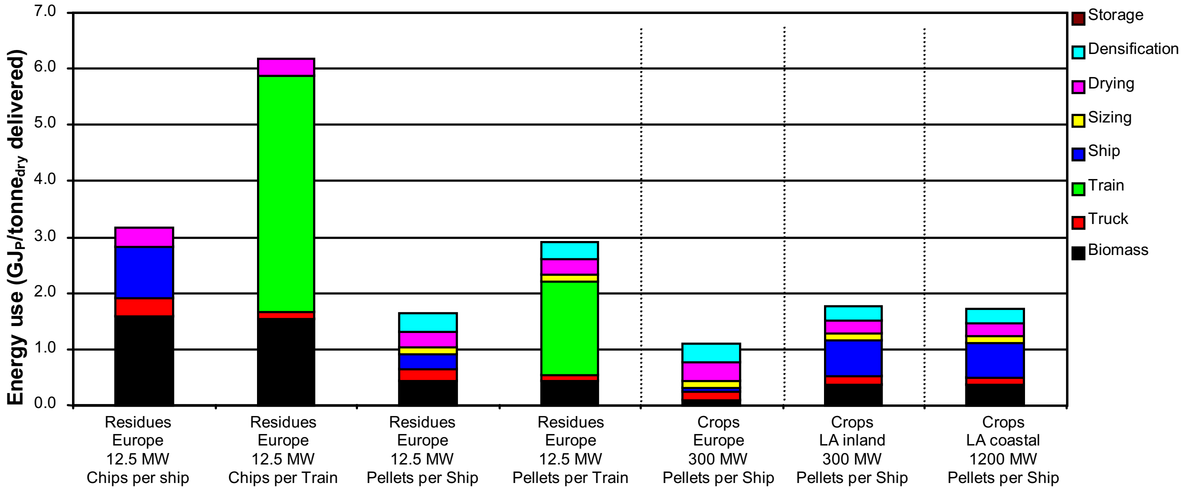

Figure 8. Breakdown of primary energy use for chains delivering solid biomass (Scandinavian forestry residues, Scandinavian, Latin American (LA) and Eastern European energy crops) to a Dutch power plant. Bioenergy in lost biomass and conversion efficiencies affect the amount of biomass delivered, instead of the chain's energy use.

longer distances lead to higher emissions. Electricity from European (or Latin American) pellets can be produced against 2 (or 9) kg CO_2/GJ_e or 7 (32) kg/MWh. Compare this to 370 – 1200 kg CO_2/MWh when electricity is produced from gas (at $\eta = 55$ %) or coal ($\eta = 28$ %).

Carbon storage in soil and litter depends on local climate, soil type, biomass species and harvesting method. The closed cycle is a justified assumption when plantations have reached an equilibrium state and do not sequester carbon in the soil, or extract it from the soil. But when starting a plantation the carbon storage per ha will change, because the land use is changed. The annual CO_2 sequestration in soil and litter decreases to zero after a 10-30 year period for production forests of short (<10 years) rotation, and the carbon content of soil plus litter becomes constant [52]. Forests planted on former agricultural plots sequester C in litter and soil during the first 10-30 years. Where plantations replace existing forest, the C that was stored in the replaced forest is released immediately. Furthermore C is released over time from the existing soil and litter because of the differences between the replaced and production forest. Here also a 10-30 year period is needed to reach equilibrium. The net CO_2 fixation or release can thus be considered as one cumulative number over the plantation lifetime, and be recalculated to an annual CO_2 credit/debit. However, the exact carbon fixation in a plot before and after land use change is impossible to estimate if the local situation is unknown. Assuming a generic cumulative carbon debit of 130 tonne carbon/ha (Estimated from exemplary figures in [52]) when a plantation replaces forest, and a plantation lifetime of 100 years, the CO_2 debit is 0.2 $tonne_{CO2}/tonne_{fresh}$. This adds about 130 kg CO_2/MWh bio-electricity. When a plantation is built on formerly agricultural land, the credit is about 20 tonne carbon/ha, or 0.03 $tonne_{CO2}/tonne_{fresh}$, or about 19 kg CO_2/MWh. Obviously this CO_2 debit/credit from land use change must play an important role in evaluation of biomass supply chains.

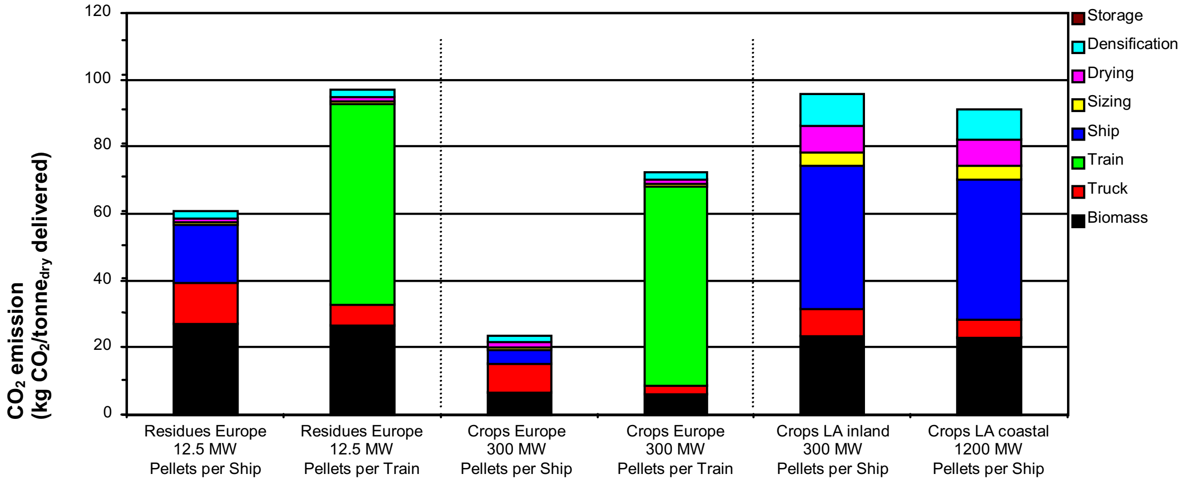

Figure 9. CO_2 emissions for chains delivering pellets from Scandinavian forestry residues, Scandinavian, Latin American (LA) and Eastern European energy crops, to a Dutch power plant.

5 Summary and Conclusion

To supply biomass to energy importing regions (Western Europe), long distance international transport is necessary. This implies extra costs, energy consumption, material loss and complex logistics. A broad variety of supply chains was envisioned and compared, including transport of raw biomass (logs, bales, chips), refined biomass (pellets, pyrolysis oil) and high quality liquid biofuels (methanol). In a modular approach the costs, energy use and CO_2 emissions of each step in the selected chains were calculated, and the resulting biomass characteristics (amount, moisture, composition…) were used for the next step. In this study we focussed on biomass from Scandinavia, Eastern Europe and Latin America, transported to the Netherlands. The applicable transport links were approached in a generic way.

Typical dry matter losses for solid biomass transports are about 15 %, mostly depending on the amount of storage steps. Densification strongly reduces the number of transport movements, although at a certain point the weight becomes restrictive instead of the volume and further densification becomes redundant. On the shorter distance (European biomass residues) the total costs are dictated by the (present) high biomass production costs, and the relatively high truck transport costs. The first truck transport in the chain, from forest to gathering point, is a dedicated transportation step (no return freight) and with large spatial spreading, the travelled distances are quite long or the applicable scale remains small. International ship transport only makes out a modest part of the total costs. On the other hand, train transport is so expensive that it neutralises the gains from bypassed transport steps. Overall chain costs do not get cheaper at larger scales, since the supplied biomass (crops instead of residues) is more expensive and truck transport efficiency does not improve. On the longer term, improved production methods (reduced energy use, and higher yield per area, and larger scale), and a shift to production in Eastern Europe, may reduce biomass and truck transport costs.

Crops are produced in Latin America against much lower costs. Solid biomass delivered at a Dutch power plant may cost about 40 €/tonne$_{dry}$, or 2.1 €/GJ$_{HHV}$, half of the price for European residues and one third of European Energy Crops. Electricity produced by BIG/CC from 300 MW$_{HHV}$ Latin American biomass may cost as little as 3.5 €cent/kWh, competitive with fossil electricity. Methanol produced in Latin America delivered to Europe may cost 8 – 10 €/GJ$_{HHV}$, depending on scale. Production in the Netherlands – even at larger scales – hardly reduces the costs. Future cost reductions do reside in both cheaper delivered biomass (above) and technological learning in methanol production. Methanol delivering costs would be the lowest when a very large conversion facility is build at the export harbour, with the advantage of flexibility in supplying the eventual fuel market which may be anywhere around the world.

When comparing the analysed chains, the importance of densification becomes clearer for chains with larger scales and longer distances, although the transport of loose chips should always be avoided. Increasing local biomass productivity (tonne/ha) allows for an increasing scale of the entire system, because the relatively expensive local truck transports limit the first-transport distance. The location of production sites near the coast reduces the amount of transport steps, but also limits the production area size, and the resulting cost reduction is therefore small. The relative importance of ship transport costs decrease with distance, when the scale increases. Biomass production costs remain decisive for the final bioenergy costs, still accounting for 25 – 40 % of the delivered biomass costs of cheap biomass (front end 0.7 – 1.1 €/GJ_{HHV}) in large-scale long-distance chains.

Energy use in and CO_2 emissions from supply chains are critical aspects in discussing bioenergy sustainability. The energy requirement for energy is ~1.25 $MJ_{primary}/MJ_{delivered}$ for European energy crop pellets. Part of the "extra" energy is fossil fuel and electricity input, part is biomass loss. The CO_2 performance is calculated direct from the primary energy use, but reckoning with local fuel mixes and efficiencies. Electricity from European or Latin American pellets can be produced against 2 or 9 kg CO_2/GJ_e, or 7 or 32 kg/MWh). A reduction of over 90 % compared to electricity produced from fossil fuel. The changing carbon storage in soil and litter as a result of changed land use is not accounted for, assuming an equilibrium situation or minimum impact. However, placing the plantation on former forestland could upturn the picture.

Other relevant sustainability aspects of bio-energy production and transport are the effects for local economies, local environment and the safety of long distance transport. These effects have not been evaluated here, but deserve attention too in discussions about sustainability of biomass energy consumption. Local GDP and employment may profit from bio-energy plantations, and electricity costs can go down if part of the biomass is used locally [27]. Van den Broek also lists a whole range of potential environmental impacts: emissions from processing (other GHG gases, SO_2 and dust) and direct impacts from cropping (erosion, loss of soil quality, groundwater eutrophication, emission of toxic substances, groundwater depletion and loss of biodiversity). For evaluation, the alternative land use should be known and well defined. Furthermore, the relative importance of the impacts is subjective. Shipwrecking and spills are important safety issues that should be assessed when considering ship transport of pyrolysis oil, methanol or pellets.

The use of biomass for energy leads to large CO_2 reductions per unit of energy produced, compared to fossil fuels. Biomass can be gathered or produced at large scale against favourable costs, and transportation over long distances should not be considered as an obstacle. The eventual costs of electricity may be competitive with present day fossil electricity. Biofuels remain slightly more expensive than fossil automotive fuels, but the gap can probably be bridged when system scales are increased and bio-processing technology improves. Governments should stimulate this development so that the biomass market size grows, dedicated energy crop plantations emerge and eventually bioenergy can significantly contribute to a sustainable society. International bio-energy trade has very promising prospects and could be a key component of the future world's energy system.

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Paper 2

Future prospects for production of methanol and hydrogen from biomass

Contents

Abstract **85**

1 Introduction **86**

2 Production of biofuels **87**

2.1 Production and conditioning of synthesis gas 87

2.2 MeOH Production 90

2.3 H_2 production 92

2.4 Electricity co-production 93

3 Selected systems **95**

4 System calculations **96**

4.1 Modelling 96

4.2 System calculation results 100

5 Economics **102**

5.1 Method 102

5.2 Results 106

5.3 Biofuel FCV economy 110

6 Discussion and conclusions **111**

7 Acknowledgements **113**

References **113**

Future prospects for production of methanol and hydrogen from biomass[*]

Abstract

Technical and economic prospects of the future production of methanol and hydrogen from biomass have been evaluated. A technology review, including promising future components, was made, resulting in a set of promising conversion concepts. Flowsheeting models were made to analyse the technical performance. Results were used for economic evaluations. Overall energy efficiencies are around 55 % HHV for methanol and around 60 % for hydrogen production. Accounting for the lower energy quality of fuel compared to electricity, once-through concepts perform better than the concepts aimed for fuel only production. Hot gas cleaning can contribute to a better performance. 400 MW_{th} input systems produce biofuels at 8 – 12 US$/GJ, this is above the current gasoline production price of 4 – 6 US$/GJ. This cost price is largely dictated by the capital investments. The outcomes for the various system types are rather comparable, although concepts focussing on optimised fuel production with little or no electricity co-production perform somewhat better. Hydrogen concepts using ceramic membranes perform well due to their higher overall efficiency combined with modest investment. Long term (2020) cost reductions reside in cheaper biomass, technological learning, and application of large scales up to 2000 MW_{th}. This could bring the production costs of biofuels in the 5 – 7 US$/GJ range.

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[*] Modified version of Hamelinck CN and Faaij APC, 2002, Future prospects for production of methanol and hydrogen from biomass, Journal of Power Sources 111(1):1-22.

1 Introduction

Methanol and hydrogen produced from biomass are both well suited for use in Fuel Cell Vehicles (FCVs), which are expected to reach high efficiencies, about a factor 2-3 better than current Internal Combustion Engine Vehicles (ICEVs). In addition FCVs are quiet and clean, emitting none of the air pollutants SO_x, NO_x, VOS or dust. When methanol and hydrogen are derived from sustainably grown biomass, the overall energy chain can be greenhouse gas neutral. Such a scheme could provide a major alternative for the transport sector world-wide in a greenhouse gas constrained world [1-3].

Methanol and hydrogen can be produced from biomass via gasification. Several routes involving conventional, commercial, or advanced technologies, which are under development, are possible. Methanol or hydrogen production facilities typically consist of the following basic steps (see Figure 1): Pre-treatment, gasification, gas cleaning, reforming of higher hydrocarbons, shift to obtain appropriate H_2:CO ratios, and gas separation for hydrogen production or methanol synthesis and purification. Optional are a gas turbine or boiler to employ the unconverted gas, and a steam turbine; resulting in electricity co-production.

Many process configurations are possible, however. Gasification can be atmospheric or pressurised, direct or indirect, resulting in very different gas compositions; different options are available for gas cleaning, processing and purification; generation of power is optional. Altogether in theory a very large number of concepts to produce methanol or hydrogen is possible.

Previous analyses by Katofsky [1] and Williams *et al.* [4] have shown that methanol can be produced from biomass at 14 – 17 US$[†]/GJ (biomass delivered at 2.3 US$/GJ), with a net HHV energy efficiency between 54 and 58 %. Hydrogen production costs can be 10 – 14 US$/GJ, with a net HHV energy efficiency of 56 – 64 %. Those cost levels are not competitive with current gasoline and diesel production costs (about 4-6 US$/GJ [5]). The evaluations of Katofsky and Williams focused on technologies that are likely to be commercial on the short term. The scale of production was fixed on about 400 MW_{th}. Komiyama *et al.* [6] calculate hydrogen from biomass to cost 5.1 US$/GJ and methanol 5.2 US$/GJ; the biomass input is about 530 MW_{th} HHV, however a significant amount of energy is added as LPG and process efficiencies and biomass cost are not given. All this leads to the key question *whether advanced, future technologies , larger scales and alternative concepts may enable competitive production of methanol and hydrogen on longer term.*

Therefore, the key objective of this work is to identify biomass to methanol and hydrogen conversion concepts that may lead to higher overall energy efficiencies and lower costs on longer term. Improved performance may be obtained by:

- Applying improved or new (non commercial) technologies. Examples are the use of Autothermal Reforming (instead of steam reforming), improved shift processes, once through Liquid Phase MeOH process, high temperature gas cleaning, high temperature hydrogen separation and improved oxygen production processes.

[†] All costs are in $US\$_{2001}$.

- Combined fuel and power production by so-called 'once through' concepts. Combined fuel and power production may lead to lower cost and possibly higher overall thermal efficiencies because of cheaper reactor capacity and reduction of internal energy consumption of the total plant.
- Economies of scale; various system analyses have shown that the higher conversion efficiencies and lower unit capital costs that accompany increased scale may well outweigh increased energy use and costs for transporting larger quantities of biomass. Furthermore, it should be noted that paper & pulp mills, sugar mills, and other facilities operate around the world with equivalent thermal inputs in the range of 1000-2000 MW_{th}. Such a scale could therefore be considered for production of energy/fuel from (imported) biomass as well.

These strategies are explicitly taken into account in the present work:

1. Technology assessment (section two) and selection of various concepts (section three). The review includes technologies that are not applied commercially at present.
2. Consulting of manufacturers and experts to obtain or verify performance and cost data of various components.
3. Creation of Aspen+ models to evaluate performance of the selected process configurations, and carry out sensitivity analyses. Particular attention is paid to the heat integration of the concepts (section four).
4. Cost analyses based on component costs; including scale factors and capacity ranges (section five).
5. The work is finalised by an overall discussion and conclusion (section six).

2 Production of biofuels

2.1 Production and conditioning of synthesis gas

Syngas, a mixture of CO and H_2, is needed to produce methanol or hydrogen. A train of processes to convert biomass to required gas specifications precedes the methanol reactor or hydrogen separation – as depicted in Figure 1.

Gasification. Many gasification methods are available for syngas production. Based on throughput, cost, complexity, and efficiency issues, only circulated fluidised bed gasifiers are suitable for large-scale fuel gas production. Direct gasification with air results in nitrogen dilution, which in turn strongly increases downstream equipment size. This eliminates the TPS and Enviropower gasifiers, which are both direct air blown. The MTCI gasifier is indirectly fired, but produces a very wet gas and the net carbon conversion is low. Two gasifiers are selected for the present analysis: the IGT (Institute of Gas Technology) pressurised direct oxygen fired gasifier, and the BCL (Battelle Columbus) atmospheric indirectly fired gasifier. The IGT gasifier can also be operated in a *maximum hydrogen* mode, by increasing the steam input. Both gasifiers produce medium calorific gas, undiluted by atmospheric nitrogen, and represent a very broad range for the H_2:CO ratio of the raw fuel gas.

The main performance characteristics of both gasifiers are given in Table 2. The IGT gasifier produces a CO_2 rich gas. The CH_4 fraction can be reformed to hydrogen, or be used in a gas turbine. The H_2:CO ratio

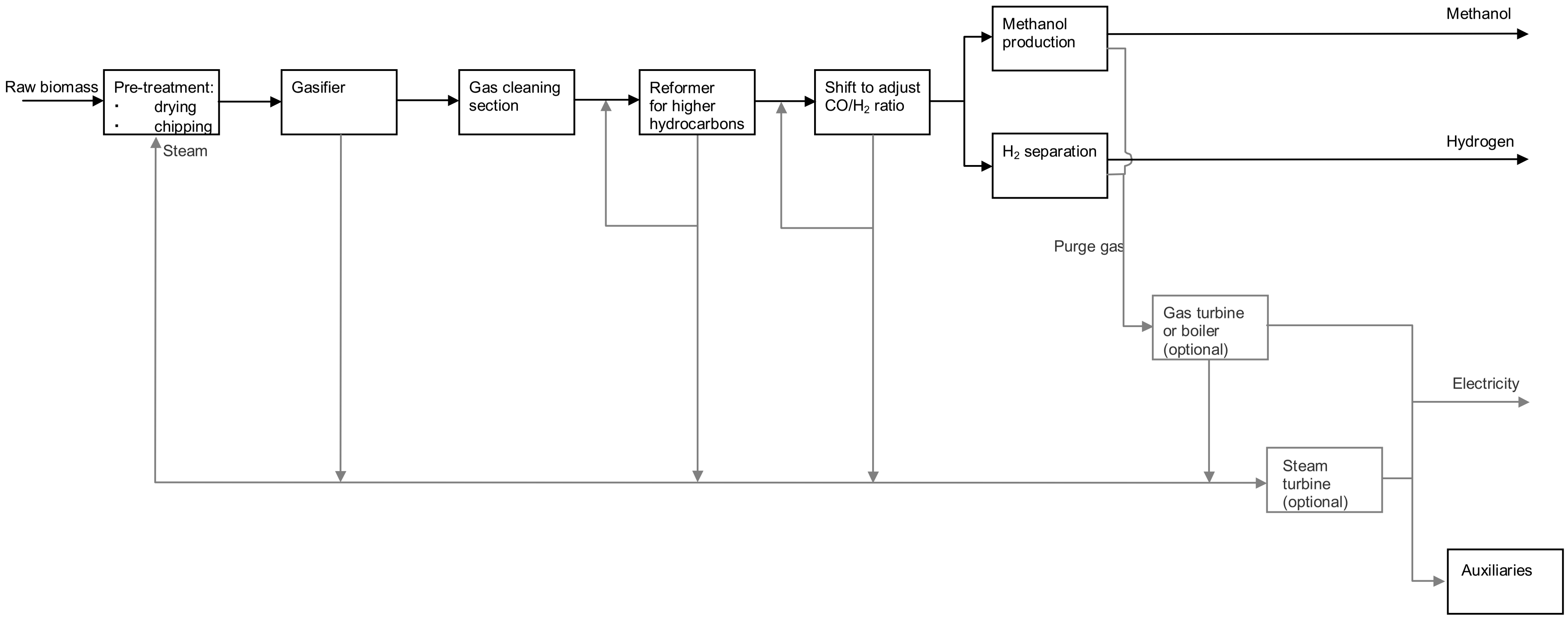

Figure 1. Key components in biomass to methanol/hydrogen production concepts.

(1.4 : 1) is attractive to produce methanol, although the large CO_2 content lowers the overall yield of methanol or hydrogen. For hydrogen production, the H_2:CO ratio should be shifted. The pressurised gasification allows a large throughput per reactor volume and diminishes the need for pressurisation downstream, so less overall power is needed. The maximum hydrogen mode is especially useful for hydrogen production, and also the H_2:CO ratio is still better for methanol production. However, the gasifier efficiency is lower and much more steam is needed. In both modes the IGT uses oxygen to reduce downstream equipment size.

The indirectly heated BCL is fired by air; there is no risk of nitrogen dilution nor need for oxygen production. It produces a gas with a low CO_2 content, but contains more heavier hydrocarbons. Therefore, reforming is a logical subsequent step in order to maximise CO and H_2 production. The tars present need to be cracked and the large CO fraction needs to be shifted to yield hydrogen. The reactor is fast fluidised, allowing throughputs equal to the bubbling fluidised IGT, despite the atmospheric operation. The atmospheric operation decreases cost at smaller scale, and the BCL has some commercial experience (demo in Burlington USA, [7]).

Gas cleaning. The produced gas contains tars, dust, alkali compounds and halogens, which can block or poison the catalysts downstream, or corrode the gas turbine. The gas can be cleaned using available conventional technology, by applying gas cooling, low temperature filtration, and water scrubbing at 100 – 250 °C. Alternatively, hot gas cleaning can be considered, using ceramic filters and reagents at 350 – 800 °C. The considered pressure range is no problem for either of the technologies. Hot gas cleaning is advantageous for the overall energy balance when a reformer or a ceramic membrane is applied directly after the cleaning section, because these processes require a high inlet temperature. However, not all elements of hot gas cleaning are yet proven technology, while there is little uncertainty about the cleaning effectiveness of low temperature gas cleaning. Both cleaning concepts are depicted in Figure 2.

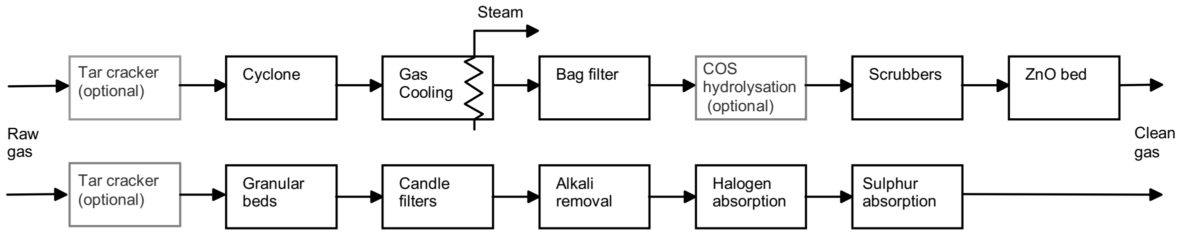

Figure 2. Conventional low temperature wet cleaning (top) and advanced high temperature dry cleaning. The tar cracker is required after atmospheric gasification. COS hydrolysation becomes redundant when Amine technology is applied for CO_2 removal downstream [8].

In low temperature wet cleaning [1; 8-14], particulates are completely removed by the cyclone, the bag filter and the scrubbers. Essentially all alkali and the bulk of sulphuric and nitrogenous compounds are removed by consecutive scrubbers. The ZnO bed or solvent absorption unit brings the sulphur concentration below 0.1 ppm. The effectiveness of cold gas cleaning has been proven for coal gasification combined cycle and Fischer-Tropsch synthesis applications [8]. Hot gas cleaning [1; 8; 10; 13; 15-22] removes particles for about 99.8 % by granular beds and ceramic candle filters. Simultaneously SO_x and NO_x are removed by injection of sorbents. Alkali removal via physical adsorption or chemisorption can be implemented at 750 – 900 °C, although lead and zinc cannot be removed at this temperature. After sulphur is removed by chemisorption, 99.5 % of the NH_3 can be decomposed over a nickel catalyst (sensitive to H_2S). Only HCN may be insufficiently removed by hot gas cleaning, leading to shorter catalyst life in downstream reactors.

Tijmensen [8] assumes maximum acceptable values of the contaminants for catalysts and equipment to lie in the 10 – 20 ppb range. The proposed cleaning technologies are appropriate and sufficient to meet these constraints.

Syngas processing. The syngas can contain a considerable amount of methane and other light hydrocarbons, representing a significant part or the heating value of the gas. Steam reforming (SMR) converts these compounds to CO and H_2 over a nickel catalyst. Autothermal reforming (ATR) combines partial oxidation in the first part of the reactor with steam reforming in the second part, thereby optimally integrating the heat flows. It has been suggested that ATR, due to a simpler concept, could become cheaper than SMR [1], although others give much higher prices [23]. There is dispute on whether SMR can deal with the high CO and C+ content of the biomass syngas. Where Katofsky writes that no additional steam is needed to prevent coking or carbon deposition in SMR, Tijmensen [8] poses that this problem does occur in SMR and that ATR is the only technology able to prevent coking.

The syngas produced by the BCL and IGT gasifiers has a low H_2:CO ratio. The water gas shift (WGS) reaction is a common process operation to shift the energy value of the CO to H_2, which can than be separated using pressure swing adsorption. If the stoichiometric ratio of H_2, CO and CO_2 is unfavourable for methanol production, the water gas shift can be used in combination with a CO_2 removal step. The equilibrium constant for the WGS increases as temperature decreases. Hence, to increase the production to H_2 from CO, it is desirable to conduct the reaction at lower temperatures, which is also preferred in view of steam economy. However, to achieve the necessary reaction kinetics, higher temperatures are required [24; 25].

2.2 MeOH Production

Fixed bed technology. Methanol is produced by the hydrogenation of carbon oxides over a Cu/Zn/Al catalyst. The synthesis reactions are exothermic and give a net decrease in molar volume. Therefore the equilibrium is favoured by high pressure and low temperature. During production, heat is released and has to be removed to maintain optimum catalyst life and reaction rate. The catalyst deactivates primarily because of loss of active copper due to physical blockage of the active sites by large by-product molecules, poisoning by halogens or sulphur in the synthesis gas, and sintering of the copper crystallites into larger crystals [26].

Conventional methanol reactors [26; 27] use fixed beds of catalyst pellets and operate in the gas phase. Two reactor types predominate in plants built after 1970. The ICI low-pressure process is an adiabatic reactor with cold unreacted gas injected between the catalyst beds (Figure 3, left). The subsequent heating and cooling leads to an inherent inefficiency, but the reactor is very reliable and therefore still predominant. The Lurgi system (Figure 3, right), with the catalyst loaded into tubes and a cooling medium circulating on the outside of the tubes, allows near-isothermal operation. Conversion to methanol is limited by equilibrium considerations and the high temperature sensitivity of the catalyst. Temperature moderation is achieved by

Figure 3. Methanol reactor types: quench (left) and steam raising (right).

recycling large amounts of hydrogen rich gas, utilising the higher heat capacity of H_2 gas and the higher gas velocities to enhance the heat transfer. Typically a gas phase reactor is limited to about 16% CO gas in the inlet to the reactor, in order to limit the conversion per pass to avoid excess heating.

Slurry technology. Processes under development at present focus on shifting the equilibrium to the product side to achieve higher conversion per pass. Examples are the gas/solid/solid trickle flow reactor, with a fine adsorbent powder flowing down a catalyst bed and picking up the produced methanol; and liquid phase methanol processes where reactants, product, and catalyst are suspended in a liquid. In liquid phase processes [26; 28] heat transfer between the solid catalyst and the liquid phase is highly efficient, thereby allowing high conversions per pass without loss of catalyst activity. Different reactor types are possible for liquid phase methanol production, such as a fluidised beds and monolithic reactors. The slurry bubble column reactor of the LPMEOH process (registered trademark of Air Products and Chemicals, Inc.; Figure 4) was invented in the late 1970s and further developed and demonstrated in the 1980s. Reactants from the gas bubbles dissolve in the liquid and diffuse to the catalyst surface, where they react. Products then diffuse through the liquid back to the gas phase. Heat is removed by generating steam in an internal tubular heat exchanger.

Conversion per pass depends on reaction conditions, catalyst, solvent and space velocity. Experimental results show 15 – 40 % conversion for CO rich gases and 40 – 70 % CO for balanced and H_2 rich gases.

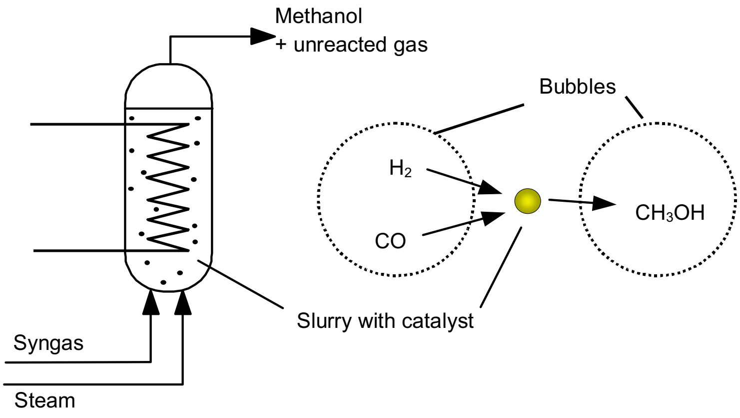

Figure 4. LPMEOH reactor with detail of reaction [28].

Computation models predict future CO conversions of over 90 %, up to 97 % respectively [26]. Researchers at the Brookhaven National Laboratory have developed a low temperature (active as low as 100 °C) catalyst that can convert 90 % of the CO in one pass [1]. With steam addition the reaction mixture becomes balanced through the water gas shift reaction, so that the initial hydrogen to carbon monoxide ratio is allowed to vary from 0.4 to 5.6 without a negative effect on performance [28].

Investment costs for the LPMEOH process are expected to be 5 – 23 % less than for a gas phase process of the same MeOH capacity. Methanol from a 420 MW electricity and 450 – 770 tpd methanol co-producing plant may cost under 0.50 US$/gallon. Methanol from an all methanol plant is expected to cost about 0.60 – 0.70 US$/gallon [28]. This compares with new methanol plants which produce methanol at 0.55 – 0.60 US$/gallon [28].

2.3 H_2 production

Pressure swing adsorption. After reforming and shifting to a hydrogen rich synthesis gas, hydrogen can be separated and compressed. Different process concepts are used in hydrogen plants in operation today. In the conventional design, CO_2 was removed and traces of CO and CO_2 were converted to easily removable methane to give hydrogen with 98 % purity. This process is no longer dominating, but many plants using this concept are still operating. New hydrogen plants are almost invariably designed using Pressure Swing Adsorption (PSA) for final hydrogen purification. The quality of the hydrogen produced is a major issue for its eventual automotive application. Specifically, CO is a strong poison to polymer electrolyte membrane (PEM) fuel cells. Studies indicate that levels as low as 1-2 pp, will deactivate the platinum catalyst of such fuel cells [1]. PSA is based on the difference in adsorption behaviour between different molecules [1; 29], it separates components of a gas stream by selective adsorption to a solid at high pressure, and subsequent desorption at low pressure. This adsorption/desorption is in fact a batch process, but by placing two beds in parallel it operates nearly continuous. While adsorption takes place in one bed, the other is desorbed [30].

PSA (see Figure 5) has been thoroughly described by Katofsky [1]. First activated carbon in the set of beds ‘A’ selectively adsorbs nearly all CO_2 and all H_2O. The remaining gas then passes to the second set of beds ‘B’ containing a zeolite molecular sieve, which selectively adsorbs essentially all the remaining compounds

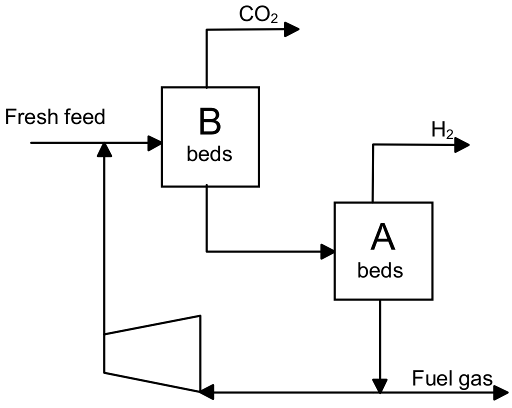

Figure 5. PSA for hydrogen purification [1].

and some hydrogen. The overall recovery of hydrogen is increased by recycling some of the desorbed gas from the 'B' beds. There is a trade-off in that the recycled gas must be recompressed and cooled to near ambient temperature, adding to capital and operating costs, and also the size of the PSA unit will be slightly larger. As with the methanol synthesis loop, some of the recycled gas must be purged to prevent the build-up of methane and other non-hydrogen gases. Recovery rates of 90% and up are achievable; the product purity is extremely high: 99.999%.

Ceramic membranes. Membranes are a promising technology for gas separation. They are attractive because of their simple design and may have the ability of combining shift and separation in one reactor. Membranes for e.g. nitrogen separation are already applied at several small size facilities, where they have better economics than traditional separation technologies [1]. Membranes for hydrogen are evaluated as being an advanced option.

Membrane separation of gas mixtures is based on the difference in mobility of compounds through a surface. The driving force for transport of a component through the membrane is a difference in partial gas density, of this component on the two sides of the membrane. The membranes themselves affect the rates at which different gas molecules are transported through the membrane, depending on the physical and chemical interaction of the gases with the membrane.

Much R&D effort is put in decreasing the pore size to the size of molecules so that membranes can be molecular sieves, allowing only one component through (the smallest). These membranes with small pore sizes are expected to perform better at high temperature [31], when surface flow is eliminated. This implies an important energy advantage when ceramic membranes are combined with hot gas cleaning, because than between gasification and gas turbine, no temperature drop would have to occur. Furthermore the membrane surface catalyses the water gas shift reaction, this reaction is than driven to hydrogen as it is removed by the membrane permeable to H_2 but not to other gases. The shift reaction is demonstrated in the Hydrogen Separation Device (HSD) made by Oak Ridge National Laboratory [32]. Since most of the information is confidential, it is not clear whether the catalytic activity stems from a catalyst condensed on the membrane surface or from the surface itself. If the former is the case, than sulphur removal upstream as not to poison this catalyst may be necessary [17]. The energy of the entering gas is shifted to hydrogen of which eventually 95 % is separated at a purity of 99.5 %.

Ceramic membranes have the advantage of a broad temperature and pressure operating range. Construction of membrane separation devices is potentially very simple and cheap when compared with other separation technologies such as pressure swing adsorption. Moreover, membranes do not suffer the efficiency losses and high capital costs for heat exchangers, associated with the need to cool the synthesis gas [17].

2.4 Electricity co-production

Gas turbines. Unconverted fuel gasses that remain after the methanol or hydrogen production section can still contain a significant amount of chemical energy. These gas streams may be combusted in a gas turbine, although they generally have a much lower heating value (4 – 10 MJ/Nm^3) than natural gas or distillate fuel

(35 – 40 MJ/Nm^3) for which most gas turbine combustors have been designed. When considering commercially available gas turbines for low calorific gas firing, the following items deserve special attention [12; 33; 34]: The combustion stability, the pressure loss through the fuel injection system, and the limits to the increasing mass flow through the turbine.

Different industrial and aeroderivative gas turbines have been operated successfully with low LHV gas, but on the condition that the hydrogen concentration in the gas is high enough to stabilise the flame. Up to 20 % H_2 is required at 2.9 MJ/Nm^3. Hydrogen has a high flame propagation speed and thus decreases the risk of extinguishing the flame [33].

Injecting a larger fuel volume into the combustor through a nozzle originally designed for a fuel with much higher energy density can lead to pressure losses, and thus to a decreased overall cycle efficiency. Minor modifications are sufficient for most existing turbines. In the longer term, new turbines optimised for low heating value gas might include a complete nozzle combustor re-design [33].

The larger fuel flow rate also implies an increase in mass flow through the turbine expander, relative to natural gas firing. This can be accommodated partly by increasing the turbine inlet pressure, but this is limited by the compressor power available. At a certain moment the compressor cannot match this increased pressure any more and goes into stall: the compressor blocks. To prevent stall, decreasing the combustion temperature is necessary; this is called derating. This will lower the efficiency of the turbine, though [12; 33]. Higher turbine capacity would normally give a higher efficiency, but as the derating penalty is also stronger the efficiency gain is small [34].

Due to the set-up of the engine the compressor delivers a specific amount of air. However, to burn one Nm^3 of fuel gas less compressed air is needed compared to firing natural gas. The surplus air can be bled from the compressor at different pressures and used elsewhere in the plant, e.g. for oxygen production [12]. If not, efficiency losses occur.

All the possible problems mentioned for the currently available GTs, can be overcome when designing future GTs. Ongoing developments in gas turbine technology increase efficiency and lower the costs per installed kW over time [12]. Cooled interstages at the compressor will lower compressor work and produce heat, which can be used elsewhere in the system. Also gas turbine and steam turbine could be put on one axis, which saves out one generator and gives a somewhat higher efficiency. And application of large scales will give increased turbine efficiency. The short term restraints and long term possibilities of turbine efficiency are both dealt with.

Turbines set limits to the gas quality. The gas cleaning system needs to match particles and alkali requirements of the GTs. When these standards are exceeded wearing becomes more severe and lifetime and efficiency will drop [12]. However, the fuel gas that passed various catalysts prior to the gas turbine has to meet stricter demands. Contaminants are therefore not a real problem in the gas turbine.

Heat integration. As was pointed out in Figure 1 heat is supplied or needed at several points in the biofuel production process. It is of great importance for the process efficiency that supply and demand are carefully matched, so that more high quality heat is left to raise and superheat high-pressure steam for electricity production in a steam turbine.

There usually is a supply of heat after the gasifier and reformer, where the gas streams are cooled prior to gas cleaning or compression. Furthermore, heat is recovered from flue gas from the gas turbine or boiler. There generally is a heat demand for the gas stream entering the reformer, and a steam demand for drying, for the gasifier, the reformer and the shift reactor.

3 Selected systems

Following the train of components of Figure 1 and given the potential options for gasification, gas cleaning and conditioning, fuel synthesis and separation, many routes to produce methanol or hydrogen from biomass can be imagined. As was explained in Chapter two, the IGT direct oxygen fired pressurised gasifier, in the normal and maximised H_2 option, and the Battelle indirect atmospheric gasifier are considered for synthesis gas production, because they deliver a medium calorific nitrogen undiluted gas stream and cover a broad range of gas compositions.

Some concepts chosen resemble conventional production of fuels from natural gas, making use of wet gas cleaning, steam reforming, shift, and either solid bed methanol reactor or hydrogen PSA. Similar concepts have previously been analysed by Katofsky [1]. Advanced components could offer direct or indirect energy benefits (liquid phase methanol synthesis, hot gas cleaning), or economic benefits (ceramic membranes, autothermal reforming). Available process units are logically combined so the supplied gas composition of a unit matches the demands of the subsequent unit, and heat leaps are restricted if possible. The following considerations play a role in selecting concepts:

- Hot gas cleaning is only sensible if followed by *hot* process units like reforming or (intermediate temperature) shifting. Hot gas cleaning is not applied after atmospheric gasification since the subsequent pressurisation of the syngas necessitates cooling anyway.
- For reforming fuel gas produced via an IGT gasifier before methanol synthesis, an autothermal reformer is chosen, because of the higher efficiency, and lower costs. The high hydrogen yield, possible with steam reforming is less important here since the H_2:CO ratio of the gas is already high. The BCL gasifier, however, is followed by steam reforming to yield more hydrogen. For hydrogen production the IGT gas is not reformed, due to the low hydrocarbon content.
- Preceding liquid phase methanol synthesis, shifting the synthesis gas composition is not necessary since the reaction is flexible towards the gas composition. When steam is added, a shift reaction takes place in the reactor itself. Before gas phase methanol production the composition is partially shifted and because the reactor is sensible to CO_2 excess, part of the CO_2 is removed.
- For hydrogen production, the gas is fully shifted to maximise the H_2 yield. Ceramic membranes, however, do not need a preceding shift because the membrane surface is expected to have shifting capabilities.
- After the methanol once through options, the gas still contains a large part of the energy and is expected to suit gas turbine specifications. The same holds for unreformed BCL and IGT gases, which contain energy in the form of C_2+ fractions. When the heating value of the gas stream does

Table 1. Selected methanol and hydrogen production concepts.

Methanol						
	Gasifier	Gas cleaning	Reforming	Shift	MeOH	Power generation
1	IGT – max H_2	Wet	-	-	Liquid phase	Combined cycle
2	IGT	Hot (550 °C)	ATR	-	Liquid phase, with steam addition	Combined cycle
3	IGT	Wet	-	-	Liquid phase, with steam addition	Combined cycle
4	BCL	Wet	SMR	-	Liquid phase, with steam addition and recycle	Steam cycle
5	IGT	Hot (550 °C)	ATR	Partial	Conventional solid bed, with recycle	Steam cycle
6	BCL	Wet	SMR	Partial	Conventional solid bed, with recycle	Steam cycle
Hydrogen						
	Gasifier	Gas cleaning	Reforming	Shift	H_2 separation	Power generation
1	IGT	Hot (350 °C)	-	Dual	PSA	Combined cycle
2	IGT – max H_2	Hot (800 °C)	-	-	Ceramic membrane + internal shift	Purge gas expansion
3	IGT	Hot (350 °C)	-	-	Ceramic membrane + internal shift	Combined cycle
4	BCL	Wet	SMR	Dual	PSA	Steam cycle
5	BCL	Wet	-	Dual	PSA	Combined cycle

not allow stable combustion in a gas turbine, it is fired in a boiler to raise process steam. The chemical energy of IGT+ gas is entirely in hydrogen and carbon monoxide. After once through methanol production the gas still contains enough chemical energy for combustion in a gas turbine. After a ceramic membrane though, this energy has fully shifted to the produced and separated hydrogen; only expansion is applied to liberate the physical energy.

- Heat supply and demand within plants are to be matched to optimise the overall plant efficiency.

These considerations lead to a selection of 11 conversion concepts. The eleven concepts selected potentially have low cost and/or high energy efficiency. The concepts are composed making use of both existing commercially available technologies, as well as (promising) new technologies.

4 System calculations

4.1 Modelling

The selected systems are modelled in Aspen+, a widely used process simulation program. In this flowsheeting program, chemical reactors, pumps, turbines, heat exchanging apparatus, etc are virtually connected by pipes. Every component can be specified in detail: reactions taking place, efficiencies, dimensions of heating surfaces and so on. For given inputs, product streams can be calculated, or one can evaluate the influence of apparatus adjustments on electrical output. The plant efficiency can be optimised by integrating the heat supply and demand. The resulting dimensions of streams and units and the energy balances can subsequently be used for economic analyses.

The pre-treatment and gasification sections are not modelled, their energy use and conversion efficiencies are included in the energy balances, though.The models start with the synthesis gas composition from the gasifiers as given in Table 2. Only the base scale of 80 dry tonne/hour (430 MW_{th}) biomass is modelled. Modelling assumptions for the process units are given in Table 3.

Oxygen is used as oxidant for the IGT gasifier and the autothermal reformer. The use of air would enlarge downstream equipment size by a factor 4. Alternatively, oxygen enriched air could be used. This would probably give an optimum between small equipment and low air separation investment costs, but it is not considered in this study.

Gas turbines are modelled both as existing and advanced engines. The performance of the low calorific gas in existing gas turbines is calculated using GT Pro, a simulation program with an extensive database on available engines. Results from these calculations, on efficiency, flow dimensions and duct burning, were translated to Aspen+. On the longer term dedicated turbines for low calorific gas have higher efficiencies [12]. It is assumed that increase of scale can barely further improve these efficiencies.

Table 2. Key characteristics of selected gasifiers.

	IGT 1) bubbling fluidised bed		**IGT max H_2 2) bubbling fluidised bed**		**BCL 3) Indirectly heated fast fluidised bed**	
Initial moisture content (%)	30		30		30	
Dry moisture content (%)	15		15		10	
steam (kg/kg dry feed)	0.3		0.8		0.019	
oxygen (kg/kg dry feed)	0.3		0.38		0	
air (kg/kg dry feed)	0		0		2.06	
Product temperature (°C)	982		920		863	
exit pressure (bar)	34.5		25		1.2	
gas yield (kmol/dry tonne)	82.0		121		45.8	
composition: mole fraction on wet basis (on dry basis)						
H_2O	0.318	(-)	0.48	(-)	0.199	(-)
H_2	0.208	(0.305)	0.24	(0.462)	0.167	(0.208)
CO	0.15	(0.22)	0.115	(0.221)	0.371	(0.463)
CO_2	0.239	(0.35)	0.16	(0.308)	0.089	(0.111)
CH_4	0.0819	(0.12)	0.005	(0.009)	0.126	(0.157)
C_2H_4	0.0031	(0.005)	0		0.042	(0.052)
C_2H_6	0		0		0.006	(0.0074)
O_2	0		0		0	
N_2	0		0		0	
LHV_{wet} syngas (MJ/Nm^3)	6.70		3.90		12.7	
Cold gas efficiency (%)	HHV 82.2 / LHV 78.1		HHV 72.1 / LHV 60.9		HHV 80.5 / LHV 82.5	

1) [35] quoted by [4].

2) [36].

3) Compiled from [37] and [38] by [4].

Table 3. Unit modelling assumptions used in Aspen+ calculations.

General	
Heat exchanger[1,2,3]	$\Delta p/p = 2\%$; Minimum $\Delta T = 15°C$ (gas-liquid) or 30 °C (gas-gas); If $T > 300$ °C then heat losses are 2% of heat transferred.
Centrifugal pump[4]	$\eta = 0.65 - 0.9$; $\eta_{driver} = 1$
Blower[4]	$\Delta p < 0.5$ bar; $\eta_{isentropic} = 0.72$; $\eta_{mech} = 1$
Compressor[2]	$\eta_{polytropic} = 0.80$; $\eta_{mech} = 0.90$
Multistage compressor[5]	$\eta_{Isentropic} = 0.76 – 0.78$ (for 1.0e4 – 1.7e5 m3/hour input volume); $\eta_{mech} = 1$; Compression ratio is same for each stage, maximum is 4, such that outlet temperature does not exceed 250°C; Intercooling to 25 °C – 130°C; last stage no duty
Gas Cleaning[6]	
Quench Scrubber[2,7]	Modelled as Two Outlet Flash drum; $T_{in, gas} = 250 – 240 – 120$ °C (for 34.5 – 24 – 1.2 bar); $T_{in, water} = 25$ °C; T = 200 – 180 – 60 °C preferably (for 34.5 – 24 – 1.2 bar) by adjusting amount of water; Minimally 1 m^3 water per 1000 m^3 gas; Q = 0 W; $\Delta p/p = 3\%$
Hot Gas Cleaning[3,8]	Modelled as Valve:T_{in} = 350 °C / 550 °C / 800 °C, $\Delta p = -5$ bar
Reformer	
Steam Reformer[9]	SMR1 provides heat to SMR2 by combusting flue gas, if this is not sufficient, then part of gasifier product is combusted as well. SMR1: Stoichiometric Reactor; T = 890°C; $\Delta p/p = 2\%$; Air is stoichiometric SMR2: Gibbs free energy minimisation Reactor; T_{in} = 860°C; p_{in} = 15,5 bar; Δp = -0,5 bar; T = 890 °C; T Approach = -10°C; 3.5 mole steam injected per mole carbon
Autothermal Reformer[10]	ATR1 provides heat ATR2 requires; T_{in} = 550°C; adjust ratio ATR1/ATR2 to T_{out} = 1000°C; Overall 2 mole steam injected per mole carbon; some gas streams do not require additional steam for reforming ATR1: Stoichiometric Reactor; T = 1000°C; Δp = -0.5 bar; complete combustion of CH_4, C_2H_4 and C_2H_6 using stoichiometric amount of air ATR2: Gibbs free energy minimisation Reactor; 2% of CH_4 is inert; Ar and N_2 are inert; C_2H_4 and C_2H_6 react completely
Shift	
Partial Shift Reactor[1]	Part of stream splits to SHIFT reactor such that ratio (H2 – CO2) / (CO + CO2) = 2.05 ± 0.02 after downstream Selexol Modelled as Gibbs free energy minimisation Reactor; T_{in} = 330 °C; T approach = +10°C; Q = 0 W; Δp = -0.5 bar Inertia: CH_4, C_2H_4, C_2H_6, Ar, N_2; Steam injected is 3 times CO - H_2O
Dual shift reactor[1]	HT Shift: Gibbs free energy minimisation reactor; T_{in} = 350 °C maximally; T approach = +10°C; Q = 0 W Δp = -0.5 bar; Inertia: CH_4, C_2H_4, C_2H_6, Ar, N_2; Steam injected is 3 times CO – H_2O LT Shift: Gibbs free energy minimisation reactor; T_{in} = 260°C; ΔT = +20°C; Δp = -0.5 bar; Inertia: CH_4, C_2H_4, C_2H_6, Ar, N_2
Chemical Reactors	
Conventional Solid Bed Methanol[1,11]	Modelled as Gibbs free energy minimisation Reactor; p_{in} = 106 bar; Δp = -8 bar; Q = 0 W; T_{in} = 50 °C and 250 °C; Inertia: CH_4, C_2H_4, C_2H_6 MeOH in reactor product = 6 mol% by adjusting T Approach; T = 260 °C ± 2.6 by adjusting cold / hot feed ratio; Recycle to Feed ratio = 5
Liquid Phase Methanol[12]	Modelled as Gibbs free energy minimisation Reactor p_{in} = 90 bar; T_{in} = 240 ± 2.4 °C by adjusting T before compression; Δp = -2 bar; T = 250°C; Inertia: CH_4, C_2H_4, C_2H_6; balanced / H_2-rich syngas (H_2:CO > 2) has 75% conversion in CO; CO-rich syngas (2 > H_2:CO > 1) has 60% conversion in CO by adjusting T Approach Optional Steam Addition H2/CO ratio at reactor outlet is adjusted to 2.05 ± 0.02; Syngas becomes balanced; real CO level = (CO_{in} + $H2_{in}$) / 3 Optional Recycle with recycle to feed ratio = 2 or lower

Table 3 continued.

Purification	
Methanol separator1)	Modelled as Two Outlet Flash Drum; 45 °C >T_{in} > 30 °C; Q = 0 Watt ; -50 bar < Δp < -5 bar Subsequent Separator for 100 % pure MeOH
Selexol1,13)	98% of CO_2 and 100% of H_2O separation; T_{in} = 127 °C; Δp = -0.5 bar; CO_2 released at 1.5 bar
Water separator14)	Modelled as Two Outlet Flash Drum T_{in} = 40 °C; Q = 0 Watt ; Δp as HX or more when desired before PSA
PSA system1)	system operating at 14-28 bar, 40°C; recycling 80% PSA-A: Δp = -0.35 bar; 100% CO_2 + H_2O adsorption; desorption at 1.3 bar PSA-B: Δp = -0.35 bar; adsorption of all gas but 84% of H_2; desorption at 1.3 bar
Ceramic Membrane1,15)	system operating at elevated pressure: 20 bar or higher T_{in} = T_{out} = 800 °C; Catalytic molecular sieve: shift all CO on surface to H_2, therefore H_2O:CO => 1 at entrance, transport 95 % of H_2 and 0% of others to product stream; Product at 1.2 bar; Δp depleted stream = -0.1 bar
Power generation	
Advanced Gas turbine16)	Compressor; p_{fuel} = 50 bar, p_{air} = 40 bar; $\eta_{isentropic}$ = 0.91; η_{mech} = 0.99 Combustor modelled as Stoichiometric reactor; Δp = 0 bar; Q = 0 W; T after turbine expander = 550 ± 2 °C by adjusting Air; Temperature is set by adjusting air to compressor Expander: p = 1.2 bar; $\eta_{isentropic}$ = 0.89; η_{mech} = 0.99; T after heat exchanger = 100°C
Existing Gas turbine	Data on pressures, efficiencies, turbine cooling, etc. from GT PRO
Boiler	2-3 MJ/m^3 is lower limit for normal combustion Stoichiometric Reactor p_{in} = 1.2 bar; Δp = -0.1 bar; T = 1200 °C by superheating steam; O_2 = 5% ± 0.05 by adjusting Air
HRSG17)	Gas T_{out} = 100 °C ; Water T_{in} = 15 °C
Steam Turbine6)	Steam of preferably 86.2 bar, 510 °C is expanded Intermediate steam extraction is possible: p = $p_{gasifier}$ (34.5 bar / 250 °C, 25 bar / 240 °C, 1.2 bar / 120°C); p = 12 bar for drier (200 °C); p = 0.04 bar; $\eta_{isentropic}$ = 0.89; η_{mech} = 0.99
Air composition	O_2 = 0.2075; H_2O = 0.0101; CO_2 = 0.0003; N_2 = 0.7729; Ar = 0.0092; T = 15 °C, p = 1 atm

1) [1].

2) [33].

3) [8].

4) Aspen+ default value.

5) [39]

6) The tar cracker following the atmospheric gasification (BCL) is not modelled. It is assumed to be integrated with the gasifier.

7) [9]

8) Hot gas cleaning modelled as a black box. Operating temperature as defined in Table 1.

9) Steam reformer operates at 1 – 3.5 MPa, with molar steam:carbon ratios in the range 3–5 : 1. Typical reformer temperature is between 830 °C and 1000 °C [1]. The inlet stream is heated by the outlet stream up to 860 °C to match reformer heat demand and supply. The furnace would typically use 10 % excess air for C_1 to C_4 firing, correlating to approximately 1.7 % oxygen in the flue gas, to ensure that the burners do not limit plant throughput, and for safety reasons [40]. The modelled SMR furnace is sized as to exactly match the heat demand, without an excess air.

10) Autothermal reformer operates at 20 – 70 bar, 850 – 1100 °C, steam to carbon ratio ranges from 0.5 to 3.5 [41]. Oxygen is set stoichiometric for oxidation part of ATR.

11) Conventional gas phase methanol reactor modelled as quench type (ICI low pressure methanol process). Typical methanol synthesis temperature is between 230 and 270 °C. Temperature is set 260 °C by adjusting the cold/hot feed ratio. The reactor operates adiabatic. Pressure is typically 50 to 150 bar, pressure drop is 5 to 8 bar. Recycle to feed ratio is typically between 2.3 and 6 [1; 13; 26; 27]. Side reactions to dimethyl-ether, formaldehyde or higher alcohols are not modelled.

12) Liquid phase methanol reactor. Experimental results show 15 – 40 % conversion for CO rich gases and 40 – 70 % CO for balanced and H_2 rich gases, but computation models predict future CO conversions of over 90 %, up to 97 % respectively [26; 28; 42]. Side reactions to dimethyl-ether, formaldehyde or higher alcohols are not modelled.

13) Selexol. Actually, half of the CO_2 is released at 1 bar and half at 4 bar. The net energy demand of a 436 tonne of CO_2 per hour unit amounts 9 MW_e [43].

14) Over 99 % of the water is separated, over 99.99 % of the combustible gasses passes through.

15) Ceramic membranes modelled as hydrogen separation device or HSD (developed by Oak Ridge National Laboratory). Operation at high temperature, surface has shifting capabilities [32; 44-46].

16) [12].

17) HRSG after GT or boiler. The flue gas can be cooled down to 100 °C without corrosion problems, since the gas is expected to contain less than 100 ppb sulphur [12].

The heat supply and demand within the plant is carefully matched, aimed at maximising the production of superheated steam for the steam turbine. A summation of all heat inputs and outputs in a *heat bin* is too simple, since it does not take the quality of heat into account. Pinch analysis, as was also done by Katofsky, gives the ultimate optimisation of energy streams within plants, but also leads to too optimistic ideal outcomes and possibly very large number of heat exchangers. Therefore heat integration of heat demand and supply within the considered plants here is done by hand. The intention is to keep the integration simple by placing few heat exchangers per gas/water/steam stream. Of course, concepts with more process units demanding more temperature altering are more complex than concepts consisting of few units. First, an inventory of heat supply and demand is made. Streams matching in temperature range and heat demand/supply are combined: e.g. heating before the reformer by using the cooling after the reformer. When the heat demand is met, steam can be raised for power generation. Depending on the amount and ratio of high and low heat, process steam is raised in heat exchangers, or drawn from the steam turbine: If there is enough energy in the plant to raise steam of 300 °C, but barely superheating capacity, than process steam of 300 °C is raised directly in the plant. If there is more superheating than steam raising capacity, than process steam is drawn from the steam cycle. Steam for gasification and drying is almost always drawn from the steam cycle, unless a perfect match is possible with a heat-supplying stream. The steam entering the steam turbine is set at 86 bar and 510 °C.

4.2 System calculation results

Table 4 summarises the outcomes of the flowsheet models. The overall energy efficiencies are expressed in different ways. The most direct is the net overall Fuel + Electricity efficiency, but this definition gives a distorted view, since the quality of energy in fuel and electricity is considered equal, while in reality it is not. The Fuel only efficiency assumes that the electricity part could be produced from biomass at 45 % HHV in an advanced BIG/CC [47]. This definition compensates for the inequality of electricity and fuel in the most justified way, but the referenced electric efficiency is of decisive importance. Expressing the performance in primary energy avoided divides the co-generation benefit over fuel and electricity. Another qualification for the performance of the system could use exergy: the amount of work that could be delivered by the material streams.

In some concepts still significant variations can be made. In Methanol concept 4, the reformer needs gas for firing. The reformer can either be entirely fired by purge gas (thus restricting the recycle volume), or by part of the gasifier gas. The first option gives a somewhat higher methanol production and overall plant efficiency. The Hydrogen concept 4 offers a similar choice between reformer combustor feeding directly from the gasifier, or from the purge gas. But in this concept combusting part of the gasifier gas gives the higher efficiency. In Methanol concept 5 one can choose between a larger recycle, and more steam production in the boiler. A recycle of five times the feed volume, instead of four, gives a much higher methanol production and plant efficiency. Per concept only the most efficient variation is reported in Table 4.

Based on experiences with low calorific combustion elsewhere [12; 33] the streams in this study, which were projected to be combusted in a gas turbine, will give stable combustion. Only the performance of the

Table 4. Results of the Aspen+ performance calculations, for 430 MW_{th} input HHV systems (equivalent to 380 MW_{th} LHV for biomass with 30% moisture) of the methanol and hydrogen production concepts considered.

		HHV Output (MW)		**HHV Efficiency (%)**		**Primary Energy Avoided[3] (%)**
		Fuel	**Net electricity[1] (gross – internal)**	**Fuel + E**	**Fuel only[2]**	
Methanol						
1	IGT – max H2, Scrubber, Liquid Phase Methanol Reactor, Combined Cycle	161	53 (71 – 18)	50%	52%	83%
2	IGT, Hot Gas Cleaning, Autothermal Reformer, Liquid Phase Methanol Reactor with Steam addition, Combined Cycle	173	62 (82 – 20)	55%	59%	91%
3	IGT, Scrubber, Liquid Phase Methanol Reactor with Steam addition, Combined Cycle	113	105 (118 – 14)	51%	58%	87%
4	BCL, Scrubber, Steam Reformer, Liquid Phase Methanol Reactor with Steam Addition and Recycle, Steam Cycle	246	0 (25 – 25)	57%	57%	90%
5	IGT, Hot Gas Cleaning, Autothermal Reformer, Partial Shift, Conventional Methanol Reactor with Recycle, Steam Turbine	221	15 (38 – 23)	55%	56%	88%
6	BCL, Scrubber, Steam Reforming, Partial Shift, Conventional Methanol Reactor with Recycle, Steam Turbine	255	-17 (10 – 27)	55%	54%	86%
Hydrogen						
1	IGT, Hot Gas Cleaning, Dual Shift, Pressure Swing Adsorption, Combined Cycle	176	73 (93 – 21)	58%	66%	85%
2	IGT – max H2, High Temperature Dust Filter, Ceramic Membrane (Internal Shift), Expansion Turbine	259	-1 (25 – 26)	60%	60%	79%
3	IGT, Hot Gas Cleaning, Ceramic Membrane (Internal Shift), Combined Cycle	177	84 (103 – 19)	61%	74%	91%
4	BCL, Scrubber, Steam Reformer, Dual Shift, Pressure Swing Adsorption	303	-22 (0 – 22)	65%	63%	83%
5	BCL, Scrubber, Dual Shift, Pressure Swing Adsorption, Combined Cycle	149	72 (97 – 25)	52%	56%	77%

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[1] Net electrical output is gross output minus internal use. Gross electricity is produced by gas turbine and/or steam turbine. The internal electricity use stems from pumps, compressors, oxygen separator, etc.

[2] The electricity part is assumed to be produced from biomass at η_e = 45 % HHV efficiency [47]. The Fuel only efficiency is calculated by $\eta = \text{Fuel}/(MW_{th,in} – \text{Electricity}/\eta_e)$.

[3] The mix fuel + electricity could also be produced from natural gas at η_e = 54 % and η_f = 63 % for methanol or η_f = 76 % for hydrogen [48]. Primary Avoided is calculated by $PA = (\text{Electricity}/\eta_e + \text{Fuel}/\eta_f) /(MW_{th,in})$.

gas turbine in the Hydrogen 3 concept is unsure, having a low calorific value combined with little hydrogen. In GT PRO (Heat balance engineering software from Thermoflow, inc) gas turbines are chosen with dimensions matching the heat flow of the purged gas, and with high combined cycle efficiencies. Gas turbine only efficiencies are 33 – 47 %; the high value is found for pressurised hot gas after the ceramic membrane. Advanced turbine configurations, with set high compressor and turbine efficiencies and no dimension restrictions, give gas turbine efficiencies of 41 – 52 % and 1 – 2 % point higher overall plant efficiency than conventional configurations. Table 4 only includes the advanced turbines.

Based on the overall plant efficiency the methanol concepts lie in a close range: methanol 50 – 57 % and hydrogen 52 – 61 %. Liquid phase methanol production preceded by reforming (concepts 2 and 4) results in somewhat higher overall efficiencies and primary energy avoided. After the pressurised IGT gasifier hot gas cleaning leads to higher efficiencies than wet gas cleaning, although not better than concepts with wet gas cleaning after a BCL gasifier. The conventional hydrogen concept 4 has the highest overall plant efficiency, but depends heavily on the import of electricity to the plant. If compared on a fuel only basis, its performance is the worst of the hydrogen concepts. The most advanced concept Hydrogen 3 than is the most efficient.

Several units may be realised with higher efficiencies than considered here. For example new catalysts and carrier liquids could improve liquid phase methanol single pass efficiency up to 95 % [42]. The electrical efficiency of gas turbines will increase by 2 – 3 % points when going to larger scale [49].

5 Economics

5.1 Method

An economic evaluation has been carried out for the concepts considered. Plant sizes of 80, 400, 1000 and 2000 MW_{th} HHV are evaluated, 400 MW_{th} being the base scale. The scale of the conversion system is expected to be an important factor in the overall economic performance. This issue has been studied for BIG/CC systems [47; 50], showing that the economies of scale of such units can offset the increased costs of biomass transport up to capacities of several hundreds of MW_{th}. The same reasoning holds for the fuel production concepts described here. It should however be realised that production facilities of 1000 – 2000 MW_{th} require very large volumes of feedstock: 200 – 400 dry tonne/hour, or 1.6 – 3.2 dry Mtonne per year. Biomass availability will be a limitation for most locations for such large-scale production facilities, especially in the shorter term. In the longer term (e.g. 2020), if biomass production systems become more commonplace, this can change. Very large scale biomass conversion is not without precedent: various large-scale sugar/ethanol plants in Brazil have a biomass throughput of 1 – 3 Mtonne of sugarcane per year, while the production season covers less than half a year. Also large paper and pulp complexes have comparable capacities. The base scale chosen is comparable to the size order studied by Williams *et al.* [4] and Katofsky [1], 370 – 385 MW_{th}.

The methanol and hydrogen production costs are calculated by dividing the total annual costs of a system by the produced amount of fuel. The total annual costs consist of:

- Annual investments
- Operating and Maintenance
- Biomass feedstock
- Electricity supply / demand (fixed power price)

The total annual investment is calculated by a factored estimation [51], based on knowledge of major items of equipment as found in literature or given by experts. The uncertainty range of such estimates is up to ± 30 %. The installed investment costs for the separate units are added up. The unit investments depend on the size of the components (which follow from the Aspen+ modelling), by scaling from known scales in literature (see Table 5), using Equation 1:

$$Cost_a / Cost_b = (Size_a / Size_b)^R \qquad \text{Equation 1}$$

with R = Scaling factor

Various system components have a maximum size, above which multiple units will be placed in parallel. Hence the influence of economies of scale on the total system costs decreases. This aspect is dealt with by assuming that the base investment costs of multiple units are proportional to the cost of the maximum size: the base investment cost per size becomes constant. The maximum size of the IGT gasifier is subject to discussion, as the pressurised gasifier would logically have a larger potential throughput than the atmospheric BCL.

The total investment costs include auxiliary equipment and installation labour, engineering and contingencies. If only equipment costs, excluding installation, are available, those costs are increased by applying an overall installation factor of 1.86. This value is based on 33% added investment to hardware costs (instrumentation and control 5%, buildings 1.5% grid connections 5%, site preparation 0.5%, civil works 10%, electronics 7%, and piping 4%) and 40 % added installation costs to investment (engineering 5%, building interest 10%, project contingency 10%, fees/overheads/profits 10%, start-up costs 5%) [47].

The annual investment cost follows from Equation 2, which takes into account the residual (salvage) value at the end of the economic lifetime of the installation. This residual value follows from the constant annual depreciation over the technical lifetime [51; 52].

$$I_{annual} = \frac{IR}{1-\frac{1}{(1+IR)^{t_e}}} \times I_t \cdot \left(1 - \frac{1}{(1+IR)^{t_e}} \cdot \frac{t_t - t_e}{t_t}\right) \qquad \text{Equation 2}$$

with I_{annual} = Annual investment costs

IR = Interest rate = 10 %

I_t = Total investment (sum of unit investments)

t_e = Economical lifetime = 15 years

t_t = Technical lifetime = 25 years

Operational costs (maintenance, labour, consumables, residual streams disposal) are taken as a single overall percentage (4 %) of the total installed investment [47; 53]. Differences between conversion concepts are not anticipated.

It is assumed in this study that enough biomass will be available at 2 US$/GJ (HHV), this is a reasonable price for Latin and North American conditions. Costs of cultivated energy crops in the Netherlands amount approximately 4 US$/GJ and thinnings 3 US$/GJ [54], and biomass imported from Sweden on a large scale is expected to cost 7 US$/GJ [55]. On the other hand biomass grown on Brazilian plantations could be delivered to local conversion facilities at 1.6 – 1.7 US$/GJ [4; 56].

Electricity supplied to or demanded from the grid yields or costs 0.03 US$/kWh. The annual load is 8000 hours.

Table 5. Costs of system components used for cost analyses of considered concepts, in MUS$\$_{2001}$[1].

Unit	Base Investment Cost (fob)	Scale Factor	Overall installation factor [24]	Base Scale	Maximum Size [25]
Pre-treatment [2]					
Conveyers [3]	0.35	0.8	1.33 (v) * 1.4	33.5 wet tonne/hour	110
grinding [3]	0.41	0.6	1.33 (v) * 1.4	33.5 wet tonne/hour	110
storage [3]	1.0	0.65	1.33 (v) * 1.4	33.5 wet tonne/hour	110
dryer [3]	7.6	0.8	1.33 (v) * 1.4	33.5 wet tonne/hour	110
iron removal [3]	0.37	0.7	1.33 (v) * 1.4	33.5 wet tonne/hour	110
feeding system [3,4]	0.41	1	1.33 (v) * 1.4	33.5 wet tonne/hour	110
Gasification System					
BCL [5]	16.3	0.65	1.69	68.8 dry tonne/hour	83
IGT [6]	38.1	0.7	1.69	68.8 dry tonne/hour	75
Oxygen Plant (installed) [7]	44.2	0.85	1	41.7 tonne O_2/hour	-
Gas Cleaning					
Tar Cracker [3]	3.1	0.7	1.33 (v) * 1.4	34.2 m^3 gas/s	52
Cyclones [3]	2.6	0.7	1.33 (v) * 1.4	34.2 m^3 gas/s	180
High-temperature heat exchanger [8]	6.99	0.6	1.31 (v) * 1.4	39.2 kg steam/s	-
Baghouse filter [3]	1.6	0.65	1.33 (v) * 1.4	12.1 m^3 gas/s	64
Condensing Scrubber [3]	2.6	0.7	1.33 (v) * 1.4	12.1 m^3 gas/s	64
Hot Gas Cleaning [9]	30	1.0	1.23 (v) * 1.4	74.1 m^3 gas/s	-
Syngas Processing					
Compressor [10]	11.1	0.85	1.23 (v) * 1.4	13.2 MW_e	-
Steam Reformer [11]	9.4	0.6	1.64 (v) * 1.4	1390 kmol total/hour	-
Autothermal Reformer [12]	4.7	0.6	1.64 (v) * 1.4	1390 kmol total/hour	-
Shift Reactor (installed) [13]	36.9	0.85	1	15.6 Mmol CO+H_2/hour	-
Selexol CO_2 removal (installed) [14]	54.1	0.7	1	9909 kmol CO_2/hour	-
Methanol Production					
Gas Phase Methanol [15]	7	0.6	1.5 (v) * 1.4	87.5 tonne MeOH/hour	-
Liquid Phase Methanol [16]	3.5	0.72	1.5 (v) * 1.4	87.5 tonne MeOH/hour	-
Refining [17]	15.1	0.7	1.5 (v) * 1.4	87.5 tonne MeOH/hour	
Hydrogen Production					
PSA units A+B [18]	28.0	0.7	1.69	9600 kmol feed/hour	-
Ceramic Membrane (installed) [19]	21.6	0.8	1	17 tonne H_2/hr	-
Power Isle [20]					
Gas Turbine + HRSG [3,21]	18.9	0.7	1.33 (v) * 1.4	26.3 MW_e	-
Steam Turbine + steam system [3,22]	5.1	0.7	1.33 (v) * 1.4	10.3 MW_e	-
Expansion Turbine [23]	4.3	0.7	1.33 (v) * 1.4	10.3 MW_e	-

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[1] Annual GDP deflation up to 1994 is determined from OECD [57] numbers. Average annual GDP deflation after 1994 is assumed to be 2.5 % for the US, 3.0 % for the EU. Cost numbers of Dutch origin are assumed to be dependent on the EU market, therefore EU GDP deflators are used. 1 €$_{2001}$ = 0.94 US$\$_{2001}$ = 2.204 Dfl$_{2001}$.

[2] Total pre-treatment approximately sums up to a base cost of 8.15 MUS$\$_{2001}$ at a base scale of 33.5 tonne wet/hour with an R factor of 0.79.

[3] Based on first generation BIG/CC installations. Faaij *et al.* [58] evaluated a 29 MW_e BIG/CC installation (input 9.30 kg dry wood/s, produces 10.55 Nm^3 fuel gas/s) using vendor quotes. When a range is given, the higher values are used [47]. The scale factors stem from Faaij *et al.* [47].

[4] Two double screw feeders with rotary valves [58].

[5] 12.72 MUS$\$_{1991}$ (already includes added investment to hardware) for a 1650 dry tonne per day input BCL gasifier, feeding not included, R is 0.7 [4]. Stronger effects of scale for atmospheric gasifiers (0.6) were suggested by Faaij *et al.* [47]. Technical director Mr. Paisley of Battelle Columbus, quoted by Tijmensen [8] estimates the maximum capacity of a single BCL gasifier train at 2000 dry tonnes/day.

[6] 29.74 MUS$\$_{1991}$ (includes already added investment to hardware) for a 1650 dry tonne/day input IGT gasifier, R = 0.7 [4]. Maximum input is 400 MW_{th} HHV [8].

[7] Air Separation Unit: Plant investment costs are given by Van Dijk [13]: $I = 0.1069 \cdot C^{0.8508}$ in MUS$\$_{1995}$ installed, C = Capacity in tonne O_2/day. The relation is valid for 100 to 2000 tonne O_2/day. Williams *et al.* [4] assume higher costs for small installations, but with a stronger effect of scale: $I = 0.260 \cdot C^{0.712}$ in MUS$\$_{1991}$ fob plus an overall installation factor of 1.75 (25 and 40%). Larson *et al.* [53] assume lower costs than Van Dijk, but with an even stronger scaling factor than Williams: 27 MUS$\$_{1997}$ installed for an 1100 tonne O_2 per day plant and R=0.6. The first formula (by Van Dijk) is used in the present study. The production of 99.5% pure O_2 using an air separation unit requires 250 – 350 kWh per tonne O_2 [13; 59].

[8] High temperature heat exchangers following the gasifier and (in some concepts) at other locations are modelled as HRSG's, raising steam of 90 bar/520 °C. A 39.2 kg steam/s unit costs 6.33 MUS$\$_{1997}$ fob, overall installation factor is 1.84 [53].

9) Tijmensen [8] assumes the fob price for Hot Gas Cleaning equipment to be 30 MUS$$_{2000}$ for a 400 MW$_{th}$ HHV input. This equals 74.1 m^3/s from a BCL gasifier (T=863°C, 1.2 bar). There is no effect of scaling.

10) Katofsky [1] assumes compressors to cost 700 US$$_{1993}$ per required kW$_{mech}$, with an installation factor of 2.1. The relation used in the present study stems from the compressor manufacturer Sulzer quoted by [8]. At the indicated base-scale, total installed costs are about 15 % higher than assumed by Katofsky. Multiple compressors, for fuel gas, recycle streams, or hydrogen, are considered as separate units. Overall installation factor is taken 1.72 because the base unit matches a 400 MW$_{th}$ plant rather than a 70 MW$_{th}$ plant.

11) Investments for steam reformer vary from 16.9 MUS$$_{1993}$, for a throughput of 5800 kmol methane/hour with an overall installation factor of 2.1 [1] to 7867 k$$_{1995}$ for a 6.2 kg methane/s (1390 kmol/hour), overall installation factor is 2.3 [13]. These values suggest a strong effect of scaling R=0.51, while Katofsky uses a modest R=0.7. In the present study the values of Van Dijk are used in combination with an R factor of 0.6. The total amount of moles determines the volume and thus the price of the reactor.

12) Autothermal Reforming could be 50 % cheaper than steam reforming [1], although higher costs are found as well [23].

13) Investment for shift reactors vary from 9.02 MU$$_{1995}$ for an 8819 kmol CO+H$_2$/hr reactor, and an overall installation factor is 1.81 [4] to 30 MUS$$_{1994}$ installed for a 350000 Nm^3/hr CO+H$_2$/hr (15625 kmol/hr) reactor [43]. Williams assumes an R=0.65, but comparison of the values suggest only a weak influence of scale (R=0.94), in the present study the values from Hendriks are used, R is set 0.85. A dual shift is costed as a shift of twice the capacity.

14) Costs for CO$_2$ removal through Selexol amounts 14.3 MUS$$_{1993}$ fob (overall installation factor is 1.87) for an 810 kmol CO$_2$/hr unit, R = 0.7 [1] up to 44 MUS$$_{1994}$ installed for a 9909 kmol CO$_2$/hour unit [43]. The value from Hendriks is assumed to be right, since his research into CO$_2$ removal is comprehensive.

15) Van Dijk *et al*. [13] estimate that a Methanol Reactor for a 2.1 ktonne methanol per day plant costs 4433 kUS$$_{1995}$ (fob) or 9526 kUS$$_{1995}$ installed (overall installation factor is 2.1). The total plant investment in their study is 138 MUS$$_{1995}$, or 150 MUS$$_{2001}$. Katofsky [1] estimates the costs for a 1056 tonne methanol/day plant to be 50 MUS$$_{1995}$ fob, this excludes the generation and altering of syngas, but includes make-up and recycle compression and refining tower. Correspondence with mr. Van Ooijen [60] of Akzo Nobel and mr. De Lathouder [61] of DSM Stamicarbon revealed that a 1000 tpd plant costs about 160 MUS$$_{2001}$, and a 2000 tpd plant 200 MUS$$_{2001}$ (this suggests a total plant scale factor of 0.3). These values come near the ones mentioned by Katofsky. This implies that the values given by Van Dijk are too optimistic and should be altered by a factor 1.33. It is therefore assumed that the base investment for the methanol reactor only is 7 MUS$$_{2001}$, the installation factor is 2.1.

16) Installed costs for a 456 tonne per day Liquid Phase Methanol unit, are 29 MU$$_{1997}$, excluding generation and altering of syngas, but including make-up and recycle compression, and refining tower. R = 0.72 [62]. Corrected for scale and inflation this value is about half the cost of the conventional unit by Katofsky and the corrected costs of Van Dijk. It is therefore assumed that the price of a Liquid Phase Methanol reactor is 3.5 MUS$$_{2001}$ for a 2.1 ktonne per day plant, installation factor is 2.1.

17) Cost number for methanol separation and refining is taken from Van Dijk, increased with 33 % as described in note 15.

18) PSA units (excluding the recycle compressor) cost 23 MUS$$_{1993}$ for a 9600 kmol feed/hour throughput, R= 0.7 [1].

19) Membrane costs 68 US$$_{1997}$/(kW/bar), but these costs are only 9 % of the total installed cost for a Hydrogen Separation Device. Investment costs stem from Parsons I&TG [32]. The economies of scale of the membrane surface are low because the required surface area is proportional to the throughput, this slightly influences the overall R factor of the complete HSD.

20) For indication: A complete Combined Cycle amounts about 830 US$$_{1997}$ per installed kW$_e$. Quoted from [63] by [23].

21) Scaled on Gas Turbine size.

22) Steam system consists of water and steam system, steam turbine, condenser and cooling. Scaled on Steam Turbine size.

23) Expansion turbine costs are assumed to be the same as steam turbine costs (without steam system).

24) Overall installation factor. Includes auxiliary equipment and installation labour, engineering and contingencies. Unless other values are given by literature, the overall installation factor is set 1.86 for a 70 MW$_{th}$ scale [47]. This value is based on 33% added investment to hardware costs (instrumentation and control 5%, buildings 1.5% grid connections 5%, site preparation 0.5%, civil works 10%, electronics 7%, and piping 4%) and 40 % added installation costs to investment (engineering 5%, building interest 10%, project contingency 10%, fees/overheads/profits 10%, start-up costs 5%). For some components, the added investments to hardware decrease slightly with larger scale (indicated by *v*), R = 0.8.

25) Maximum sizes from Tijmensen [8].

5.2 Results

Results of the economic analysis are given in Table 6 and Figure 6. The 400 MW_{th} conversion facilities deliver methanol at 8.6 - 12 US$/GJ, the hydrogen cost range is 7.7 – 11 US$/GJ. Considering the 30 % uncertainty range one should be careful in ranking the concepts. Some concepts (Methanol 4 and 6 and Hydrogen 2, 3 and 4) perform somewhat better than the other concepts due to an advantageous combination of lower investment costs and higher efficiency. The lowest methanol production price is found for concepts using the BCL gasifier, having lower investment costs. In hydrogen production the ceramic membrane concepts perform well due to their higher overall efficiency and modest investments. The combination of an expensive oxygen fired IGT gasifier with an combined cycle is generally unfavourable, since the efficiency gain is small compared to the high investment. Only in Hydrogen 3 (using a ceramic membrane) the extra investment seems justified.

Investment redemption accounts for 42 – 76 % of the annual costs and is influenced by the unit investment costs, the interest rate and the plant scale. The build-up of the total investment for all concepts is depicted in Figure 7. It can be seen that the costs for the gasification system (including oxygen production), syngas processing and power generation generally make up the larger part of the investment. For autothermal reforming higher investment costs [23], would increase the methanol price from considered concepts by about 1.5 US$/GJ. Uncertainties in the investment costs for ceramic membranes, however do not have a large influence. Developments in gasification and reforming technology are important to decrease the investments. On the longer term capital costs may reduce due to technological learning: a combination of lower specific component costs and overall learning. A third plant build may be 15 % cheaper leading to a 8

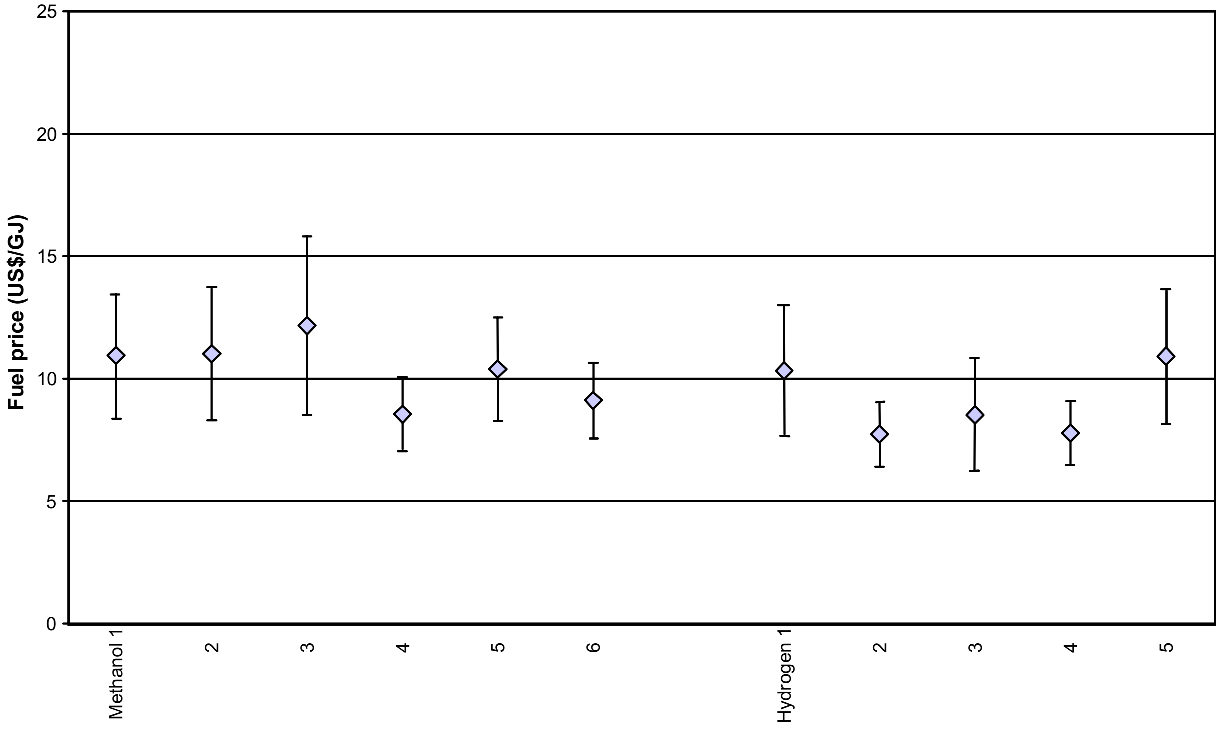

Figure 6. Fuel price for 400 MW_{th} input concepts, with 30 % uncertainty on investment costs.

– 15 % fuel cost reduction.

The interest rate has a large influence on the fuel production costs. At a rate of 5 % methanol production costs decrease with about 20 % to 7.2 – 9.0 US$/GJ, hydrogen to 6.6 – 8.5 US$/GJ. At a high interest rate (15%) methanol production costs become 9.9 – 14 US$/GJ, hydrogen 8.9 – 14 US$/GJ. The last rows of Table 6 show potential fuel production costs in smaller or bigger installations. Going to 1000 and 2000 MW_{th} scales the fuel production costs reach cost levels as low as 7.1 – 9.5 US$/GJ for methanol and 7.0 – 8.4 US$ /GJ for hydrogen.

Feedstock costs account for 36 – 62 % of the final fuel costs for the mentioned technologies. If a biomass price of 1.7 US$/GJ could be realised (a realistic price for e.g. Brazil), methanol production costs would become 8.0 – 11 US$/GJ, and hydrogen production costs 7.2 – 10 US$/GJ for 400 MW_{th} concepts. On the other hand, when biomass costs are 3 US$/GJ (short term Western Europe) the cost of produced methanol will increase to 10 – 16 US$/GJ, and hydrogen to 9.4 – 14 US$/GJ.

If the electricity can be sold as green power, including a *carbon neutral* premium, the fuel production costs for power co-producing concepts drops, where the green premium essentially pays a large part of the fuel production costs. A power price of 0.08 US$/GJ would decrease methanol costs to -0.6 – 9.5 US$/GJ and hydrogen costs to 1.9 – 4.6 US$/GJ. Of course the decrease is the strongest for concepts producing more electricity. A green electricity scenario, however, may be a typical western European scenario. As such it is unlikely that it can be realised concurrent with biomass available at 1.7 US$/GJ.

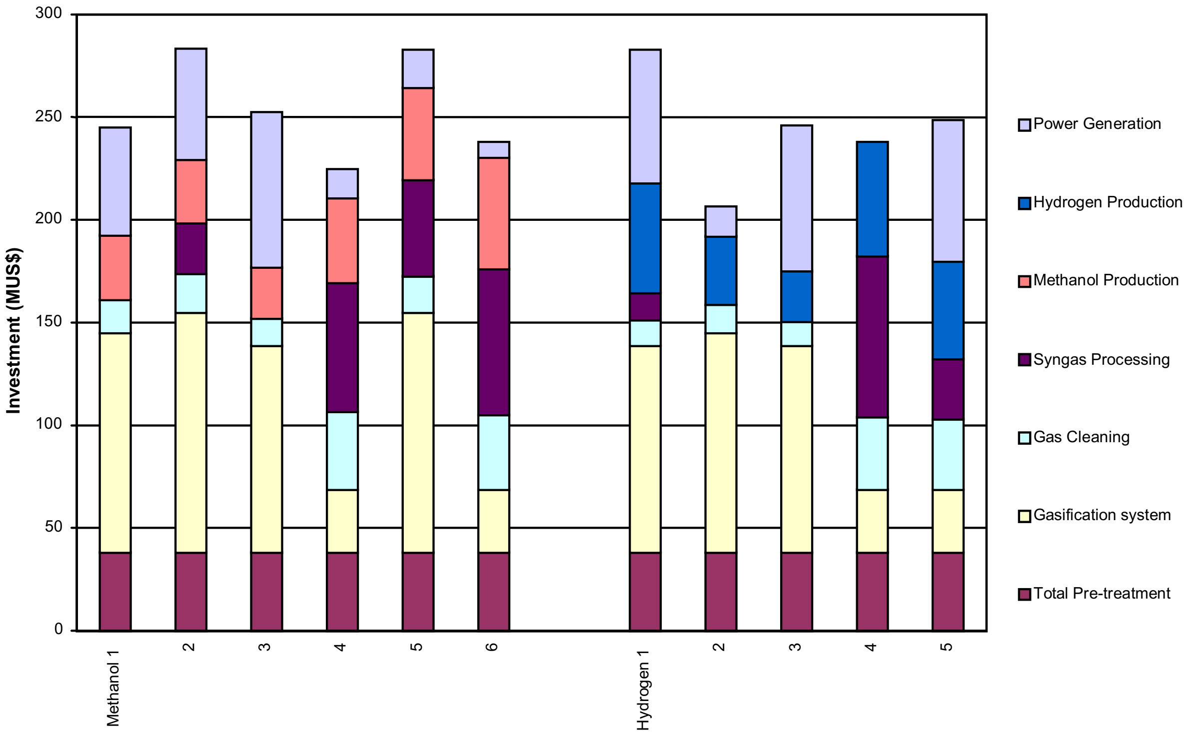

Figure 7. Breakdown of investment costs for 400 MW_{th} concepts.

Table 6. Economic analyses for the concepts considered. Costs in US$$_{2001}$.

			Methanol						**Hydrogen**				
	Unit		**1**	**2**	**3**	**4**	**5**	**6**	**1**	**2**	**3**	**4**	**5**
	Total Pre-treatment	MUS$	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2
Gasification system	BCL	·	0	0	0	30.4	0	30.4	0	0	0	30.4	30.4
	IGT	·	73.0	73.0	73.0	0	73.0	0	73.0	73.0	73.0	0	0
	Oxygen	·	33.8	43.8	27.7	0	43.7	0	27.7	33.8	27.7	0	0
Gas Cleaning	Tar cracker	·	0	0	0	9.2	0	9.2	0	0	0	9.2	9.2
	Cyclones	·	1.8	1.2	1.2	6.8	1.2	6.8	1.2	1.8	1.2	6.8	6.8
	HT Heat Exchanger (total installed)	·	10.9	14.6	9.6	11.2	13.6	10.3	8.9	4.4	7.4	9.0	6.9
	Baghouse Filter	·	1.4	0	0.9	4.1	0	3.8	0	0	0	3.8	4.3
	Condensing Scrubber	·	2.1	0	1.3	6.8	0	6.4	0	0	0	6.4	7.2
	Hot Gas Cleaning	·	0	3.0	0	0	3.0	0	2.3	7.5	3.0	0	0
Syngas Processing	Compressor	·	0	0	0	16.9	0	16.5	0	0	0	14.9	17.9
	Steam Reformer	·	0	0	0	45.9	0	43.3	0	0	0	42.7	0
	Catalytic Autothermal Reformer	·	0	24.5	0	0	24.5	0	0	0	0	0	0
	Shift Reactor(s)	·	0	0	0	0	5.0	1.9	13.3	0	0	21.0	11.5
	Selexol CO_2 removal	·	0	0	0	0	17.4	9.5	0	0	0	0	0
Methanol Production	Make Up Compressor	·	13.3	12.4	10.5	17.4	11.4	17.5	0	0	0	0	0
	Gas Phase Methanol	·	0	0	0	0	9.1	9.8	0	0	0	0	0
	Liquid Phase Methanol	·	3.3	3.5	2.6	4.4	0	0	0	0	0	0	0
	Recycle Compressor	·	0	0	0	0.4	6.5	7.2	0	0	0	0	0
	Refining	·	14.7	15.3	11.8	19.1	17.8	19.5	0	0	0	0	0
Hydrogen Production	PSA Units A + B	·	0	0	0	0	0	0	31.7	0	0	35.1	26.9
	HT Ceramic Membrane	·	0	0	0	0	0	0	0	10.1	7.5	0	0
	Recycle Compressor	·	0	0	0	0	0	0	15.3	0	0	6.2	13.1
	Product Compressor	·	0	0	0	0	0	0	6.3	23.2	17.1	14.1	7.4
Power Generation	Gas turbine + HRSG	·	35.4	31.5	54.5	0	0	0	46.6	0	54.3	0	54.9
	Steam Turbine + steam system	·	17.1	22.4	21.1	13.9	18.3	7.6	18.2	0	16.6	0	14.0
	Expansion Turbine	·	0	0	0	0	0	0	0	14.5	0	0	0
Total Installed Investment		MUS$	245.0	283.3	252.5	224.6	282.8	237.9	282.7	206.5	245.9	237.9	248.6
Total Installed Investment corrected for lifetime		MUS$	221.5	256.2	228.3	203.1	255.7	215.1	255.6	186.7	222.4	215.1	224.8

Table 6 continued.

	Biomass input	dry tonne/hour	80.0	80.0	80.0	80.0	80.0	80.0		80.0	80.0	80.0	80.0	80.0
	Biomass input	MWth	428.4	428.4	428.4	432.4	428.4	432.4		428.4	428.4	428.4	428.4	428.4
	Load	hours	8000	8000	8000	8000	8000	8000		8000	8000	8000	8000	8000
	Biomass input	GJ/year	12.3	12.3	12.3	12.5	12.3	12.5		12.3	12.3	12.3	12.3	12.3
Annual Costs	Capital	MUS$	29.1	33.7	30.0	26.7	33.6	28.3		33.6	24.5	29.2	28.3	29.6
	O&M	.	9.8	11.3	10.1	9.0	11.3	9.5		11.3	8.3	9.8	9.5	9.9
	Biomass	.	24.7	24.7	24.7	24.9	24.7	24.9		24.7	24.7	24.7	24.7	24.7
	Costs/Income Power	.	-12.8	-14.8	-25.1	0.0	-3.6	4.2		-17.4	0.2	-20.3	5.4	-17.3
Total Annual Costs		MUS$	50.8	54.9	39.7	60.6	66.0	66.9		52.1	57.7	43.5	67.8	46.8
Production	Fuel output	MW HHV	161.1	172.7	113.4	246.3	220.6	254.8		175.5	259.2	177.1	303.0	149.0
	Power output	MWe	53.3	61.8	104.5	-0.1	14.9	-17.3		72.7	-0.7	84.4	-22.4	72.2
	Efficiency fuel	%	37.6	40.3	26.5	57.0	51.5	58.9		41.0	60.5	41.3	70.7	34.8
	Efficiency power	%	12.4	14.4	24.4	-0.0	3.5	-4.0		17.0	-0.2	19.7	-5.2	16.9
	Efficiency total HHV	%	50.0	54.7	50.9	56.9	55.0	54.9		57.9	60.3	61.0	65.5	51.6
Costs of fuel produced	80 MWth	US$/GJ	16.01	16.78	19.75	12.31	14.80	12.93		15.37	9.89	12.93	10.68	16.65
	400 MWth scale	**US$/GJ**	**10.95**	**11.03**	**12.16**	**8.55**	**10.39**	**9.11**		**10.32**	**7.72**	**8.53**	**7.77**	**10.92**
	1000 MWth	US$/GJ	9.85	9.67	10.45	7.61	9.36	8.14		9.11	7.36	7.57	7.03	7.03
	2000 MWth	US$/GJ	9.21	8.90	9.46	7.14	8.77	7.65		8.39	7.13	7.01	6.65	6.65

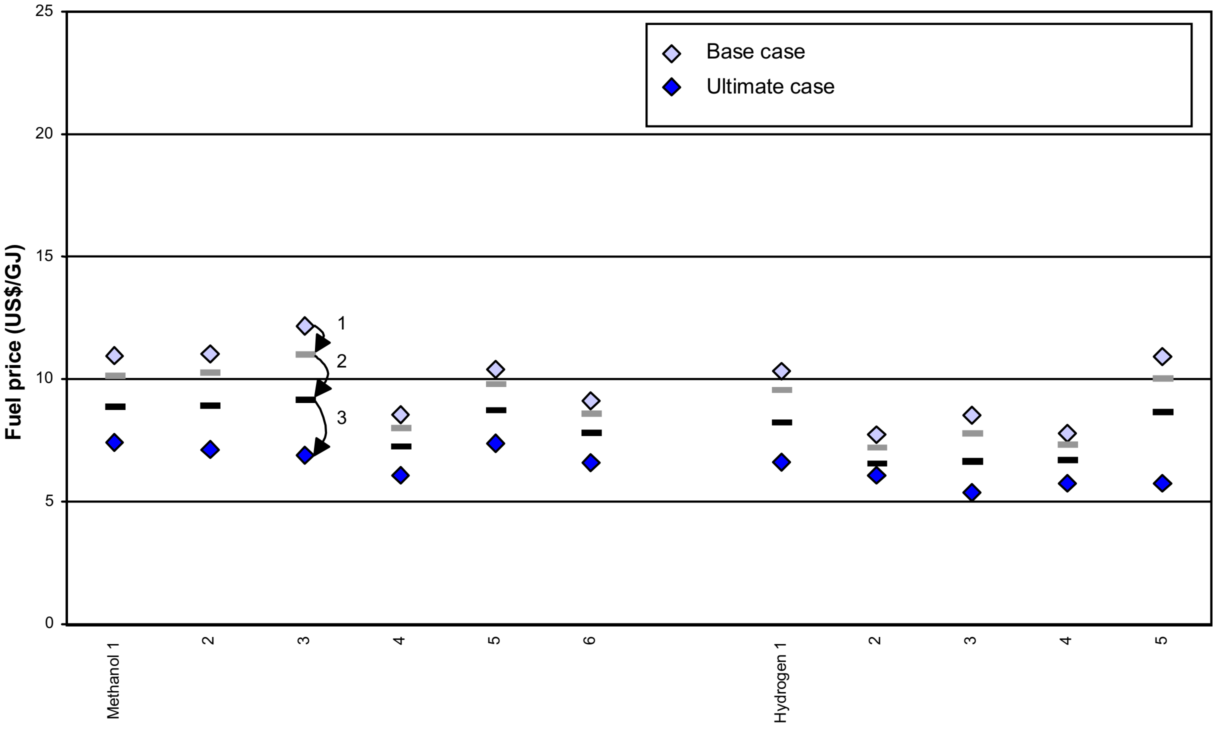

Figure 8. Optimistic view scenario. Different cost reductions are foreseeable: (1) biomass costs 1.7 US$/GJ instead of 2 US$/GJ, (2) technological learning reduces capital investment by 15 % and (3) application of large scale (2000 MW_{th}) reduces unit investment costs.

On the long term different cost reductions are possible concurrently [8]. Biomass could be widely available at 1.7 US$/GJ, capital costs for a third plant built are 15 % lower, and the large (2000 MW_{th}) plants profit from economies of scale. These reductions are depicted in Figure 8: methanol concepts produce against 6.1 – 7.4 US$/GJ, and hydrogen concepts against 5.4 – 6.6 US$/GJ. The influence of capital redemption on the annual costs has strongly reduced and the fuel costs of the different concepts lie closer together. Methanol 4 and Hydrogen 3 give the lowest fuel costs.

In this long-term scenario, methanol produced from biomass costs considerably less than methanol at the current market, which is priced about 10 US$/GJ [64]. Hydrogen from biomass resides in the cost range of hydrogen at a large natural gas reformer plant 5-9 US$/GJ (small amounts of excess hydrogen from large refineries could cost down to 3 US$/GJ [65]). Diesel and gasoline production costs vary strongly depending on crude oil prices, but for an indication: current gasoline market prices lie in the range 4 – 6

US$/GJ [5]. Current diesel prices are around 5 US$/GJ. Longer-term projections give estimates of roughly 8 – 11 US$/GJ [48].

5.3 Biofuel FCV economy

For a well to wheel comparison of biofuels with gasoline, the distribution costs and performance in cars has to be considered, next to the biofuel production cost. This has been done in more detail by Ogden [3] and Faaij [66]. Over long distances hydrogen is preferably distributed in liquefied form by tank trucks or, in future, pipelines. Costs for distribution and for the refuelling station are summarised in Table 7. Compared to gasoline ICEVs, methanol and hydrogen FCVs have similar fuel costs per km driven. However, the FCV will be more expensive: the fuel cell and necessary electricity system cost more than the corresponding elements in an ICEV [3; 67]. The resulting total costs per km driven depend on many assumptions, such as annual capital redemption, annual operating and maintenance costs and the annual amount of km driven. Both Williams [4] and Ogden [3] projected these costs to amount 26 and 27 US$cent/km for methanol and hydrogen, while gasoline costs 27 US$cent/km (without tax).

Next to fuel efficiency, also storage capacity will be important, determining the range. Despite lower fuel economy, the methanol reformed H_2 FCV initially has a larger range, due to the larger storage capacity in its simple fluid form. On the other hand onboard reforming is only an option if the reformer is flexible in providing hydrogen to the fuel cell, as fast or slow as it is being consumed by the fuel cell. If additional hydrogen storage would be necessary, the onboard reformer loses its advantage [68].

For on board hydrogen storage, currently two methods receive the most attention: compressed gas in storage tanks at high pressure or liquid hydrogen in insulated storage tanks at low temperature and pressure. Other methods based on metal hydrides, solid absorbents, and glass micro-spheres have potential advantages but are not as well developed. Hydrogen storage systems can be engineered to be as safe as the fuel systems in current automobiles [68].

Table 7. Build-up of delivered fuel costs and fuel costs per km driven.

	Hydrogen FCV	Methanol FCV	Gasoline FCV
Production (US$/GJ)	5-7	6-7	5-8[1]
Distribution (US$/GJ)	1[2,3]	2[2,4]	1[2]
Tank station (US$/GJ)	5[2]	1[2]	0.5[2]
Delivered (US$/GJ)	11-13[5]	9-10[5]	6.5-9.5[6]
Fuel economy (MJ HHV/km)	0.77[2]	1.18[2]	1.21[7]
Fuel economy (US$cent/km)	1	1	1

[1] BP [5]: 5, DOE/EIA: 8.

[2] Ogden [3].

[3] Pipeline distribution of e.g. $30\cdot10^3$ Nm^3/day over 1 km; costs proportional to distance and inverse to transported volume.

[4] Tank truck distribution.

[5] Faaij [66] projects 8 US$/GJ for hydrogen and 10 US$/GJ for methanol. Ogden [3] projects 12 and 13 US$/GJ. Differences are mainly due to production costs.

[6] Or 36 US$/GJ in the Netherlands when excise duty included.

[7] Current gasoline ICEVs on average consume 2.8 MJ/km. Van Walwijk *et al* [69] projects a large increase in fuel economy to 1 MJ/km, while DOE/IEA projects only a modest increase to 2.5 MJ/km. Gasoline reformed FCVs may achieve 1.21 MJ/km [3].

6 Discussion and conclusions

Promising conversion concepts for the production of methanol and hydrogen from biomass have been evaluated. The concepts incorporate improved or new technologies for gas processing and synthesis and were selected on potential low cost or high energy efficiency. Some concepts explicitly co-produce power to exploit the high efficiencies of once-through conversion. The biofuel production plants were modelled using the Aspen+ flowsheeting program, and optimised towards internal heat demand and supply, surplus heat is converted to electricity. The models directly yielded the plant energy balance and dimensions of streams and units for the economic calculations.

Overall HHV energy efficiencies remain around 55 % for methanol and around 60 % for hydrogen production. Accounting for the lower energy quality of fuel compared to electricity, once-through concepts perform better than the concepts aiming at fuel only production. Also hot gas cleaning generally shows a better performance. Some of the technologies considered in this study are not yet fully proven/commercially available. Several units may be realised with higher efficiencies than considered here: new catalysts and carrier liquids could improve liquid phase methanol single pass efficiency. At larger scales, conversion and power systems (especially the combined cycle) may have higher efficiencies, but this has not been researched in depth.

The biofuel production costs are calculated by dividing the total annual costs of a system by the produced amount of fuel. Unit sizes, resulting from the plant modelling, are used to calculate the total installed capital of biofuel plants; larger units benefit from cost advantages. Assuming biomass is available at 2 US$/GJ, a 400 MW_{th} input system may produce methanol at 9 – 12 US$/GJ and hydrogen at 8 – 11 US$/GJ, slightly above the current production from natural gas prices. The outcomes for the various system types are rather comparable, although concepts focussing on optimised fuel production with little or no electricity co-production perform somewhat better. Hydrogen concepts using ceramic membranes perform well due to their higher overall efficiency combined with modest investment.

The calculated biofuel production cost consists for about 50 % of capital redemption, of which the bulk is in the gasification and oxygen system, syngas processing and power generation units. Further work should give more insight in investment costs for these units and their dependence to scale. The maximum possible scale of particularly the pressurised gasifier gives rise to discussion. The operation and maintenance costs are taken as a percentage of the total investment, but may depend on plant complexity as well. Long term (2020) cost reductions mainly reside in slightly lower biomass costs, technological learning, and application of large scales (2000 MW_{th}). This could bring the methanol and hydrogen production costs in the range of gasoline/diesel and even lower: to 7 and 6 US$/GJ, respectively. Availability of Liquid phase methanol synthesis and ceramic membrane technologies are crucial to reach this cost level. R&D are necessary to verify the performance of these components.

Large-scale production facilities require very large volumes of feedstock. For a small country like the Netherlands, this would imply massive biomass import. Long-distance biomass transport will influence the biomass price, and subsequently the biofuel production costs, as feedstock costs account for about 45 % of the biofuel production costs. Advanced transportation technologies and logistic organisation of large-scale biomass import are currently researched within our department.

Hydrogen as the ultimate fuel for fuel cell vehicles, has a high fuel economy and low costs per km driven, and will certainly compete with gasoline ICEVs or FCVs. However, hydrogen requires new distribution infrastructure – which is the main bottleneck – and further development of on-board storage means. A methanol distribution system is probably easier to realise and FCVs fuelled by on-board reformed methanol will initially have a greater range. More research, focussing on biofuel use through well-to-wheel analysis, is needed to allow a clearer comparison of renewable fuels with their fossil competitors.

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Paper 3

Fischer-Tropsch liquids and power from biomass via gasification: exploration of the possibilities

Contents

Abstract **121**

1 Introduction **122**
1.1 General background 122
1.2 Configurations 122
1.3 Objectives 123
1.4 Methodology 123

2 System description **124**
2.1 The Fischer-Tropsch process 124
2.2 Biomass gasification 127
2.3 Syngas processing 128
2.4 Power generation with a combined cycle 128
2.5 Gas cleaning 130
2.6 Potential system configurations and selection 131

3 System calculations **133**
3.1 Modelling and results 133
3.2 Discussion of results 134

4 Economics **136**
4.1 Basic principles 136
4.2 Calculation of production costs of FT liquids 138
4.3 Sensitivity analysis and longer term perspectives 141

5 Conclusions and recommendations **144**

6 Acknowledgements **146**

References **146**

Fischer-Tropsch liquids and power from biomass via gasification: exploration of the possibilities[*]

Abstract

This paper reviews the technical feasibility and economics of Biomass Integrated Gasification–Fischer-Tropsch (BIG–FT) processes in general, and identifies most promising system configurations and key R&D issues essential for the commercialisation of BIG–FT technology. The FT synthesis produces hydrocarbons of different length from a gas mixture of H_2 and CO. The large hydrocarbons can be hydrocracked to form mainly diesel of excellent quality. The fraction of short hydrocarbons can be used in a combined cycle together with the remainder of the syngas. Total LHV[†] conversion efficiencies (biomass to fuel and electricity), calculated with the flowsheet modelling tool Aspen Plus, could be 33 – 40 % for atmospheric and 42 – 50 % for pressurised gasification systems. Investment costs for 367 MW input BIG–FT systems are 280 – 450 MUS$[‡], depending on the configuration. In the short term, production costs of FT liquids will be about 14 US$/GJ (FT diesel 15 US$/GJ). In the longer term, with large-scale production, higher CO conversion and higher C_{5+} selectivity in the FT synthesis, production costs of FT liquids could drop to 9 US$/GJ. The resulting diesel could then cost 10 US$/GJ. Several improvement options could further reduce the costs, and more minute research could reduce the uncertainty. Therefore, the perspectives for producing FT liquids from biomass, and for their use in the transport sector could still be better. To enable large systems, research and development should focus at large pressurised biomass gasifiers, and investigate their cost/scale relation. Special research attention must be given to the gas cleaning section, to verify that all gas contaminants can be removed to specification.

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[*] Modified version of Tijmensen MJA, Faaij APC, Hamelinck CN and Van Hardeveld MRM, 2002, Exploration of the possibilities for production of Fischer-Tropsch liquids and power via biomass gasification, Biomass and Bioenergy 23(2):129-152.

[†] Energy throughout this paper is expressed on LHV basis, unless indicated otherwise.

[‡] All valuta are in US$$_{2000}$.

1 Introduction

1.1 General background

To prevent climate change induced by human activity, greenhouse gas emissions must be dramatically reduced. Renewable energy (e.g. solar, wind and biomass) could play a major role in achieving this. Biomass is a renewable energy source when carbon dioxide emissions caused by its use are absorbed by newly grown biomass. Biomass offers the possibility to produce liquid carbon neutral (on terms of lifecycle CO_2 emissions) transportation fuels, such as ethanol, methanol, synthetic hydrocarbons, and hydrogen on foreseeable term [1]. This is particularly relevant, since transport is responsible for a large part (about 20 %) of human induced global CO_2 emissions. The share of transport in the total energy consumption is increasing, especially in developing countries [2]. Some recent studies indicated that the use of Fischer-Tropsch (FT) technology for biomass conversion to synthetic hydrocarbons may offer a promising alternative to conventional diesel, kerosene and gasoline [3; 4].

The FT process produces from syngas hydrocarbons of different length, of which the liquid part can be transported by the same means as oil. The recent interest in FT synthesis is fed by the desire to capitalize natural gas from remote locations [5], and by continuing technological developments (catalyst and reactor technology). Furthermore, FT liquids are totally free of sulphur and contain very few aromatics compared to gasoline and diesel, which results in lower emissions from use in internal combustion engines.

Local conversion of stranded natural gas into shippable hydrocarbon liquids by FT is demonstrated on full commercial scale by Shell in Malaysia. Also, Sasol in South Africa applies FT synthesis on commercial scale, to produce diesel from coal. FT synthesis from *biomass* derived syngas, however, has received little attention so far: Larson and Jin [3] presented preliminary energy balances based on a simplified calculation approach, but did not yet perform economic calculations.

1.2 Configurations

In principle, numerous process configurations for the conversion of biomass to FT liquids are possible: Different gasification methods, covering atmospheric & pressurised, air-blown & oxygen-blown, indirect & direct, can produce a wide range of syngas compositions, with H_2/CO ratios varying between 0.45 and 2. Any raw biomass derived syngas contains contaminants like H_2S, NH_3, dust and alkalis. Consequently the syngas needs to be cleaned and processed to make it suitable for the Fischer-Tropsch synthesis, but different cleaning technologies are available. Reforming and shift can be applied to alter the syngas composition prior to the FT reactor. The FT synthesis can be realised in different reactor types. FT synthesis off gas can either be recycled partially, so that the maximum amount of FT liquids is produced (full conversion mode), or used in a combined cycle for electricity production, so that the syngas is most efficiently used for fuels and electricity co production (once through mode). Last but not least, economies of scale are important for this type of technology [6; 7]. In total, all those variables lead to a large number of possible process configurations to produce FT-liquids from biomass.

1.3 Objectives

The main objective of this study is to evaluate the energetic and economic performance of different options to use biomass for the production of FT liquids. This is done by:

- Exploring the technical feasibility and economics of Biomass Integrated Gasification–Fischer-Tropsch (BIG–FT) processes in general, with specific attention for gas cleaning requirements.
- Identifying the most promising system configurations; various biomass gasification processes will be combined with FT synthesis in two main categories:
  1. Full conversion Fischer-Tropsch, focussed on a maximum amount of FT liquids. Use of a combined cycle for electricity co-production is optional.
  2. Once through Fischer-Tropsch with firing the off gas (or co-firing with natural gas) in a combined cycle.
- Investigating the economies of scale of BIG–FT conversion concepts.
- Exploring the technical and economic perspectives of this process on the longer term.
- Identifying key R&D issues for the commercialisation of BIG–FT technology.

1.4 Methodology

The work consists of several steps: First, a technology assessment on gasification, gas cleaning, syngas processing, FT conversion and combined cycle is made. Besides information from literature, experts from technology manufacturers and research institutes were consulted to identify the potential problems of using of FT processes with biomass derived syngas. Manufacturers have been consulted for process data. The assessment includes some technologies that are not applied commercially at present, such as advanced high temperature gas cleaning. Second, promising system configurations were selected for performance modelling with help of the flowsheeting program Aspen Plus to calculate and optimise energy and mass balances. Third, an economic evaluation is performed. Again, manufacturers have been consulted for cost data of various components. Fourth, a sensitivity analysis is performed, including an analysis of economies of scale. And finally, the various system configurations are compared, conclusions are drawn and recommendations on R&D issues are formulated.

2 System description

Figure 1 shows a scheme of the main process steps to convert biomass to FT liquids and power.

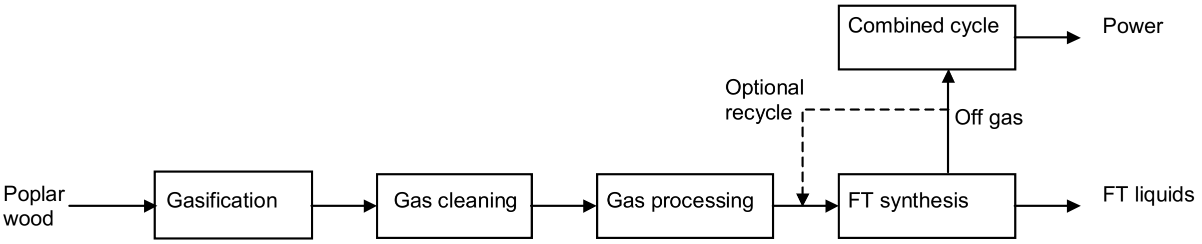

Figure 1. A basic schematic view of the key components for converting biomass to FT liquids combined with gas turbine (combined cycle) power generation.

2.1 The Fischer-Tropsch process

Reaction mechanism and selectivity. The Fischer-Tropsch reaction produces hydrocarbons of variable chain length from a gas mixture of carbon monoxide and hydrogen. The main mechanism of the FT reaction is:

$$CO \; + \; 2H_2 \; \rightarrow \; -CH_2- \; + \; H_2O \qquad \text{Equation 1}$$

The $–CH_2–$ is a building stone for longer hydrocarbons. A main performance characteristic of the FT synthesis is the liquid selectivity of the process. The liquid selectivity is the mass fraction of C_5 and longer, further called C_{5+} selectivity (S_{C5+}). S_{C5+} is determined by the chain growth probability α, the chance that a hydrocarbon chain grows with another $–CH_2–$ group, instead of terminating. The FT synthesis therefore produces hydrocarbons of different length. A high liquid selectivity (maximum amount of long hydrocarbon chains) is desired for the production of diesel. Any C_1-C_4 in the off gas may be used in a combined cycle for power generation.

The relation between the hydrocarbon yield and the chain growth probability is described by the Anderson-Schulz-Flory (ASF) distribution [3; 5; 8] describing the molar yield of each hydrocarbon fraction as:

$$fraction \quad C_n \;=\; \alpha^{n-1} \cdot (1-\alpha) \qquad \text{Equation 2}$$

with α = chain growth probability
C_n = linear hydrocarbon with n number of carbon atoms

Thus, 1-α is the chance that the chain-growth terminates. Figure 2 shows the hydrocarbon yields for different values of α.

Selectivity is influenced by a number of factors, either catalyst dependent (type of metal (iron or cobalt), support, preparation, pre-conditioning and age of catalyst) or non-catalyst dependent (H_2/CO ratio in

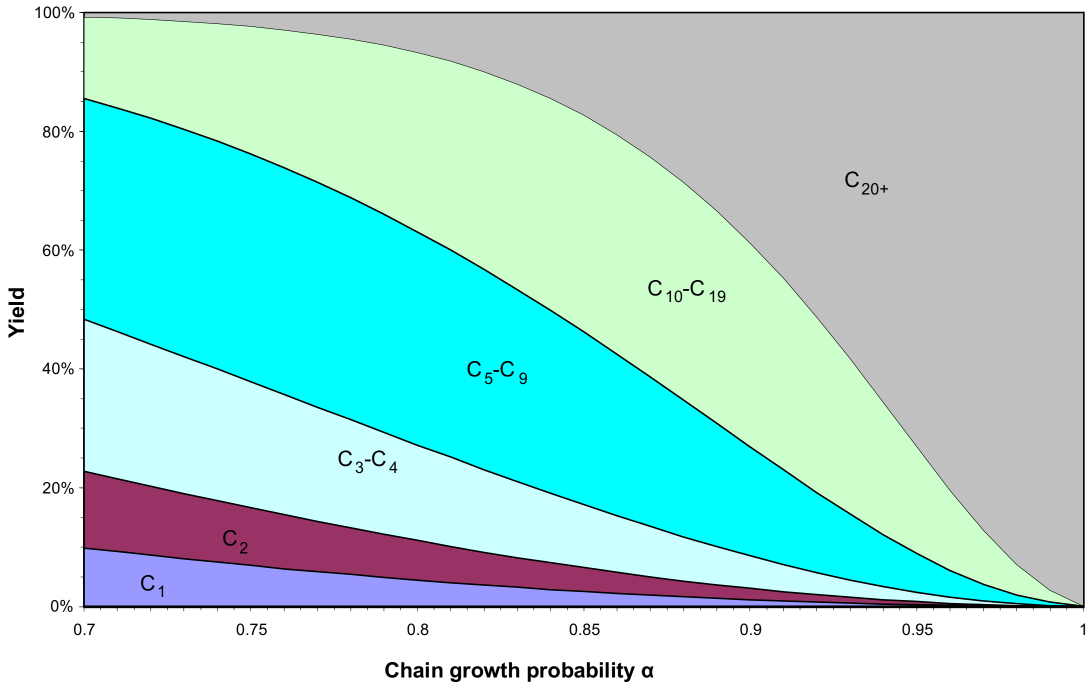

Figure 2. Product distribution for different α for Fischer-Tropsch synthesis.

the feed gas, temperature, reactant partial pressures, and reactor type). The division in iron and cobalt is relevant because the water gas shift reaction, only takes place significantly over an iron catalyst [9].

The FT synthesis uses H_2 and CO at a ratio near 2.1:1, depending on selectivity. Since biomass gasification in most cases leads to a significantly lower H_2/CO ratio in the feed gas, a shift reaction may be necessary

The FT process is generally operated at pressures ranging 20 – 40 bar and at 180 – 250 °C. Higher partial pressures of H_2 and CO lead to higher liquid selectivity S_{C5+}. More N_2 or CO_2, (dependent on gasification technology and gas processing) in the syngas will decrease partial pressures of H_2 and CO, thereby reducing S_{C5+}.

Fischer-Tropsch reactors. There are three main kinds of FT reactors: the fluidised bed reactor, the fixed bed reactor and the slurry phase reactor. The fixed bed (Figure 3, left) and the slurry reactor (Figure 3, right) are the most promising according to many authors [e.g. 10], some favouring the slurry phase reactor [e.g. 3] and some favouring the fixed bed [8]. Specific biomass related advantages for either fixed bed or slurry cannot be given clearly, though sensitivity for inert (relevant for some biomass derived syngas compositions) seems less in a slurry reactor. The main disadvantage of the slurry reactor is the need for catalyst/wax separation, of which no public information appears to be available. Table 1 summarises some key differences between the fixed bed and slurry phase reactors [11].

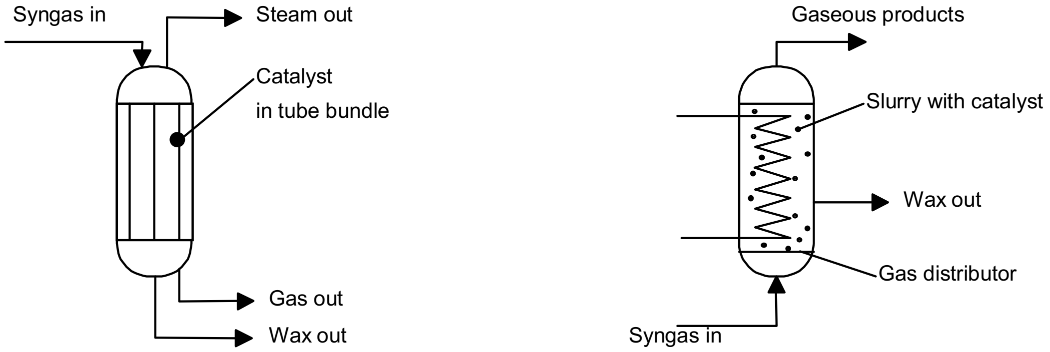

Figure 3. Slurry bed (right) and tubular fixed bed (left) Fischer-Tropsch reactors [11].

Table 1. Differences between fixed bed and slurry FT synthesis processes.

	Fixed bed	Slurry
Economic:		
O&M	- Maintenance and labour intensive and long down time due to periodical catalyst replacement.	+ Little down time due to on-line catalyst replacement. + Lower catalyst consumption [9; 12].
Economies of scale	+ Scale up is straightforward, by multiplying number of tubes - Economies of scale limited [10]	± Possible, but scale up is difficult and not clearly reported [10; 13]
Conversion efficiency:		
Once through conversion	+ Up to 80% is assumed to be possible.	+ High average conversions (once through), up to about 80 % [9; 13]
C_{5+} selectivity	+ Over 90% possible - Negatively influenced by inert	+ Over 90% possible ± Negative influence of inert seems lower [14]
Pressure drop	3-7 bar	< 1 bar [12]
Technical aspects:		
Wax/catalyst separation	+ Performed easily and at low costs.	- Barrier for commercial application although solutions are reported [9]
Process control		+ Easy
Sulphur poisoning		- 1.5 – 2 times higher, so more thorough cleaning required.
Status:		
	+ Proven technology. + Advanced reactors are likely to have higher once through conversion.	+ Overall considered proven technology.

Table 2. Typical product distribution for different wax hydrocracking conditions (weight %). In addition a small percentage C_3 and C_4 are formed [14].

Product split	Gas oil (Diesel) mode	Kerosene mode
Naphtha	15%	25%
Kerosene	25%	50%
Diesel	60%	25%

Hydrocracking. When diesel is the desired final product, the FT product requires hydrocracking. Hydrogen is added to remove double bonds, after which the FT liquids are cracked catalytically with hydrogen. Depending on the wax cracking conditions, mainly diesel or kerosene is produced. The overall carbon efficiency of the hydrocracking step is close to 100 % [12]. Also, hydrocracking conditions can be altered relatively simple to obtain a desired product mix (Table 2).

The FT products are totally free of sulphur, nitrogen, nickel, vanadium, asphaltenes, and aromatics which all are typically found in mineral oil products. FT diesel, with a very high cetane number[§], can also be used as a blendstock to improve the quality of normal diesel. The FT naphtha has a much lower octane[**] number than fossil fuel derived naphtha. FT kerosene for aviation still needs approval of several product specifications. Based on the current product specifications and requirements, these products could therefore have less value than 'normal' naphtha and kerosene. But like FT diesel they contain no sulphur or other contaminants. Besides reducing emissions to air, FT liquids may be well suited for use in fuel cell vehicles (FCVs), which require very clean fuel to prevent damage to the catalyst in reformer and fuel cell. This is a very important characteristic for the longer term when FCVs start penetrating the market [15], as first generation FCVs are likely to onboard reform diesel or gasoline.

2.2 Biomass gasification

Pre-treatment of feedstock. A wide variety of biomass resources can be used as feedstock. Wood, agricultural wastes, organic wastes, and sludges are potential feedstock. However, in this study clean (poplar) wood is assumed to be used. Clean wood gives a relatively clean syngas, with low levels of contaminants. On the longer term wood from dedicated plantations may become a major source of renewable biomass [e.g. 16].

Pre-treatment prior to gasification is required and generally consists of screening, size reduction, magnetic separation, wet storage, drying and dry storage [6]. Moisture content of the 'wet' poplar chips delivered is assumed to be 30 %. Drying (to 10 – 15 % moisture) is generally the most important pre-treatment operation, necessary for high cold gas efficiency in gasification [17]. This can either be done with flue gas or with steam. Since, as will be discussed, significant amounts of low quality steam are generated in the FT process, steam drying is preferable (e.g. a Niro steam dryer [6]). Also, steam drying yields less emission and may eventually be safer with respect to risks for dust explosion, compared to flue gas drying.

Gasification. Conversion of biomass to an H_2 and CO containing feed gas that is suited for FT synthesis takes places through gasification. Gasification can be done at different pressures, directly or indirectly heated (lower temperatures), and with oxygen or air. Since for economic and efficiency

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[§] Cetane number is a primary indicator of diesel fuel quality. It is essentially a measure of the delay before ignition. The shorter the delay the better – and the higher the cetane number

[**] Octane number is a quality rating for gasoline, indicating the ability of the fuel to resist premature detonation and to burn evenly when exposed to heat and pressure in an internal combustion engine. Normal gasoline has a octane number of 87-89.

considerations, the capacities investigated in this analysis start at 100 MW input, only CFB gasifiers have been taken into account [1; 7; 18]. Some key advantages and disadvantages of each gasification method are shown in Table 3.

Table 3. Main technical aspects gasification method.

Pressurised	**Atmospheric**
+ Pressurised equipment upstream is smaller and generally more economical at larger scales (see 4.1) - Higher costs of gasifier at small scale - High risk in keeping constant mass flow in gasifier, operational experience so far limited to demonstration projects [19]	- Larger equipment upstream needed + Less costs at small scale (see 4.1) + Significant commercial experience
Oxygen	**Air**
- Air separation plant needed, especially at small scales relatively expensive + No dilution of syngas by N_2	+ Cheaper - N_2 diluted syngas, has negative influence on C_{5+} selectivity - Larger equipment needed upstream
Direct	**Indirect**
+ Less tars produced (tars presence in syngas is one of the biggest problems when gasifying biomass)	+ No N_2 dilution even if air is used - Bigger tar problem

To cover the wide range of gasification methods, different gasifiers currently available and/or under development were selected for further study (see Table 4). These gasifiers produce a wide range of syngas compositions representing the reasonably maximum possible variation in CO:H_2 ratios that can by obtained.

The syngas produced by the different gasifiers contain various contaminants: particulates, condensable tars, alkali compounds, H_2S, HCl, NH_3, HCN and COS [20]. However, no full data sets of syngas compositions including all these contaminants are available for the gasifiers considered. Therefore, some estimations and assumptions are required (see section 2.5).

2.3 Syngas processing

The syngas, produced by the gasification of biomass, consists mainly of H_2, CO, CO_2 and CH_4. Their shares in the syngas can be tailored to the needs of the FT process by reforming (converts CH_4 with steam to CO and H_2), a shift reaction (adjusts the H_2/CO ratio by converting CO with steam to H_2 and CO_2) and CO_2 removal, which reduces the amount of inert for the FT synthesis. For reforming, the autothermal reformer (ATR) is selected. An amine treating process is used for CO_2 removal. More extensive component descriptions and performance data can be found in literature [7; 18; 21].

2.4 Power generation with a combined cycle

It is possible to use the off gas from the FT reactor in a combined cycle for additional electricity production. At high pressure, the off gas is mixed with pressurised air and combusted at about 1100 – 1300 °C. Expansion of the resulting hot flue gas in a gas turbine generates electricity. Next the flue gas superheats steam, which generates electricity in a condensing steam turbine. Various other (gas) streams in the whole process require cooling, e.g. the syngas after gasification or reforming. Also at these places

Table 4. Operating characteristics of the gasifiers evaluated in this paper, based on poplar wood.

Gasifier	**BCL**[a]		**IGT**		**IGT+**[a]		**EP**[c]		**TPS**[d]	
Process type	**Indirect**		**Direct**		**Direct**		**Direct**		**Direct**	
	Atmospheric		**Pressurised**		**Pressurised**		**Pressurised**		**Atmospheric**	
	Air blown		**Oxygen blown**		**Oxygen blown**		**Air blown**		**Air blown**	
Characteristics:										
P (bar)	2		34		20.3		22		1.3	
T (K), exit	1136		1255		1241		1223		1173	
Moisture dry biomass	10%		15%		15%		15%		15%	
Pilot size (dry tons/day)	10		100		No pilot		-		270	
Flow rate dolomite (kg/kg wet)	-		0		0		0.0095		0.0268	
Flow rate air/oxygen[e]	1.46 kg/kg dry		0.3 kg/kg dry		0.3 kg/kg dry		-		1.4 kg/kg wet	
Steam (kg/kg wet input)	0.19		0.34		0.6		0,34		0.34	
Yield (kmol/dry tonne)	45.8		82.0		123.1		113.3		112.1	
LHV syngas (MJ/Nm^3 wet gas)	13.9		7.3		4.8		5.8		5.2	
Gasifier efficiency[f]	86.8		80.7		80.9		88.6		80.0	
H_2/CO ratio	0.45		1.39		2.0		0.73		0.77	
Composition (mol %)	*wet*	*(dry)*								
H_2O	19.9	(0)	31.8	(0)	50.6	(0)	13.6	(0)	13.6	(0)
H_2	16.7	(20.8)	20.8	(30.5)	15.7	(31.7)	10.0	(11.6)	13.3	(15.3)
CO	37.1	(46.3)	15.0	(22.0)	7.8	(15.9)	13.8	(16.0)	17.2	(19.9)
CO_2	8.9	(11.1)	23.9	(35.0)	17.7	(35.9)	15.4	(17.8)	12.2	(14.1)
CH_4	12.6	(15.7)	8.2	(12.0)	5.7	(11.6)	7.3	(8.4)	2.8	(3.26)
C_{2+}	4.8	(6.0)	0.3	(0.5)	0		0.5	(0.6)	1.0	(1.1)
C_2H_4	4.2	(5.2)							0.9	
C_2H_6	0.6	(0.74)							0.02	
N_2+Ar	0		0		0.40	(0.80)	38.9	(45.3)	39.2	(45.3)
Others	< 0.3		< 0.3		< 0.3		< 0.3		< 0.3	

[a] Batelle Columbus gasifier [1]. The operation pressure is an assumption.

[b] Institute of Gas Technology [18]. Two gas compositions are included for pressurized, oxygen blown gasification: one is at 'standard' conditions (IGT), the other at operating conditions leading to maximised hydrogen production, which is obtained by increased steam addition and reduced pressure (IGT+). This is a more theoretical case, but allows for exploring the consequences of a more optimal CO:H_2 ratio at the cost of higher steam consumption, lower pressure and a lower heating value of the (wet) syngas.

[c] Enviro Power with dolomite tar cracker [6]. The pilot size is unknown, the steam amount is an assumption.

[d] Thermiska Processer with dolomite tar cracker. The amount of steam is an assumption.

[e] Oxygen of 99.5 % purity is used. Oxygen of 95 % purity would require 305 kWh per tonne, of 99.5 % will require 15 % extra energy [22].

[f] Gasifier efficiency is defined as energy content of the syngas divided by the energy content of the biomass input, based on LHV. Energy content of additional steam, air and/or oxygen is not accounted for.

superheated steam is generated. It is first used in the gasifier and reformer; any remaining superheated steam is expanded in the steam turbine. Low temperature steam can also be used for drying and other steam demanding processes, such as the shift reaction. Part of the generated electricity is used within the process.

In case of BIG–FT systems, the caloric value of the off gas may be too low for combustion in a gas turbine. Typically, minimum heating values required for commercial gas turbines are 4 – 6 MJ/Nm^3, assuming some modifications on burners and fuel manifolds [6; 23]. Co-firing of natural gas avoids problems by raising the heating value of the gas. Moreover, the larger throughput enables the use of more efficient turbines. Co-firing is therefore included in this analysis.

2.5 Gas cleaning

General. The syngas produced by the gasification process contains different kinds of contaminants: Particulates, condensable tars, alkali compounds, H_2S, HCl, NH_3 and HCN [20]. These contaminants can lower activity in the FT synthesis by catalyst poisoning. Sulphur is an irreversible poison for the cobalt and iron catalysts (and to a smaller extent for the shift and reformer catalysts), because it will stick to active site, and shortens catalyst life. Tolerance for contaminants is low and thorough cleaning is required. Two distinct cleaning routes will be considered in this study: Wet low temperature cleaning and dry high temperature cleaning.

Conventional wet low temperature cleaning. Conventional cleaning (Figure 4) was proposed to clean the fuel gas for BIG/CC installations [6; 20]. However, cleaning requirements for the FT synthesis are much more stringent. Specifications for FT are given in Table 5 and compared to syngas compositions typical for CFB gasification of clean wood. At present, relatively clean natural gas is the common feedstock for FT synthesis, and actual cleaning specifications for some specific biomass contaminants are not known. Therefore, some specifications are estimates. The specification for sulphur, however, is explicitly known, since it is also present in natural gas and known to irreversibly poison the FT catalyst.

Although with proper sizing and possible addition of active coal filters it is likely that the specifications can be met, this needs to be tested and verified in practice. In particular potential problems with tars, which foul downstream equipment and sorbents, require further research.

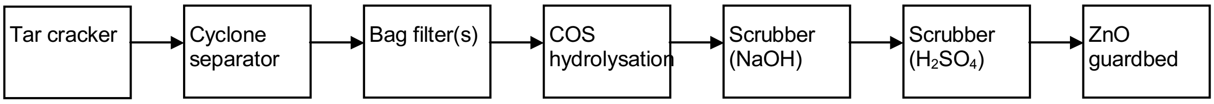

Figure 4. Schematic set-up of ‘wet’ low temperature cleaning.

Advanced ‘dry’ hot gas cleaning. Hot gas cleaning consists of several filters and separation units in which the high temperature of the syngas can partly be maintained, potentially resulting in an efficiency benefit and lower operational costs. Hot gas cleaning has an efficiency advantage when preceding a reformer or shift reactor, because these process steps have high inlet temperatures. When FT synthesis would be applied directly after the gas cleaning, the syngas has to be cooled to 200 °C anyway, and the potential benefits are expectedly less apparent. Hot gas cleaning after atmospheric gasification does not improve efficiency, because the subsequent essential compression also requires syngas cooling.

Hot gas cleaning is not commercial practice yet; some unit operations are still in the experimental phase. It is in this light uncertain whether hot gas cleaning can meet the required high purification specifications of FT synthesis in a foreseeable timeframe. Research so far is mainly focused on developing hot gas cleaning to meet the less severe requirements of BIG/CC installations. There are no commercial processes for the high temperature removal of nitrogen compounds, halides, alkali metals and heavy metals yet, although various solutions are worked on [6; 20; 24].

Table 5. Contaminant concentrations from gasifying poplar wood (weight % in syngas), maximum values for FT synthesis (ppb), cleaning set-up in wet low temperature cleaning, and performance.

Contaminant	Concentration[a)] (wt %)	Specification (ppb)	Required cleaning	Cleaning set-up	Performance[b)]
Particulates	1.33	0	> 99.9 %	·Cyclone separator ·Bag filters/scrubber.	+
HCN+ NH_3	0.47	20	> 99.9 %	·Scrubber (possibly with H_2SO_4) ·Amine treating also removes HCN and NH_3.	+ ?
H_2S+COS	0.01	10	> 99.9 %	·Scrubber ·Possibly COS hydrolisation unit or amine treating necessary ·ZnO guard bed	+ + ZnO guard beds are also used for natural gas based FT
Alkalis	0.1	10	> 99.9 %	·During cooling down alkalis condense on particulates, Possibly also on vessels (and thereby polluting them)	+ ?
HCl	0.1	10	> 99.9 %	·Absorbed by dolomite in tar cracker (if used), reaction with particulates in bag filter, scrubber (possibly with NaOH)	+
Pb and Cu	0*	Not known	-	·Condense on particulates, but actual behaviour has not been studied	?
Tars	-	0	> 99.9 %	·Condense on particulates and vessels when syngas is cooled below 500 °C	Potential tar problem, limited experience with complete removal or conversion

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[a)] Assumed feedstock is dry poplar wood [18], except alkalis and heavy metals hold for miscanthus [17], and tars concentration is not known, but order of magnitude is g/Nm^3.

[b)] Performance is expressed as “+” requirement achieved, “?” some uncertainty remains.

Discussion. Conventional syngas cleaning is the preferred technology in the short term [20]. This technology will have some efficiency penalties though, and requires additional waste water treatment but there is little uncertainty at present about the cleaning effectiveness of such systems with respect to IG/CC installations, both for coal and biomass fired facilities [23]. However, actual testing with biomass FT systems is still necessary to ensure the effectiveness for these systems.

Within 10 years hot gas cleaning may become commercially available for BIG/CC installations [6; 25], but opinions differ. Therefore, hot gas cleaning is considered advanced option for the longer term in this analysis, to show the possible effect of hot gas cleaning on the performance of BIG/FT systems.

2.6 Potential system configurations and selection

Key elements. In theory, a large number of system configurations to convert biomass to FT liquids and power is possible (Table 6). Since it is difficult to predict a specific S_{C5+} for biomass derived syngas, e.g. due to different inert percentages, a realistic S_{C5+} range of 73.7 to 91.9 % is modelled. This corresponds to chain growth probabilities α of 0.8 to 0.9. As a conventional option a CO conversion of 40 % per pass is assumed for the full conversion concepts. To increase the overall CO conversion, gas recycle (maximally equal to feed) has been employed. As an advanced option 60 % and 80 % once through CO conversion is assumed for the once through concepts. Advanced FT reactors, either fixed bed or slurry, can be expected to obtain higher once through CO conversion [14]. For all concepts an external shift reactor will be used, implying the use of a cobalt catalyst for FT.

After the FT synthesis the product stream is cooled to 50 °C to separate the C_{5+} fraction as liquid. Further cooling to separate C_3 and C_4 could be done, but whether this is economically attractive is questionable. In the concepts modelled, the C_1-C_4 fraction is used in a combined cycle for power generation.

Table 6. Possible choices in system components leading to different system configurations for BIG FT systems.

Gasifier	Gas cleaning	Reforming	Shift	CO_2 removal	FT reactor	Recycling
·BCL ·IGT ·IGT+ ·EP ·TPS	·Low temperature ·High temperature	·Yes ·No	·Intern (iron catalyst) ·Extern (cobalt catalyst)	·Yes ·No	·Fixed bed ·Slurry	·Yes (full conversion) ·No (once through)

Full conversion Fischer-Tropsch, with possible use of a gas turbine. The goal of the full conversion concepts is to maximise the yield of FT liquids (and to a smaller extent power production). Systems both including and excluding reforming are modelled, to study the pay off between higher FT yield and lower capital investments. The IGT+ option is modelled without reformer or shift, otherwise the basic idea of creating a syngas with a 'perfect' H_2/CO ratio would not make sense. In case of the direct atmospheric gasification systems, reforming with air would lower the H_2 and CO content in the syngas since some H_2 and CO have to be combusted to obtain the necessary process heat. Therefore, this configuration is not modelled.

Amine based CO_2 removal is included in all configurations, as less CO_2 allows for a high C_{5+} selectivity and will therefore increase the amount of FT liquids produced. On the other hand, CO_2 removal is an expensive process and it is questionable whether the high costs are justified by the increased production of FT liquids. The impact of in- and excluding CO_2 removal is therefore investigated.

Once through FT, with co firing in a (150 MW_e) natural gas based gas turbine. The once through concepts produce both FT liquids and power. All are modelled without reformer or CO_2 removal, so FT synthesis is not maximised. This will result in lower capital costs and possibly somewhat higher total (FT + electricity) energy efficiencies. Only the concepts with IGT gasifiers are modelled with CO_2 removal, because of their high syngas CO_2 content.

3 System calculations

3.1 Modelling and results

Modelling of the various concepts has been performed with the flowsheeting program Aspen Plus. A basic flowsheet is presented in Figure 5. The gasification processes, however, have not been modelled in Aspen Plus, because the fuel gas compositions resulting from most biomass gasification processes are determined by kinetics instead of equilibrium conditions and difficult to model. Thus, fuel gas compositions from literature are used as starting point for the calculations. The base capacity for all system calculations is fixed at 80 tonne_{dry}/hr biomass input (at 30 % moisture, 16.5 $GJ_{LHV}/\text{tonne}_{dry}$ this is 367 MW_{LHV} input).

Table 7 summarises the key data used for Aspen modelling. The overall energy efficiencies[††] of the full conversion concepts are presented in Table 8.

The BCL-R and the EP-R concepts produce off gas with a caloric value probably too low for (direct) use in a gas turbine, as the methane and ethane from the gasifier are converted to FT liquids. Nevertheless, the gas turbine is of particular importance for those concepts because otherwise a substantial amount of energy in the form of light hydrocarbons ($C_1 – C_4$) would be wasted. Co-firing with natural gas upgrades the heating value of the off gas and increases the turbine efficiency. Low calorific gas could also be combusted in a boiler, but this is not assessed.

The overall energy efficiencies of the once through concepts are presented in Tables 9 and 10. All once through concepts make use of a 150 MW_e gas turbine, so that a certain amount of complementary natural gas is necessary. The net power output in the table is allocated to the fraction of the energy input of biomass derived off gas.

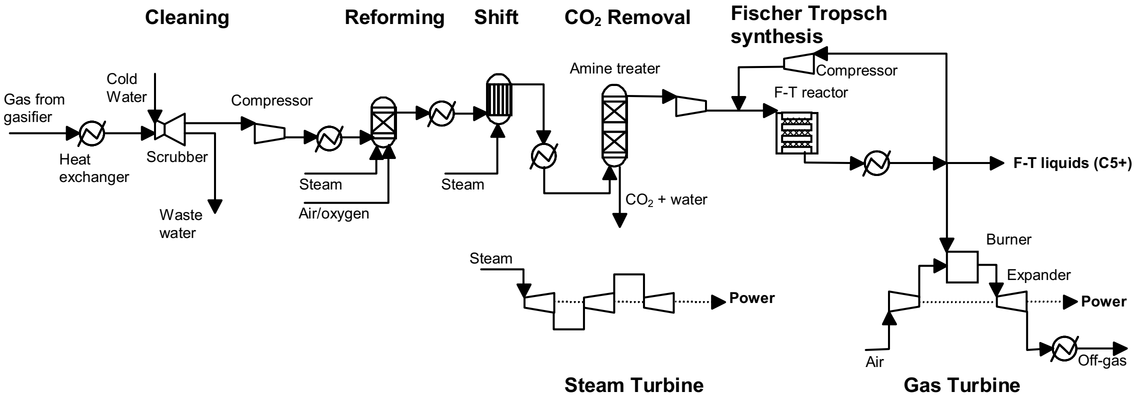

Figure 5. Base Aspen Plus flowsheet, used for the calculation of the mass and energy balances. Steam is generated and needed at various places in the facility. The net amount of valuable steam (70 bar) is used in the steam turbine.

[††] The overall energy efficiency of the systems is defined as the sum of all outputs / total biomass input.

Table 7. Generic input data for Aspen modelling.

Dryer	Per tonne biomass 0.41 tonne steam needed for drying to 10 % moisture content, 0.33 for drying to 15 %
Wet cold gas cleaning	T_{in} = 400 °C, T_{out} = 40 °C below dew point, Δp = -0.5 bar at $p > 30$ bar, Δp = -0.3 bar at 10 bar < p < 30 bar, Δp = -0.2 bar at $p < 10$ bar
Hot gas cleaning	T_{out} = 450 °C, Δp = -1 bar
ATR	p = 33 – 44 bar, Δp = -0.5 bar, T_{in} = 400 – 450 °C, T_{out} = 950 °C, T_{air} = 600 °C, T_{oxygen} = 300 °C
Shift	15 bar < p < 70 bar, Δp = -0.5 bar, T_{in} = 330 °C
FT reactor	p = 40 bar, Δp = 5 bar, T_{in} = 200 °C, T_{out} = 240 °C, effective $H_2/CO_{outlet} > 0.4$ Recycle maximally equal to feed (mol), FT steam at p = 22 bar and T = 230 °C
Gas turbine	Compressor: $\eta_{isentropic} = 0.91$, $\eta_{mechanical} = 1$ Combustor: T = 1200 °C but if LHV < 6 MJ/Nm^3 then T = 1100 °C Expander: $\eta_{isentropic} = 0.89$ (0.9 when co fired), $\eta_{mechanical} = 1$ p_{out}=1.2 bar (0.2 bar needed for heat exchanger), T_{out} (after heat exchanger) > 170 °C[a]
Steam turbine	Stages pressure (bar) & temperature (°C): 70 & 500, 41.4 & 440, 22 & 375, steam to FT reactor: 230 °C, steam to dryer: 2 bar, steam to gasifier: 1 – 34 bar, steam to shift: 15.5 – 43 bar, steam to ATR: 20 – 44 bar, Bottom steam turbine: 0.04 bar
Heat exchangers	Δp = -0.5 bar at $p > 30$ bar, Δp = -0.3 bar at 10 bar < p < 30 bar, Δp = -0.2 bar at $p < 10$ bar, maximum syngas to syngas heat exchange at 400 °C[b]

[a] Minimum outlet temperature is due to environmental constraints [6].

[b] Due to coking problems at higher temperatures [1].

3.2 Discussion of results

The modelled concepts give the following insights:

1. The concepts with high overall energy efficiency are based on pressurised gasifiers. When high C_{5+} selectivity (91.9 %) is assumed, IGT-R has an total conversion efficiency of 50.1 % (fuel + electricity); when hot gas cleaning is used this becomes 51.5 %. The 80 % once through concepts show that conversion efficiencies of near 50 % are obtainable for the EP and the IGT gasifiers, even without the use of a reformer. Larson and Jin [3] earlier found efficiencies to range from 44 – 49 % for concepts based on the atmospheric indirect heated gasifier.
2. CO_2 removal has little direct effect on overall energy efficiency – but does have an effect on the amount of inert and thus on liquid selectivity. Question remains whether the investment is justified by the improvement of selectivity.
3. Higher C_{5+} selectivity leads to higher total conversion efficiency for all concepts. The obtainable S_{C5+} is uncertain, but will be in the range of 74 – 91 % ($\alpha = 0.8 – 0.9$). When much inert is present (as is the case with air gasification), S_{C5+} will probably end up in the lower part of this range.
4. The concepts with 80 % CO conversion have higher total conversion efficiencies than the concepts with 60 % conversion. Thus, a high overall CO conversion has a beneficial effect on efficiency.
5. Pressurised concepts have higher total conversion efficiencies than atmospheric concepts. The difference is about 10 % points. This is mainly due to the high electricity consumption of syngas compression when direct, air blown, atmospheric gasifiers are used.
6. Reforming concepts are most sensitive to C_{5+} selectivity and only improve the total conversion efficiency when S_{C5+} is high. For the BCL concepts reforming does even improve efficiency at low S_{C5+}, due to the high methane and ethane content of the syngas.

Table 8. Total LHV conversion efficiencies and fuel and electricity output for the full conversion (40 % CO conversion per pass, max recycle rate is 2) concepts. α = 0.8 corresponds with S_{C5+} = 73.7 %; 0.85 with 83.5 %; 0.9 with 91.1 %.

Gasifier **Configuration**[a)]	**BCL** **R**	**BCL** **R-nt**	**BCL**	**IGT** **R**	**IGT** **R-hg**	**IGT**	**IGT+**	**EP** **R**	**EP** **R-nt**	**EP**	**TPS**
α = 0.80											
eff. (%)	45.1	30.1	35.9	44.7	46.1	46.0	44.6	41.6	25.4	42.4	32.9
FT-liquids (MW)	123.9	123.9	66.8	129.9	132.6	81.1	78.4	100	100	64.6	83.1
Power (MW)	41.4	-13.6	65.0	34.2	36.6	87.5	85.4	52.5	-6.7	90.9	37.6
α = 0.85											
eff. (%)	47.0	33.5	37.1	47.7	49.1	47.4	44.9	44.8	29.4	43.4	34.5
FT-liquids (MW)	139.1	139.1	75.9	150.7	153.4	91.9	89.8	113.3	113.3	73.2	94.2
Power (MW)	33.3	-16.3	60.4	24.4	26.8	82.2	75.0	50.9	-5.5	86.2	32.5
α = 0.90											
eff. %	48.0	38.2	38.1	50.1	51.5	48.2	47.3	45.4	32.2	44.5	35.8
FT-liquids (MW)	154.8	154.8	83.4	168.7	171.4	101.7	97.9	124.6	124.6	80.5	103.6
Power (MW)	21.3	-14.6	56.5	15.2	17.4	75.1	75.7	42.0	-6.5	82.9	27.6

[a)] Explanation of used codes for configurations: R = reformer used, nt = no gas turbine used, hg = hot gas cleaning.

Table 9. Total LHV conversion efficiencies and fuel and electricity output for the 60 % CO conversion once through concepts. α = 0.8 corresponds with S_{C5+} = 73.7 %; 0.85 with 83.5 %; 0.9 with 91.1 %.

Gasifier **Configuration**[a)]	**BCL**	**IGT**	**IGT** **A**	**IGT+**	**IGT+** **A**	**EP**	**TPS**
α = 0.80							
eff. (%)	35.8	44.1	43.9	43.5	43.6	43.6	33.9
FT-liquids (MW)	49.9	58.9	58.9	57.8	57.8	54.8	69.3
Power (MW)	81.3	103.0	102.2	101.9	102.4	105.1	55.2
α = 0.85							
eff. (%)	37.2	44.6	44.5	44.6	44.7	44.6	35.2
FT-liquids (MW)	56.6	66.8	66.8	65.4	65.4	62.0	78.5
Power (MW)	79.8	97.0	96.4	98.1	98.8	101.8	50.7
α = 0.90 (MW)							
eff. (%)	38.0	45.6	45.4	45.4	45.7	45.5	36.3
FT-liquids (MW)	62.2	73.5	73.5	72.0	72.0	72.0	86.3
Power (MW)	77.2	93.7	93.1	95.0	95.6	98.6	47.0

[a)] Explanation of used codes for configurations: A = Amine treating used.

Table 10. Total LHV conversion efficiencies and fuel and electricity output for the 80 % CO conversion once through concepts. α = 0.8 corresponds with S_{C5+} = 73.7 %; 0.85 with 83.5 %; 0.9 with 91.1 %.

Gasifier **Configuration**[a)]	**BCL**	**IGT**	**IGT** **A**	**IGT+**	**IGT+** **A**	**EP**	**TPS**
α=0.80							
eff. (%)	37.7	46.4	46.2	45.6	45.7	47.2	36.5
FT-liquids (MW)	66.6	78.6	78.6	77.0	77.0	73.0	92.4
Power (MW)	71.9	91.5	91.0	90.4	90.8	100.4	41.5
α=0.85							
eff. (%)	38.9	47.8	47.7	47.0	47.1	48.6	38.2
FT-liquids (MW)	75.4	89.1	89.1	87.3	87.3	82.7	104.6
Power (MW)	67.4	86.3	85.9	85.3	85.8	95.7	35.6
α=0.90							
eff. (%)	40.4	49.0	48.9	48.2	48.3	49.8	39.7
FT-liquids (MW)	82.9	98.0	98.0	96.0	96.0	91.0	115.1
Power (MW)	65.3	81.9	81.6	81.0	81.4	91.6	30.5

[a)] Explanation of used codes for configurations: A = Amine treating used.

7. Hot gas cleaning improves the efficiency for pressurised concepts, with on average 1 to 2 % points.

The main energy losses in the model are caused by the gasification section (η ~ 80 %), the shifting and reforming section (η ~ 90 %), the FT section (η ~ 78 % for the converted CO and H_2) and the combined cycle (η ~ 50 %). It is important to note that the small hydrocarbons ($C_1 – C_4$) formed in the FT synthesis and used in the gas turbine, lead to energy losses both in the FT section and in the combined cycle. Also, from the FT section (35 bar out) to the gas turbine (14 – 16 bar in) the syngas loses pressure without producing electricity. An expansion turbine could be used to reduce this energy loss, but is not included here.

4 Economics

4.1 Basic principles

The calculation of the Total Capital Requirement (TCR) is based on component level cost data, which were obtained from literature, vendor quotes and personal communication with experts. The specific costs of most system components are affected by their capacity. The capacity or scale of each component is derived from the energy and mass balances obtained through the Aspen Plus modelling. From an equipments base cost, base scale, scale factor R (Table 11), and the calculated scale in the concept the actual equipment cost can be calculated:

$$\frac{Cost\ a}{Cost\ b} = \left(\frac{Size\ a}{Size\ b}\right)^{R} \qquad \text{Equation 3}$$

with R = scaling factor (between 0 and 1)

A lower R indicates larger cost reductions with increasing scale. A maximum size for each unit is taken into account, above which increasing scale is no longer economically attractive or possible (see Table 11). When the total capacity of a conversion unit exceeds this maximum component capacity, it is assumed that multiple units are built to meet the desired capacity. The gasifiers used have different maximum sizes. The maximum size of a gasifier is mainly determined by two factors: whether the gasifier operates at elevated pressure and whether the plant is located near a harbour. If road transport is considered, the dimensions of the road are of importance. When transport of the gasifier to location can be done over water, much larger single units can be installed. For the TPS, BCL, IGT and EP gasifiers, the maximum sizes assumed are 122, 200, 400 and 400 MW_{HHV} respectively. Exact maximum scales cannot be given because existing plants of such size are not yet realised in practice, but it can be expected that larger single pressurised gasification reactors can be built than assumed here. This aspect requires further research.

For making cost estimates for much larger and smaller scales compared to the base capacity of 367 MW overall scale factors for the whole plant (not per equipment) are used or respectively 0.74 and 0.91, see Section 0.

Table 11. Basic costs for all units used with their maximum size (base costs are in relation to base scales). Costs expressed in MUS$.

	Base cost (MUS$)	**Scale factor**	**Installation factor**[a]	**Base scale**		**Maximum size**
Pre-treatment:						
conveyers[b]	0,33	0,8	1.53	69.5	MW_{LHV} input	367
grinding[b]	0,43	0,6	1.53	69.5	MW_{LHV} input	367
storage[b]	1,05	0,65	1.53	69.5	MW_{LHV} input	367
dryer[b]	7,71	0,8	1.53	69.5	MW_{LHV} input	367
iron removal[b]	0,33	0,7	1.53	69.5	MW_{LHV} input	367
feeding system[b]	0,38	1	1.53	69.5	MW_{LHV} input	367
Gasifiers:						
gasifier TPS[b]	3,24	0,7	1.53	69.5	MW_{LHV} input	105
gasifier BCL (incl. feeding)[c]	13,0	0,7	1.53	400	MW_{HHV} input	200
gasifier IGT and EP[d]	30	0,7	1.53	400	MW_{HHV} input	400
Gas cleaning:						
tar cracker[b]	3,24	0,7	1.53	69.5	MW_{LHV} input	105
cyclones[b]	2,57	0,7	1.53	69.5	MW_{LHV} input	367
gas cooling[b]	2,95	0,7	1.53	69.5	MW_{LHV} input	367
baghouse filter[b]	1,62	0,65	1.53	69.5	MW_{LHV} input	367
condensing scrubber[b]	2,57	0,7	1.53	69.5	MW_{LHV} input	367
hot gas cleaning[e]	14,3		1.53	400	MW_{HHV} input	
Compressors:						
compressor[f]	12,0	0,85	1.53	13,2	MW_e	
Combined cycle:						
gas turbine[b]	7,7	0,7	1.53	25	MW_e	
modifications turbine LCV gas[b]	8%	0,7	1.53	69.5	MW_{LHV} input	
HRSG[b]	3,38	0,8	1.53	47,5	tonne/hour	
Steam turbine + condenser[b]	4,48	0,7	1.53	12,3	MW_e	
water+steam system[b]	0,43	0,9	1.53	49,5	tonne/hour	
cooling[b]	0,95	0,3	1.53	50,5	tonne/hour	
Other						
oxygen plant[g]	7.7	0,75	3	24	tonne/hour	
ATR reactor[H]	10.1	0,7	3	400	MW_{HHV} input	
Shift reactor[i]	0,15	0,6	3	2400	kmol/hour	
Amine treating[j]	Confidential	0,65	3	400	MW_{HHV} input	
FT reactor[k]	5.6	1	3	100	MW_{FT}	
ZnO beds[l]	0,043	1	3	400	MW_{HHV} input	

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[a] For most components, the FOB (Free On Board) price is raised with 53 % [6] to obtain the installed costs: This installation factor consists of 33 % added investment to hardware (instrumentation & control 5 %, buildings 1,5 %, grid connections 5 %, site preparation 0,5 %, civil works 10 %, electronics 7 %, piping 4 %) and 15 % added installation to investment (engineering). For the bottom 6 components these percentages are probably not valid. Therefore installed costs were calculated directly, or the FOB price was tripled to obtain installed costs.

[b] Costs figures based on first generation BIG/CC installations [6].

[c] [26].

[d] The Enviro Power gasifier and the IGT gasifier, both operating at elevated pressure, have likewise designs. Therefore cost figures from IGT [26] are assumed applicable for EP.

[e] These costs are an assumption. No real world data are available.

[f] Vendor quote (Sulzor)

[g] 576 t/d oxygen production of 95 %v purity has a capital cost of 31,000 US$ per t/d, for 1008 t/d the costs are 27,000 per t/d [22]. Oxygen of 99.5% purity requires 5% extra capital costs [22]. Costs are indexed to 1999 US$ using a Consumer Price Index of 0.816.

[h] FOB price for the ATR is 10.1 million US$ [26].

[i] Calculations were done on basis of m^3_{NTP} flow and necessary reactor height. Given base unit gives a good representation of this.

[j] Calculations were done on basis of m^3_{NTP} flow and necessary reactor height.

[k] Calculated for a fixed bed reactor. No cost data are available for slurry reactors. Main factor used is the amount of CO converted to FT liquids (in MW, HHV based). One catalyst loading is included in these costs.

I) Assuming 1 weight % of Sulphur (excluding N_2) entering the ZnO bed, two guard beds of 3 m^3 are necessary. This will take about 2300 kilos of steel. Using a steel price of US 9.5 US$/kilo, each guardbed will cost US 22,000 US$ (FOB) or US 66,000 US$ installed.

For most components, the actual equipment cost is raised with 53 % [6] to obtain the installed cost (Table 11). For some components and installations, it is assumed, as a rule of thumb, that the FOB price should be tripled to obtain the installed cost (also Table 11). To calculate the total capital requirement (TCR), the *sum* of all the installed costs is once more raised with 35 % (10 % building interest, 10 % project contingency, 10% fees/overheads/profits, 5 % start-up costs) [6].

4.2 Calculation of production costs of FT liquids

Investment costs. Table 12 presents the calculated TCR for the various concepts. As an example, the breakdown of the capital requirement for the IGT pressurised gasification concepts is shown in Figure 6. The pre-treatment, gasification with oxygen and gas cleaning sections account for almost 75 % of total capital investment. The use of an amine treating process for CO_2 removal would add more than 10 % to the total capital investment.

Production costs of FT liquids. The calculated energy efficiency and overall total capital requirement are used to calculate the production costs of pure FT liquids, first without hydrocracking. Key assumptions regarding interest rate and variable costs are presented in Table 13. Straightforward discounting is applied by annuity (interest rate of 10 % and depreciation period of 15 years). A power reimbursement of 0.057 US$/kWh is assumed (includes a premium for green electricity applicable in

Table 12. Total capital requirement for all 367 MW_{th} LHV concepts (MUS$).

Full conversion concepts	**BCL R**	**BCL nt**	**BCL**	**IGT R**	**IGT hg**	**IGT**	**IGT+**	**EP R**	**EP R-nt**	**EP**	**TPS**
Total capital requirement	395	363	312	387	358	339	341	449	417	364	386
Idem without Amine treating	366	334	292	349	320	305	316	396	363	322	344
Once through concepts, 60% and 80% CO conversion		**BCL**	**IGT**	**IGT A**	**IGT+**	**IGT+ A**	**EP**	**TPS**			
Total capital requirement		281	305	338	297	310	325	331			

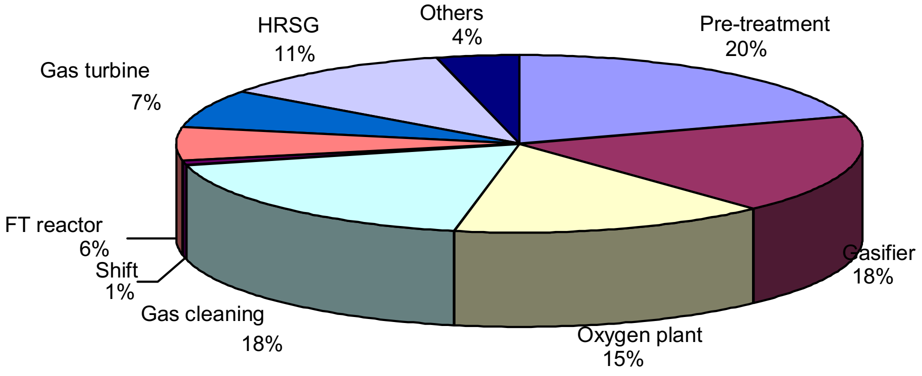

Figure 6. Breakdown of total capital for the IGT 80 % conversion concept (367 MW_{LHV} input).

the Dutch context in year 2000). Electricity from the grid or produced elsewhere on-site costs 0.03 US$/kWh.

Production costs of FT liquids in full conversion concepts are presented in Figure 7, and for the 60 and 80 % conversion once through concepts in Figure 8. The costs vary for the different concepts between 13 and 30 US$/GJ. For both once through and full conversion, the pressurised (IGT) gasifier seems to turn out best. Pressurised systems have a big advantage over atmospheric systems. Also, the co production of electricity by a gas turbine reduces FT liquids production costs.

Table 13. Main assumptions for calculating the FT liquids production costs.

Cost factor		**Input for base scale (367 MW_{LHV})**
Annuity[a]	13.1 % of investment	
Biomass costs[b]	2 US$/GJ (LHV)	21 MUS$
Operational costs		
Maintenance	3 % of investment	
Personnel[c]	0.7 MUS$ at 100 MW_{LHV} input	0.97 MUS$
Dolomite[d]	47.6 US$/tonne	1.22 MUS$
Waste water treatment[e]	0.21 MUS$ at 75 MW_{LHV} input	1.0 MUS$
NaOH consumption[e]	1.3 kUS$/tonne solid NaOH	0.058 MUS$
ZnO consumption [27]	33.3 kUS$/year	Confidential
FT catalyst consumption		Confidential
Insurance	1% of annual depreciation	

[a] Interest rate = 10% and depreciation period = 15 years.
[b] 11,55 GJ_{LHV}/$tonne_{wet}$, 30% moisture, load factor = 8000 hours/year, thus 10.6 PJ_{LHV} biomass.
[c] With a scaling factor of 0.25 [6].
[d] Dolomite (25728 tonne at 367 MW_{LHV}) is used only when a tar cracker is applied [6].
[e] Waste water treatment and NaOH consumption (0.07 kg/tonne dry fuel, or 44800 kg/yr at 367 MW_{LHV}) only if low temperature gas cleaning is applied [6].

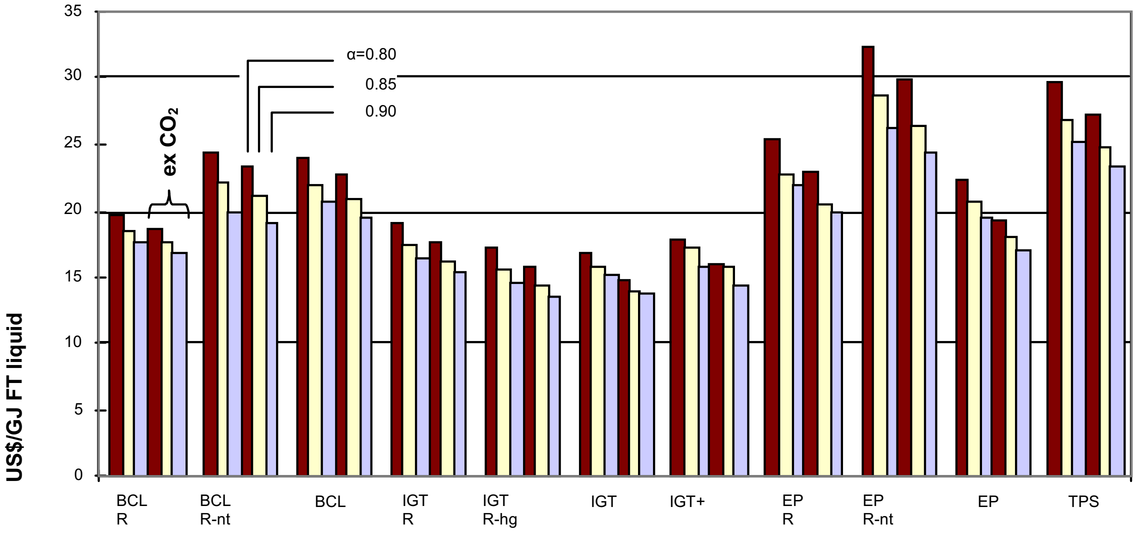

Figure 7. Production costs per GJ_{LHV} FT liquid for the full conversion concepts (40 % conversion per pass and recycling). Electricity yields 0.057 US$/kWh. Input scale is 367 MW_{LHV}. $\alpha = 0.8$ corresponds with $S_{C5+} = 73.7$ %; $\alpha = 0.85$ with $S_{C5+} = 83.5$ %; $\alpha = 0.9$ with $S_{C5+} = 91.1$ %. ex CO_2 represent concepts where CO_2 removal is omitted.

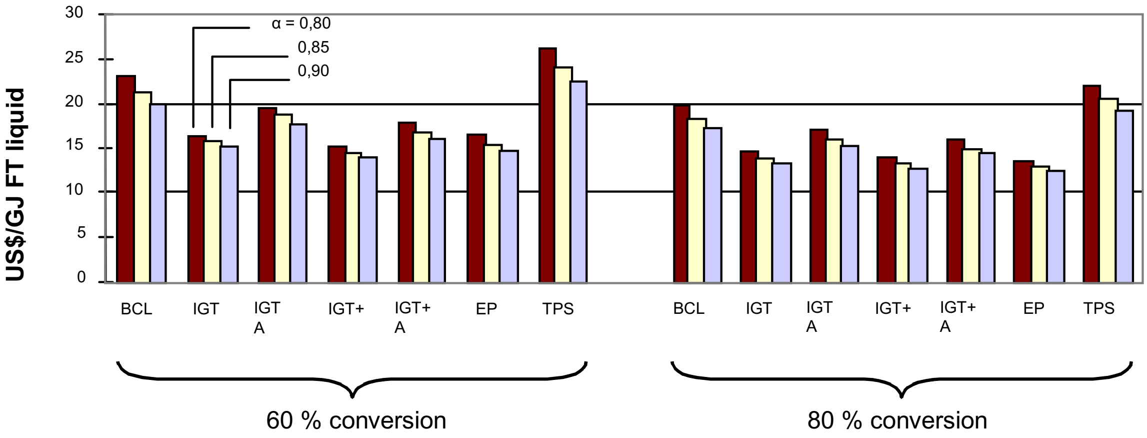

Figure 8. Production costs per GJ_{FT} for the 60 % (left) and 80 % conversion once through concepts. Electricity yields 0.057 US$/kWh. Input scale is 367 MW_{LHV}. α = 0.8 corresponds with S_{C5+} = 73.7 %; α = 0.85 with S_{C5+} = 83.5 %; α = 0.9 with S_{C5+} = 91.1 %.

When all parameters are set at their 'base' value, production costs of FT liquids are 14 US$/GJ. To calculate costs including hydrocracking, a cracking unit is assumed to have a TCR of 8.1 MUS$ per 2000 bbl/day. Using 13.1% annuity the capital costs are thus 0.26 US$/GJ FT liquid. The efficiency of the hydrocracking process is assumed to be 98 % and the H_2 consumption is 176 gram/GJ FT liquid (0.17 US$/GJ FT liquid). Overall costs for hydrocracking become 0.72 US$/GJ FT liquid, being about 5 % on top of the FT liquids production costs. To calculate the production costs of FT diesel, the co-product fractions kerosene and naphtha should be valued to their market price. Since different market prices will also influence the choice for product distribution by changing cracking conditions (refer to Table 2), this is impossible within the present study. Furthermore, the value of biomass derived kerosene and naphtha could be valued very different from fossil fuel derived kerosene and naphtha, because of the CO_2 reduction potential. Assuming that the three products are equally valued by energy content, the FT diesel production cost becomes about 15 US$/GJ. Compare this value to the current production costs of around 5 US$/GJ for diesel.

CO_2 removal has a strong impact on overall production costs. For this reason production costs are also calculated without the amine treating process, for the same efficiencies and selectivities. The heating value of the off gas, however, will be lower due to the higher CO_2 content. This could hinder direct use of the off gas in the gas turbine, but co-firing is still possible.

The concepts cannot be compared directly. Depending on the amount of inert in the FT reactor, C_{5+} selectivity will differ. For example, the EP once through concept may seem slightly better than the IGT once through concept. But the IGT syngas, with little inert, has a higher liquid selectivity than the EP syngas, which contains a large amount of inert.

From Figure 7 it appears that CO_2 removal is an expensive option for all full conversion concepts. However, if this reduction of inert results in an increasing liquid selectivity from 73.7 to 91.9 (α = 0.8 → 0.9), it *does* have a positive effect on production costs. The quantitative impact of the amount of inert on S_{C5+} is not known, however, and needs further detailed study and testing. Furthermore, the captured CO_2 could be sequestered or used, which could be financially valued by society.

When comparing once through with full conversion concepts one should realise that the once through concepts make use of more advanced FT reactors, with higher conversion per pass. The once through options also make use of a co-fired combined cycle that has a higher energy efficiency and benefits from lower specific investment costs.

4.3 Sensitivity analysis and longer term perspectives

Sensitivity analysis of key parameters. Parameters that affect the final FT liquids costs strongest are shown in Table 14, with the potential range between which these parameters may vary. Figure 9 shows the results of a sensitivity analysis over these ranges for the IGT-R concept at S_{C5+} = 83.5 % (in which case 14 % of the total energy output is power). Overall total capital requirement for this concept are about 380 MUS$ and O&M is about 14 MUS$ annually. When power output is high (54 % of the total energy output for the EP full conversion concept at S_{C5+} = 83.5 %), the sensitivity to the electricity value would be much stronger.

Economies of scale have a considerable influence on overall production costs. When scale is between 100 and 400 MW input the overall scaling factor for the entire plant (with respect to overall total investment costs) is approximately 0.74. When capacities go beyond 367 MW, the average scaling factor increases to 0.91: the cost advantage decreases. The results for doing so are shown in Figure 10 for the

Table 14. Main parameters used and ranges for the sensitivity analysis.

Parameter	Value	Range
Biomass costs	2 US$/GJ	2.1 – 8.4
Capital costs	380 MUS$ (varies per concept)	50 – 175 %
Electricity value	0.057 US$/kWh	0.03 – 0.07
Load factor	8000 hours/year	7588 – 8760
Interest rate	10 %	6.25 – 15
Depreciation period	15 years	9.4 – 22.5

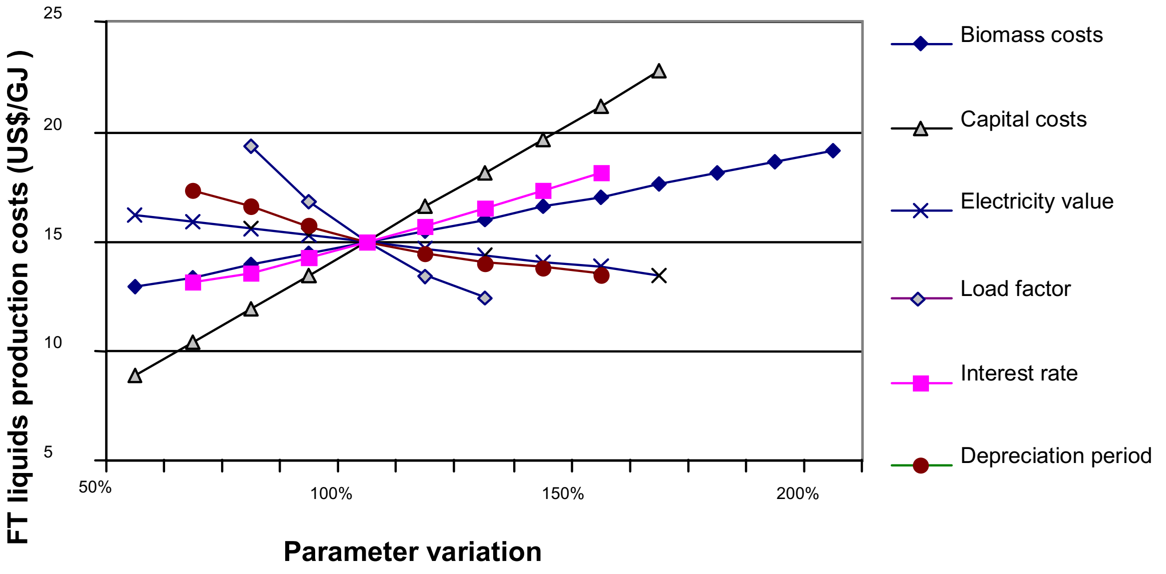

Figure 9. Sensitivity of production costs of the IGT-R concept to used parameters.

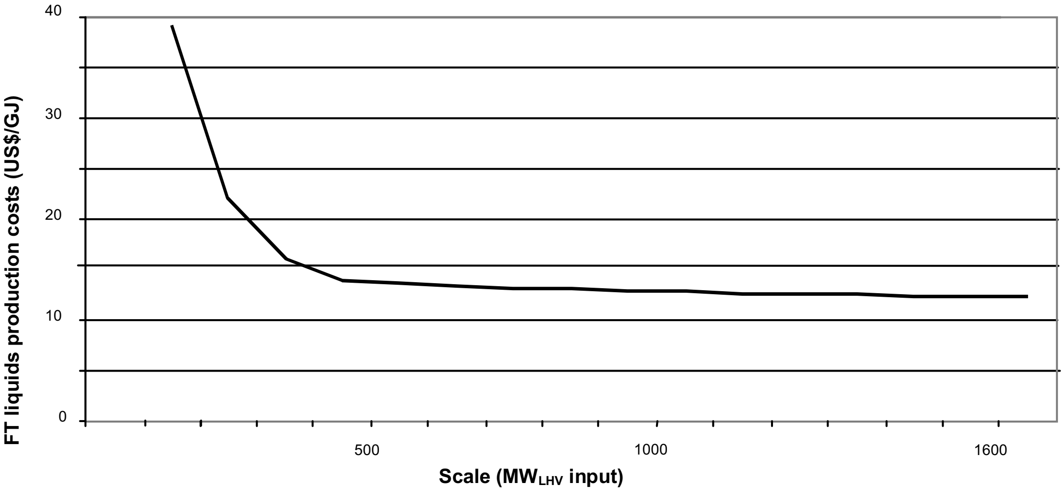

Figure 10. Effect of scale on the production costs of FT liquids; production costs of 14 US$/GJ are assumed at 367 MW input. Biomass feedstock costs are assumed constant here, where in practise biomass costs could slightly increase for larger scales due to higher logistic costs [28].

IGT-R concept. Scale up reduces FT liquids costs from 14 US$/GJ at 367 MW to 12 US$/GJ at 1600 MW, a reduction of 14 %. When the capacity is smaller than 367 MW, costs strongly increase, however, and small-scale production of FT liquids is economically not feasible.

Short and long term perspective. On the longer term various improvements may be feasible. These include: increasing C_{5+} selectivity, increasing scale, lower feedstock costs, reduction of investment and O&M costs through technological learning, and application of hot gas cleaning. For the short and long term the key assumptions made are summarised in Table 15. The type of assumptions for the short and longer term reasonably represent a first commercial plant and a plant that could be built one or two decades from now.

A cost-breakdown for the short and long term is given in Figure 11. Capital costs represent about 50 % of the overall production costs of FT liquids. Therefore, reduction of these capital costs for a third

Table 15. Assumptions for the short and long term.

Short term (first commercial plant)	Long term (third generation)
IGT full conversion (40 % once through, no CO_2 removal) is the best concept	IGT once through 80 % conversion (with high efficiency gas turbine) is the best concept[a)]
Obtainable α = 0.8	Obtainable α=0.9
Scale of the system is 367 MW input	Scale of the system is 1600 MW[b)]
Biomass costs are 2 US$/GJ	Biomass costs are US $2/GJ[c)]
	Technological learning reduces capital costs with 15%[d)]

[a)] Hot gas cleaning has not been modelled for this concept since only a shift reaction is used, which requires some cooling down after the hot gas cleaning. Consequently efficiency advantage would be smaller.

[b)] An overall scaling factor of 0.91 (with respect to total capital requirement) is used.

[c)] In the longer term biomass costs may be lower, but larger scales will increase costs again [28].

[d)] Technological learning can be assumed on longer term. This can be expressed by a progress curve; such curves are defined by a progress ratio. A progress ratio of x (between 0 and 1) implies that each doubling of cumulative output leads to a 1 - x reduction in costs. A progress ratio of 0.9 is used, applied for a third plant built. This results in 15 % lower capital costs [e.g. 6].

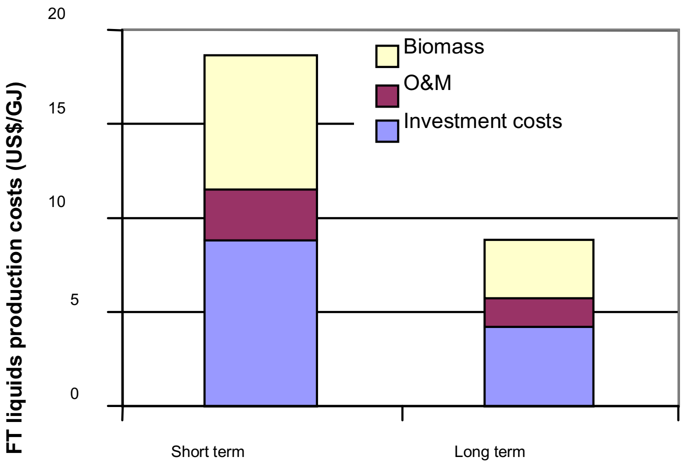

Figure 11. Cost breakdown for production of FT liquids from biomass (excluding hydrocracking) for the short and the long term.

generation plant, by scaling up (12 %) and technological learning (15 %), has a considerable impact on overall production costs. O&M costs decrease proportionally with the reduction of the capital costs. The share of biomass feedstock costs (on themselves assumed to remain constant for the short and long term, see footnotes under Table 15) will decrease per GJ FT liquid due to an increase of overall energy efficiency. Overall energy efficiency will be higher for a third generation plant, due to higher C_{5+} selectivity and higher (once through) CO conversion.

As a result of the reduction in capital costs and biomass costs per GJ FT liquid, production costs of FT liquids could drop from over 14 US$/GJ to 9 US$/GJ. FT diesel then costs 10 MUS$/GJ.

It can be concluded that in the short term pressurised BIG/FT systems have production costs of FT diesel of about 15 US$/GJ. Atmospheric, air blown gasification based systems result in much higher production costs. None of the concepts, either atmospheric or pressurised, have production costs that are competitive with current diesel costs of around 5 US$/GJ. By including a number of improvement options, production costs of FT diesel can drop to around 10 US$/GJ. Obviously, biomass derived FT liquids become more attractive with rising oil prices. Although projections for the future oil price development are highly uncertain, diesel price could go up from 5.5 US$/GJ now to 7 in 2020 [29]. Considering the inherent uncertainties in price estimates as composed from data reported in this study, the fact that not all improvement options have been considered in this study, the longer term economic perspectives for biomass derived hydrocarbons in the transport may still be attractive.

In addition it must be stressed that biomass derived FT liquids differ from fossil diesel in that they contain no contaminants like sulphur and aromatics. They are thus suited for use in Fuel Cell Vehicles when on board reforming (by means of partial oxidation) is applied. Such technology is already demonstrated at present, but requires very clean fuel to avoid poisoning of the fuel cell catalyst. On longer term, this can allow higher vehicle efficiency compared to current diesel fired internal combustion engine vehicles [e.g. 15]. In total, the real value of FT liquids can be considered higher than conventional diesel because of the inherent higher efficiency and much lower emission levels for e.g.

sulphur, soot and other contaminants. The carbon neutral character of bio derived FT liquids is of course a key difference with conventional diesel. Altogether, this may provide a basis for a premium on 'green' biomass derived FT-diesel compared to conventional transport fuels.

In the case of the Netherlands (but similar systems are observed throughout Western Europe), a tax exemption is applied for green electricity varying between 0.024 and 0.038 US$/kWh (year 2000). On energy basis this is equivalent to 6.7 – 10.5 US$/GJ fuel. As fossil diesel production costs are about 5 US$/GJ, depending on oil prices, a fuel tax exemption could make 'green' diesel, naphtha and kerosene' competitive when their production costs are between 11.7 and 15.5 US$/GJ. It is interesting to note that this tax exemption for green energy is lower than the current excise duty on diesel in the Netherlands and many other countries of the European Union. On the long term required 'premiums' could be far lower to make biomass derived FT-fuels cost competitive with conventional diesel per kilometre driven.

5 Conclusions and recommendations

This study presented a broad exploration of the possibilities to produce synthetic hydrocarbons from biomass via gasification and Fischer-Tropsch synthesis. The energy efficiencies and economics of a wide variety of potential conversion system configurations have been evaluated. Systems applying pressurised gasifiers (IGT and EP) have much better overall energy efficiencies (42 – 50 % LHV) than atmospheric systems (33 – 40 %). This is mainly due to the high electricity consumption of the syngas compressors when atmospheric gasifiers are used. High CO conversion, either once through or after recycle of unconverted gas, and high C_{5+} selectivity are important for high overall energy efficiencies.

In the *short term*, production costs of FT diesel, naphtha and kerosene could be about 15 US$/GJ. Capital costs represent about 50 % of the overall production costs of FT liquids. The pre-treatment, gasification (with oxygen) and cold gas cleaning account for almost 75 % of total capital costs. Biomass costs are 30 % of total production costs (assuming a biomass price of 2 US$/GJ), and operation & maintenance about 20 %.

In the *longer term* with large-scale production, high C_{5+} selectivity, high CO conversion and capital cost reductions caused by technological learning, production costs of FT liquids could drop to 9 US$/GJ (FT diesel 10 US$/GJ). Feedstock costs per GJ FT liquid decrease due to an increase of overall energy efficiency, especially because of higher C_{5+} selectivity and higher (once through) CO conversion. When diesel is the desired final product the FT product requires additional hydrocracking: besides 60 % diesel, 15 % naphtha and 25 % kerosene are produced. Hydrocracking will add about 5 % to production costs. 'Green' FT diesel is not competitive with conventional diesel prices, which cost about 0.14 US$/litre or 5 US$/GJ. In the longer term conventional diesel prices may go up due to higher oil prices, but still 'green' FT diesel will probably not be competitive with expected diesel prices (5 - 7 US$/GJ), unless a bonus is provided because of reduced environmental emissions.

There are several uncertainties with respect to the *technology status*. A very critical step in the whole system is gas cleaning. It still has to be proven whether the gas cleaning section is able to meet the strict cleaning requirements for FT synthesis, especially in the case of high temperature concepts having the advantage of high conversion efficiencies. Possibly, amine based CO_2 removal is required for cleaning purposes, thereby raising production costs. The *syngas compositions* as produced by the different gasifiers strongly influence the overall results. Most data are from literature and based on pilot-scale operating experience. The reliability of these data for large-scale gasifiers is not known. Pressurised (oxygen) gasifiers, having most promising economics and advantages of scale, still need further development. At present, only atmospheric air gasifiers, operating at relatively small scale, have proved to be reliable.

Not all possible *concepts* have been investigated. Separating the $C_3 – C_4$ fraction as liquid product could be more advantageous than combusting this fraction in a gas turbine. Also variable gasification temperatures could make other reforming methods possible, like partial oxidation and steam reforming, due to a different C_{2+} content. For the concepts modelled, however, reforming did not result in lower production costs. Decreasing the pressure in the FT reactor will cause a selectivity drop, but on the other hand compression costs will also be lower. Using an iron catalyst could reduce production costs due to an internal shift reaction. This is to be investigated further. In the long term the efficiency of the concepts will be higher if high selectivity can be combined with high CO once through conversion. This could be realised in either fixed bed or slurry reactors. Costs for slurry reactors could be lower than for fixed bed reactors and are very likely to have better economies of scale. Heat integration of the total plant can also be improved. Power generation in the gas turbine will improve if the scale is larger and when on the longer term more efficient turbines will enter the market [6].

This study did include co-firing with natural gas for power generation with combined cycles as an improvement option. Co-feeding natural gas to the FT synthesis was not included but could lead to some efficiency and scale benefits as well.

The overall energy efficiency, a critical parameter in obtaining good economic performance when more expensive cultivated biomass is processed, may be increased by optimised gas turbine technology [e.g. 6] and also by improved selectivity of the FT process applied. With further catalyst and process development higher selectivities may be obtained, improving the net yield of the most valuable commodity of the process: transport fuel.

Technological learning over time and economies of scale were roughly included in the cost projections for longer term, but not investigated in great detail. Especially for pressurised gasification larger scales may be more attractive than projected here and combinations with enriched air gasification (eliminating the expensive oxygen production assumed in this study) may reduce costs further. Some process components may be more suited for up scaling than others, which may lead to different ‘optimal’ technology for different capacities. Altogether, the full technological improvement potential requires further study.

Another key variable is the feedstock costs. (Cultivated) biomass is assumed to cost 2 US$/GJ. At present, this is low for Western Europe, but high compared to Brazil (Eucalyptus). Production of FT liquids in regions where biomass feedstock is cheap (or by using co-products or biomass residues) will positively affect the economics of FT production further.

Considering the inherent uncertainties in cost estimates reported in this study, the fact that not all improvement options for biomass based FT synthesis as discussed above are considered in this study, the longer term perspective for biomass derived hydrocarbons for the transport sector could still be better then projected here. Recommendations for further actions and research are:

- The gas cleaning section needs special attention. Proper data sets of contaminants in the syngas must be made, with high detection accuracy. Deep, hot gas cleaning is promising as such, but will require even more development before sufficient cleaning is guaranteed.
- Pressurised biomass gasification must be developed for large-scale FT plants, but requires demonstration at full-scale.
- For the use of biomass syngas in the FT synthesis, high liquid selectivity is desirable. The FT process (either a fixed bed or a slurry process) needs to be configured to fulfil this need.
- Development of sustainable forestry is necessary to ensure a large enough supply of clean wood. Efforts must be made to create a working biomass market and reduce prices of biomass, from various sources, over time.
- Markets should be explored to determine what the outlets are for FT naphtha and kerosene, and if a ‘green’ fuels premium will be paid for these products, which is particularly relevant for the short term.

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Paper 4

Production of Fischer-Tropsch transportation fuels from biomass; technical options, process analysis and optimisation, and development potential

Contents

Abstract **151**

1 Introduction **152**

2 System components **153**

2.1 Pre-treatment and gasification 153

2.2 Gas cleaning and conditioning 154

2.3 FT synthesis and upgrading, and recycling 157

3 System selection **158**

4 System modelling **160**

4.1 FT modelling 161

4.2 Economic calculations 164

5 System performance **168**

5.1 Physical and economic performance 168

5.2 Towards the ultimate concept 172

6 Conclusion and recommendation **175**

7 Acknowledgements **177**

References **177**

Production of Fischer-Tropsch transportation fuels from biomass; technical options, process analysis and optimisation, and development potential[*]

Abstract

Fischer-Tropsch (FT) diesel derived from biomass via gasification is an attractive clean and carbon neutral transportation fuel, directly usable in the present transport sector. System components necessary for FT diesel production from biomass are analysed and combined to a limited set of promising conversion concepts. The main variations are in gasification pressure, the oxygen or air medium, and in optimisation towards liquid fuels only, or towards the product mix of liquid fuels and electricity. The technical and economic performance is analysed. For this purpose, a dynamic model was built in Aspen Plus, allowing for direct evaluation of the influence of each parameter or device, on investment costs, FT and electricity efficiency and resulting FT diesel costs. FT diesel produced by conventional systems on the short term and at moderate scale, would probably cost 16 €/GJ. On the longer term (large scale, technological learning, and selective catalyst), this could decrease to 9 €/GJ. Biomass Integrated Gasification FT plants can only become economically viable when crude oil price levels rise substantially, or when the environmental benefits of green FT diesel are valued. Green FT diesel also seems 40 – 50 % more expensive than biomass derived methanol or hydrogen, but has clear advantages with respect to applicability to the existing infrastructure and car technology.

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[*] Manuscript, accepted for publication by Energy International Journal. Co-authors: André PC Faaij (Utrecht University), Herman den Uil (ECN), Harold Boerrigter (ECN).

1 Introduction

The transportation sector represents about 86 EJ/yr or 21 % of the world's primary energy consumption in 2000 and this share continues to increase [1]. Transportation fuels are, at present, predominately derived from fossil oil. The present consumption of fossil fuels leads to environmental concerns like the possibility of human induced climatic change through greenhouse gas emission. Concurrently the geographical imbalance between major producers and large consumers raises questions on energy security and global social stability.

Biomass derived transportation fuels can have low lifecycle CO_2 emissions if the biomass feedstock or derivatives are sustainably grown, transported, converted and consumed [2]. Also, they partly reduce local air pollution. Fischer-Tropsch (FT) diesel from biomass via gasification has received growing attention in recent years, as it offers a clean and potentially carbon neutral transportation fuel, directly usable in the present day transportation sector. Furthermore, FT diesel may be suitable as hydrogen source for fuel cell vehicles (FCVs) via on-board reforming, because it is free of fuel cell catalyst poisons, such as sulphur.

Previous research by our group [3] and Arthur D. Little [4] indicated that in Europe, it might be possible to produce FT diesel at 16.7 or 9.1 €[†]/GJ_{HHV} (short or long term). Thus without exemption of excise duty and value added tax (11.6 and 3.5 €/GJ, respectively, in the Netherlands), it cannot compete with low sulphur fossil diesel. For comparison, the US resale price over past five years was 3.7 – 7.5 €/GJ [5]. The indicated study [3] was – though more detailed than previous studies – still of a strong explorative nature, bearing rough assumptions on process equipment performance, ignoring the influence of process conditions on conversion and selectivity in the FT reactor, and rough in the economic analysis. Also, the study listed improvement options to bring the production costs of bio FT diesel to a competitive level. Various essential system components are not commercially available and require development over time. The present study [6] aims to incorporate these improvements and refines the technical and economical analysis. The main system efficiency improvements reside in combined production of FT liquids with power, application of pressurised gasifiers, and combining high selectivity with high conversion in the FT reactor. Since about 75 % of the investment costs are in the pre-treatment, gasification and gas cleaning section, the influence of the gasification pressure and medium (air / oxygen enriched air / pure oxygen) on economy of both gasifier and downstream equipment will be investigated more closely. The scale of the conversion system is expected to be an important factor in the overall economic performance [3; 7-10], but, at the same time, the perspectives of large capacity biomass gasifiers are heavily disputed. Therefore, special attention will be given to the technical perspectives for large-scale pressurised biomass gasification. Previous research also indicated that insight is needed in the sensibility of FT catalysts to contamination in the biomass-derived synthesis gas, in necessary gas cleaning equipment to avoid catalyst poisoning and ageing, and in reactor design [11]. However, many of the ruling uncertainties have been taken away by recent ECN research [12].

[†] All costs in this article are in $€_{2002}$. Energy is expressed on higher heating value basis.

This study investigates the technical and economic performances of bio FT diesel production, the potential production costs in the short, mid and long term, and the appropriate process configurations. Furthermore, the study will indicate RD&D priorities for eventual realisation of the optimal configuration(s), and identify the most probable and successful development paths.

To address these issues, the state of the art technology is extensively surveyed. In Section 2, all necessary system elements are described and their development stage and improvement options indicated. The most promising concepts are presented in a shortlist in Section 3. Section 4 describes the technical modelling of FT diesel production processes and economic calculations. An adjustable computer model is constructed, with an FT reactor in its center that incorporates the influence of different reaction mixtures and reactor conditions on the conversion extent and liquid selectivity. Special attention is given to heat integration within the proposed plants. Results on efficiencies and stream sizes are used for economic evaluations in order to calculated fuel prices. This article is concluded by a discussion of the results (Section 5) and conclusion (Section 6).

2 System components

The basic steps in production of FT liquids [3; 13] are depicted in Figure 1. After pre-treatment, biomass is gasified to produce synthesis gas (biosyngas). This gas is cleaned and the composition is modified to fit the specifications for FT synthesis in the FT reactor. FT off-gas is recycled or combusted to produce electricity. The liquid FT products are treated to yield gasoline (C_5-C_9 hydrocarbons) and/or diesel (C_{10}-C_{20}).

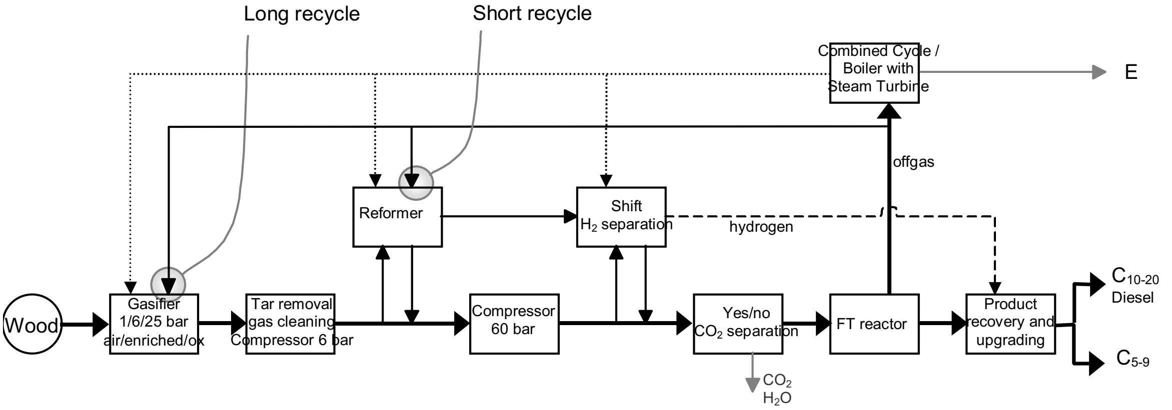

Figure 1. Overview of FT liquids producing plant. By applying or omitting side flows or recycles, different configurations are possible.

2.1 Pre-treatment and gasification

Several pre-treatment units precede gasification, such as biomass feedstock drying and size reduction. Drying requires a significant amount of energy, which can be the FT process heat, or can be extracted from the plant off-gas or from the steam cycle. The efficiency of gasification increases with drier biomass [3; 8; 9; 14]. However, the hydrogen content of the produced syngas decreases, which may be unfavourable for the FT synthesis downstream. Also, drying costs increase quickly below 10 % moist [8]. The resulting effect of a certain extent of drying on the overall plant efficiency/costs is unknown. For Biomass Integrated Gasification Combined Cycle applications 10 – 15 % is the optimum [8]; in this study, drying to a moisture content of 15 % is assumed.

Many gasification methods are available for syngas production. Based on throughput, cost, complexity, and efficiency issues, circulated fluidised bed gasifiers are very suitable for large-scale syngas production. A previous study we did, ruled out direct gasification with air, because the nitrogen dilution strongly increases downstream equipment size and costs [9]. In the present study we focus on direct fired circulated fluidised bed (CFB) gasifiers and attempt to bring the influence of gasification medium and pressure to light. Economies of scale are very important for the technologies considered [8]. Atmospheric CFB gasification up to 300 MW_{th} is technically feasible, but the upper technical limit to capacity has never been explored as generally fuel supply is considered a limiting factor. Pressurised gasification up to 25 bar may have economic advantages: the gasifier may be much smaller per throughput, so that a larger maximum capacity might be possible. Moreover, as no additional compression is required, the temperature can be kept high for high temperature operations downstream. At pressures above 25-30 bar gasifiers need to be constructed more heavily (or the throughput should be kept small) and feeding becomes expensive.

Gasification with oxygen as oxidative medium offers benefits in downstream equipment size, compression energy, and higher partial pressures for relevant components in FT. Pure oxygen is expensive, but oxygen enriched air may combine the advantages in a less expensive medium. In this article, the extremes ‘pure’ air and pure O_2 are investigated, as well as one intermediate medium.

2.2 Gas cleaning and conditioning

The gas produced by gasification contains impurities. Typical are the organic impurities tars and BTX (benzene, toluene, and xylenes), the inorganic impurities NH_3, HCN, H_2S, COS, and HCl, and furthermore volatile metals, dust, and soot. Three possible gas cleaning trains are investigated (Figure 2).

In gasification, larger hydrocarbons are formed, generally categorised as ‘tars’. When condensing, they foul downstream equipment, coat surfaces and enter pores in filters and sorbents. To avoid this, their concentration must be below condensation point at FT synthesis pressure. On the other hand they contain a lot of potential CO and H_2. They should thus preferably be cracked into smaller hydrocarbons. Fluidised beds produce tar at about 10 g/m_{NTP}^3 or 1 – 5 wt% of the biomass feed [12; 15]. BTX, accounting for 0.5 volume % of the syngas, have to be removed prior to the active carbon filters, which otherwise sorb the BTX completely and quickly get filled up [12].

Three methods may be considered for tar removal/cracking: Thermal cracking, catalytic cracking, and scrubbing. At temperatures above 1000 – 1200 °C, tars are destroyed without a catalyst, usually by addition of steam and oxygen, which acts as selective oxidant [15]. Drawbacks are the need for

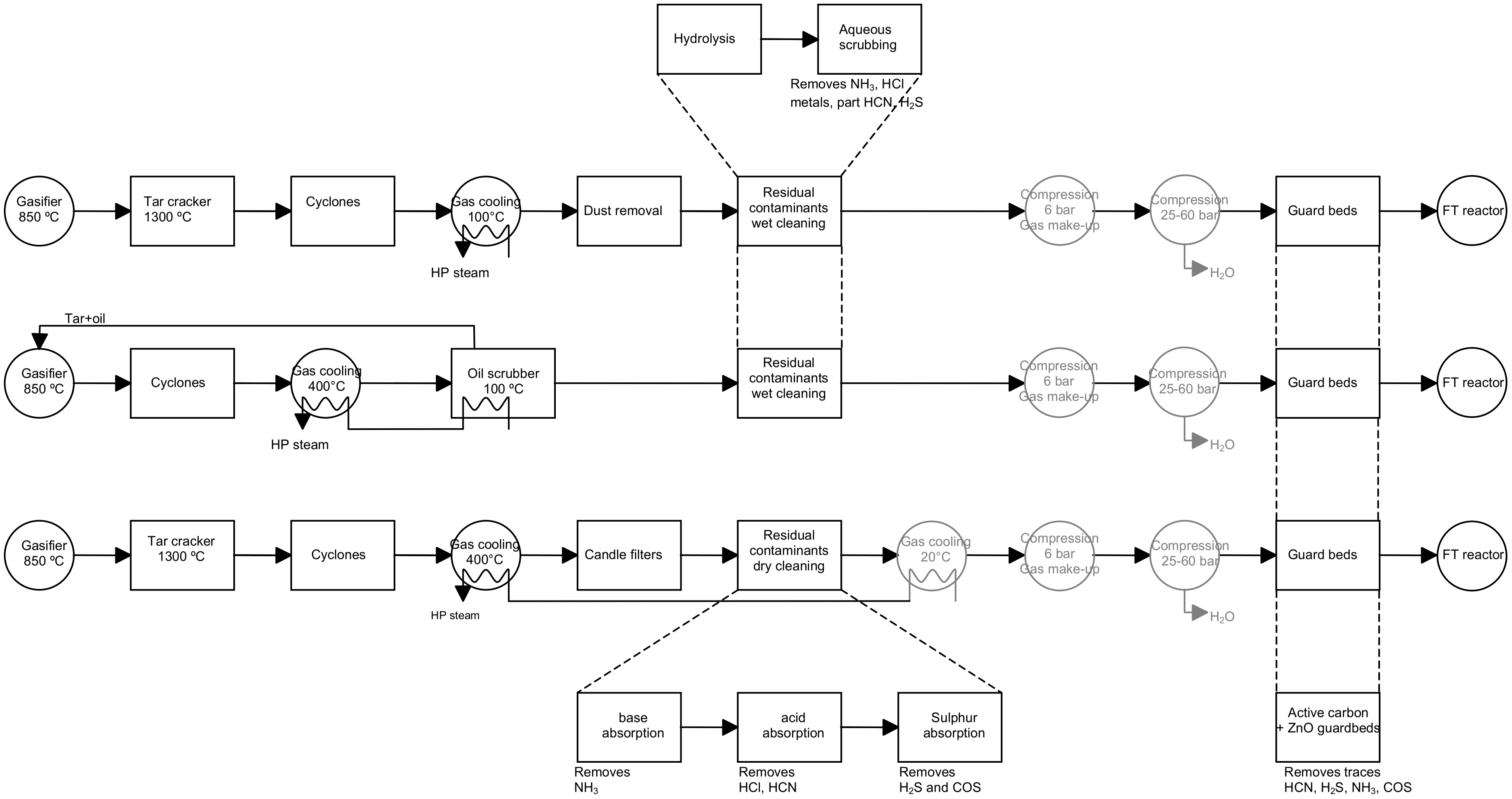

Figure 2. Three gas cleaning trains applied in this study. Top: tar cracking and conventional wet gas cleaning; middle: tar scrubbing and conventional wet gas cleaning; and bottom: Tar cracking and dry gas cleaning.

Table 1. Contaminant specification for cobalt FT synthesis[1], and cleaning effectiveness of wet and dry gas cleaning.

Contaminant	Gas phase Specification	Treatment method & remarks Existing technologies	 Dry gas cleaning[5]
soot (dust, char, ash)	-[2] 0 ppb[3]	Cyclones, metal filters, moving beds, candle filters, bag filters, special soot scrubber[3] Specifications are met.	
alkaline (halide) metals	-[2] < 10 ppb[3]	Active coal bed meets specification[2]	Sorbents Under development
Tar	below dew point at FT pressure[2], catalyst poisoning compounds < 1 ppmV[2]	All tar and BTX: Thermal tar cracker[2], Oil scrubber[2,6], Specifications are met[2].	All tar and BTX: Catalytic tar cracker, other catalytic operations. Under development.
BTX	below dew point at FT pressure[2]		
Halide compounds			
HCl (HBr, HF)	< 10 ppb[2,3]	removed by aqueous scrubber[2,3] active coal bed meets specification[2] absorbed by dolomite in tar cracker (if applicable),	In-bed sorbents or in-stream sorbents < 1 ppm Guardbeds necessary
Nitrogen compounds	total nitrogen <1 ppmV[2] < 20 ppb total N[3]		All nitrogen: catalytic decomposition, combined removal of NH_3/H_2S Selective oxidation Under development
NH_3		removed by aqueous scrubber[2] removed to specification[2,3]	
HCN		active coal bed[2] possibly preceded by hydrolysis to NH_3[3] Specifications are met.	
Sulphur compounds	total sulphur <1 ppmV[2] < 10 ppb total S[3]		All sulphur: In-bed Calcium sorbents Metal oxide sorbents < 20 ppm
H_2S		ZnO guard bed[2] in case of high sulphur loads a special removal step, *e.g.* Claus unit.	
COS		active coal bed[2] possibly preceded by hydrolysis toH_2S[3] Specifications are met.	

[1] Gas turbine specifications are met when FT specifications are.

[2] Boerrigter *et al*. [12].

[3] Assumed by Tijmensen [3].

[4] Bechtel [16].

[5] Hot gas cleaning was practiced in the Värnamo Demonstration plant, Sweden [17]. All data on dry gas cleaning here is based on the extensive research into high temperature gas cleaning by Mitchell [18; 19].

[6] Bergman *et al* [20].

expensive materials, the soot production and the low thermal efficiency. Catalytic cracking (dolomite or Ni based) is best applied in a secondary bed, and avoids the mentioned problems of thermal cracking.

However the technology is not yet fully proven [15] and the catalyst consumption and costs are matters of concern. Tars can also be removed at low temperature by advanced scrubbing with an oil based medium [12]. The tar is subsequently stripped from the oil and reburned in the gasifier. At atmospheric pressures BTX are only partially removed, from about 6 bar BTX are fully removed. The gas enters the scrubber at about 400 °C, which allows high temperature heat exchange before the scrubber.

When the tars and BTX are removed, the other impurities are removed by standard wet gas cleaning technologies or advanced dry gas cleaning technologies. Maximal acceptable contaminant concentrations for the cobalt FT catalyst are summarised in Table 1, together with the effectiveness of two gas-cleaning methods.

In dry gas cleaning, residual contaminations are removed by chemical absorbents at elevated temperature. In the FT situation, hot gas cleaning has little *energy* advantages as the FT reactor operates at 200 – 250 °C, especially when preceeding additional compression is required (efficient compression requires a cold inlet gas). However, dry gas cleaning may have lower operational costs than wet gas cleaning [19].

Early compression reduces the size of gas cleaning equipment, however, sulphur and chloride compounds condense when compressed and they may corrode the compressor. Therefore, intermediate compression to 6 bar takes place after bulk removal and 60 bar compression just before the guardbed.

The syngas can contain a considerable amount of methane and other light hydrocarbons, representing a significant part of the heating value of the gas. Reforming to convert these compounds to CO and H_2 is optional, driven by steam addition over a nickel catalyst. Autothermal reforming is applied in the present study. Compared with steam reforming it is of simpler design, possibly cheaper [21], and able to prevent carbon deposition [3]. The difference in impact of different reforming technologies on H_2/CO ratios in bio-syngas is small.

The syngas produced by CFB gasification (especially at atmospheric pressures) generally has a low H_2/CO ratio. Although FT selectivity is favourable at a low ratio, for high conversion effectively two H_2 molecules are needed per CO. This can be realised by water gas shift with subsequent H_2 separation. Water in the FT reactor acts as inert, thereby decreasing the partial pressure of carbon monoxide and hydrogen. Therefore, prior to the FT reactor the syngas is cooled and water is knocked out.

The syngas from the gasifier contains a considerable amount of CO_2. After reforming or shifting, this amount increases. CO_2 is no reactant in the FT reaction (assuming no shift reaction) but its presence negatively influences the C_{5+} selectivity, at higher concentrations it is removed by Selexol.

2.3 FT synthesis and upgrading, and recycling

In the FT reactor, a high selectivity towards liquid products (longer hydrocarbons) should be combined with a high conversion. To produce long chains, the growth probability α should be close to 1. α is largely catalyst dependent [22], but furthermore depends on temperature, local partial pressures of reactants and inert, and the applied FT technology [3; 23; 24]. For example, α increases with decreasing H_2/CO ratio, decreasing reaction temperature and increasing pressure. Also α is higher with Ru and Co catalysts than with Fe catalysts [25]. Conversion rate and extent depend especially on availability of reactants and the reactor size, via space velocity (inverse residence time) and catalyst density.

There are three main kinds of commercially applied FT reactors: the fixed bed reactor, the fluidised bed reactor and the slurry phase reactor. Comparisons between the reactor types are broadly found [3; 6; 10; 25].

The conversion extent in the FT reactor is limited, depending on catalyst type and reactor size and technology. The reactor product stream thus contains unreacted carbon monoxide and hydrogen in addition to the FT products. The C_{5+} products are easily separated by a condensation step and sent to the recovery and upgrading section. Recovering the very small fraction of C_4 is energy consuming and in general not economic [26].

To maximise the production of FT liquids, the off-gas containing unreacted H_2 and CO and produced lower alkanes can (partly) be recycled to the entrance of the reactor. The recycle can contain a reformer to reconvert C_{1-4} back into syngas, and a shift reactor. Furthermore the FT off-gas may be recycled to the gasifier and subsequent tar cracker, which will work as reformer. This latter option is only sensible when the gasification pressure is the same as the FT pressure. With higher once through conversions in future, the recycling option could become obsolete.

Instead of maximised fuel production, the system can also be optimised towards combined fuel and electricity production. In this case the syngas passes 'once through' the FT reactor. The FT off-gas is not recycled, but completely purged to a combined cycle for electricity production. Investments are assumed to be lower (e.g. no reformer) and presence of inert is less problematic. Co-production of FT liquids and electricity may combine a high overall efficiency with lower investment costs.

The hydrocarbon recovery plant recovers the C_{5+} naphtha, distillate and wax fractions for further processing. First the C_{5-9} fraction is separated from the heavier products. Although the C_{5-9} fraction cannot be applied as green petrol (too low octane number), it may have a high market value in future, as it is suitable for FCVs with onboard reformer, and can serve as raw material in green plastics production. The waxy part C_{10+} of the raw synthesis product is selectively hydro cracked to the desired middle distillate products C_{10-20}. Simultaneously, the product is isomerised to improve the cold flow properties, and subsequently fractionated in a conventional distillation column [22]. In this study the product range comprises both C_{5-9} and C_{10-20}.

3 System selection

Given all the options for gasification, gas cleaning and conditioning, fuel synthesis and upgrading, and power generation, many routes to produce FT Diesel can be imagined (Figure 1). Not all combinations may be sensible from efficiency or economic viewpoint. Besides, not all sensible combinations can be modelled due to time constraints. Of the many considerations [6] the following are most important.

- Both once through and recycle options are evaluated to investigate both optimisation towards FT products, and combined FT and electricity production. The short recycle option basically produces hydrogen from the off-gas instead of from the feed gas, keeping the hydrogen/carbon monoxide ratio below the consumption ratio of these compounds over reactor length and for hydro cracking purposes. Full recycle concepts recycle off-gas to the gasifier, which acts as a reformer for lower alkanes. To avoid accumulation of inert, recycle options are only evaluated for syngas from oxygen blown gasifiers. The full recycle is operated with both gasifier and FT reactor at 25 bar. The recycle is maximally equal to the feed stream.

- The gasifier chosen in this study is a direct fired circulating fluidised bed. Gasification can be at atmospheric, intermediate, or high pressure (*i.e.* 1,6, and 25 bar, respectively). The gasification medium is air, oxygen enriched air, or pure oxygen. The atmospheric air blown gasifier serves as reference point.
- Gas cleaning consists basically of tar removal and residual contaminations removal. Three configurations are considered (Figure 2).
- At atmospheric pressures an oil scrubber would remove BTX to a small extent only. Therefore, after atmospheric gasification only a tar cracker is applied for tar removal. After elevated pressure gasification, both tar cracking and oil scrubbing are evaluated. Heat recovery before tar removal – in the case of oil scrubbing – is possible as long as the temperature remains above the tar condensation point (~ 400 °C).
- After atmospheric gasification the feed gas is compressed to intermediate pressures between tar cracker and residual contaminations removal, and further to FT pressure after gas cleaning. Elevated pressure gasification concepts need only one compression step: after the gas cleaning train.
- Reforming is incorporated in the recycle stream of short recycle options and may be applied in once through options. The reformer is operated at 6 or 25 bar. Higher pressures negatively affect the equilibrium.
- Hydrogen needs to be separated for hydro cracking in the upgrading section. Water gas shift and separation can consecutively be done by a partial shift reactor followed by a pressure swing adsorption unit, or concurrently by a ceramic membrane. Hydrogen separation takes place directly preceding the FT reactor, in a side stream to limit the dimensions of reactors.
- Both fixed bed and slurry FT reactor are analysed.
- The heat produced on various places in the plant is carefully matched with the heat demand at other places in the plant. Leftover heat is used to raise steam of different qualities, which is eventually used for power production through a steam turbine. There are no net heat streams to or from the plant.

These considerations lead to a limited selection of promising biomass to FT conversion concepts, summarised in Table 2. The main variations between these concepts stem from the different gasifier configurations and the gas cleaning train. Some concepts represent the long term (dry cleaning, ceramic membrane), while most are technically feasible at present with state of the art technology.

Table 2. Selected FT liquids production concepts.

	Gasification		Tar removal	Gas cleaning	Reforming Shift and H_2 separation	FT
Once through1) concepts						
I	1 bar	air	tar cracker 1300 °C	HX 100 °C wet cleaning	(ATR reforming) side stream: shift reactor + PSA	60 bar3)
II		enriched2)				
III		oxygen				
IV	6 bar	enriched				
V		oxygen				
VI	25 bar	enriched				
VII		oxygen				
VIII	1 bar	air		HX 400 °C dry cleaning	(ATR reforming) side stream: Ceramic Membrane	
IX		enriched				
X		oxygen				
XI	6 bar	enriched				
XII		oxygen				
XIII	25 bar	enriched				
XIV		oxygen				
XV	6 bar	enriched	HX 400 °C Oil scrubber 100 °C	wet cleaning	(ATR reforming) side stream: shift reactor + PSA	
XVI		oxygen				
XVII	25 bar	enriched				
XVIII		oxygen				
Short recycle1) concepts (to reformer)						
XIX	1 bar	oxygen	tar cracker 1300 °C	HX 100 °C wet cleaning	In recycle: ATR reformer shift reactor + PSA	25 bar
XX	6 bar					
XXI	25 bar					
XXII	1 bar			HX 400 °C dry cleaning	In recycle: ATR reformer ceramic membrane	
XXIII	6 bar					
XXIV	25 bar					
XXV	6 bar		HX 400 °C Oil scrubber 100 °C	wet cleaning	In recycle: ATR reformer shift reactor + PSA	
XXVI	25 bar					
Long recycle1) concepts (to tar cracker)						
XXVII	25 bar	oxygen	tar cracker 1300 °C	HX 100 °C wet cleaning	No reformer side stream: shift reactor + PSA	25 bar
XXVIII				HX 400 °C dry cleaning	No reformer side stream: ceramic membrane	
XXIX			HX 400 °C Oil scrubber 100 °C	wet cleaning	No reformer side stream: shift reactor + PSA	

---

1) See Figure 1 for once through, short and long recycle.

2) Oxygen enriched air consists of 80 % oxygen and 20 % air.

3) For sensitivity analysis the FT reactor pressures in once through concepts may also be 40 bar (after oxygen enriched air firing) or 25 / 40 bar (after oxygen firing).

4 System modelling

To evaluate the chosen configurations, one general flowsheet is built in Aspen Plus [27]. By "opening or closing valves", the different system configurations arise, allowing efficient and quick modelling, and direct comparisons. The gasifier, reformer and gas turbine deliver heat, whereas the dryer, gasifier, reformer, and shift require steam. The supply and demand of heat is added to or drawn from the steam turbine, such that the surplus heat is turned into electricity.

Using a gasifier databank (see Table 3), different gasifiers can be plugged into the plant. The pressure and oxygen purity determine the amount of oxidation medium and steam. Relations from literature determine the yields of methane and heavier components, the other products follow from the water gas shift equilibrium, see footnotes under the Table. It is also possible to choose between differently performing FT reactors (catalysts) and different combined cycles, in aid of the sensitivity analysis.

4.1 FT modelling

Selection and conversion in FT synthesis are a function of feed stream composition, pressure, temperature, catalyst and reactor type and size. Figure 3 depicts how the selectivity and reaction rate are influenced by reactant concentration and ratio and vice versa.

Table 3. Mass flows for circulating fluidised bed gasifier (850 °C), for different media and pressures. Input scale is 440 MW HHV biomass (421 MW LHV). It is assumed that the results may be scaled linear towards other capacities.

Oxidative medium[1)]	**Air**			**Enriched air**			**Oxygen**		
pressure (bar)	**1.3**	**6**	**25**	**1.3**	**6**	**25**	**1.3**	**6**	**25**
In (kg/s)									
Biomass[2)]	26.5	26.5	26.5	26.5	26.5	26.5	26.5	26.5	26.5
Oxidative medium[3)]	32.5	30.2	28.0	8.50	7.76	7.09	6.79	6.21	5.67
Steam[4, 5)]	0	0	0	14.7	13.1	11.6	11.0	9.68	8.49
Out[6)] **(kg/s)**									
H_2	0.661	0.559	0.462	0.808	0.663	0.533	0.791	0.648	0.520
CO	10.7	9.67	8.62	5.84	5.16	4.51	6.74	5.98	5.25
CO_2	13.3	13.6	14.0	19.5	19.1	18.9	18.3	18.0	17.9
H_2O	5.32	5.12	4.88	17.5	15.9	14.5	13.9	12.7	11.5
CH_4	1.76	2.28	2.78	2.32	2.90	3.42	2.34	2.91	3.44
C_2H_4	0.328	0.310	0.29	0.315	0.293	0.272	0.279	0.260	0.242
C_6H_6	0.365	0.346	0.33	0.351	0.326	0.303	0.311	0.289	0.270
N_2	25.3	23.5	21.8	1.77	1.64	1.51	0.271	0.268	0.266
Tar[7)]	0.338	0.338	0.338	0.338	0.338	0.338	0.338	0.338	0.338
Ash	0.428	0.428	0.428	0.428	0.428	0.428	0.428	0.428	0.428
C	0.551	0.551	0.551	0.551	0.551	0.551	0.551	0.551	0.551
Total (kg/s)	59.0	56.7	54.5	49.7	47.4	45.2	44.3	42.4	40.6

[1)] Air at 450 °C, Enriched air (80 % O_2) at 200 °C, oxygen (99.5 % O_2) at 200 °C.

[2)] Feedstock is willow wood: moisture 15 wt %, ash 1.9 wt % d.b., C 49.9 wt % d.a.f., H 6.11 wt % d.a.f., O 42.9 wt % d.a.f. N 0.62 wt % d.a.f. HHV = 19.88 MJ/kg d.a.f. [28].

[3)] It is assumed that carbon conversion is 95 % and heat loss is 2 % of the thermal input. The amount of medium is adjusted such that the desired temperature is reached.

[4)] Steam at 450 °C.

[5)] Steam in oxygen and enriched gasification, replaces the heat capacity of nitrogen in air gasification (to keep product gas at same temperature), the steam is also necessary for fluidisation.

[6)] The water-gas shift reaction is assumed to be at equilibrium. Tar yield from gasification is assumed to be 1.5 wt % of the dry feed, ethane and benzene concentration in the product gas are assumed to be 0.5 and 0.2 vol % respectively [12]. The methane yield is assumed to be in pseudo-equilibrium [29]: $K \cdot \log\left(\frac{p_{H_2}^{1.3}}{p_{CH4}}\right) = 3.0 \times 10^4 \exp\left(\frac{-1.17 \times 10^4}{T}\right)$

[7)] Tar is defined as $C_{14}H_{10}$, represented in Aspen Plus by phenanthrene.

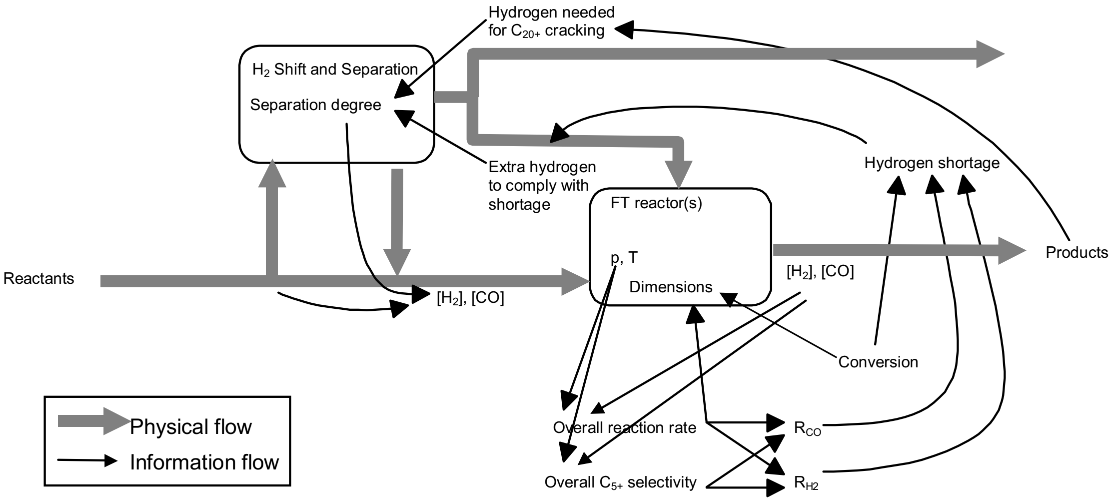

Figure 3. Schema for incorporation of selectivity and conversion in Aspen Plus model. C_{5+} selectivity is influenced by either entrance or exit concentrations, depending on the reactor type: solid bed respectively slurry phase. It is also possible to fix the reactor dimension and calculate the conversion.

Selectivity. In literature, relevant relations for the selectivity depending on these parameters are mostly described in a qualitative way. Selectivity e.g. decreases with hydrogen content, decreases with temperature, and increases with pressure [30]. Assuming that linear relations hold (within boundaries) for selectivity dependency on temperature, ratio, and syngas pressure, the following general relation holds:

$$S_{C5+} = a_1 + a_2 \cdot T + a_3 \cdot \frac{[H_2]}{[CO]} + a_4 \cdot ([H_2] + [CO]) + a_5 \cdot p_{Total} \qquad \text{Equation 1}$$

with S_{C5+} = the mass fraction C_{5+} in the hydrocarbon product
a_i = parameters
$[H_2]$, [CO] = concentrations expressed as fraction of the feed gas
T, p = Temperature (K), pressure (bar)

A least sum of squares fit with (proprietary) data yields Equation 2. The Equation realistically shows the extent and direction of parameter influence; it accords with results presented by Dry [23] on cobalt catalysts.

$$S_{C5+} = 1.7 - 0.0024 \cdot T - 0.088 \cdot \frac{[H_2]}{[CO]} + 0.18 \cdot ([H_2] + [CO]) + 0.0078 \cdot p_{Total} \qquad \text{Equation 2}$$

In the computer model, the selectivity for slurry reactors is calculated from exit conditions, and for solid bed from entrance conditions, change of selectivity along the solid bed is assumed negligible. The product distribution in the relevant range of C_5-C_{20} is assumed to obey the Anderson-Schulz-Flory distribution [30]:

$$\alpha_{Cn} = \alpha^{n-1} \cdot (1-\alpha) \qquad \text{Equation 3}$$

with α_{Cn} = molar fraction C_n in the hydrocarbon product

The relation between α and S_{C5+} can be approached by [6]:

$$\alpha \approx 0.75 - 0.373 \cdot \sqrt{-\log(S_{C5+})} + 0.25 \cdot S_{C5+} \qquad \text{Equation 4}$$

Now the computer model can calculate the C_{1-19} from Equation 3. The fraction C_{4-} is redistributed to 74 mol% C_1, 16 mol% C_2, 6 mol% C_3, and 4 mol% C_4 [31]. C_{20+} is expressed in weight fractions C_{20} and C_{32}, by first calculating the average molecular mass. H_2O, CO_2, and N_2 entering the reactor are considered inert, thus decreasing the partial pressures of the reactants. Unreformed methane, ethane and longer hydrocarbons that enter the FT reactor (after recycle) are inert as well and will not grow to longer chains.

Conversion. The reaction rate determines the conversion extent or the reactor size. Many different relations for reaction rate are found in literature, for different catalysts and sometimes incorporating water inhibition. Yates and Satterfield [32] extensively reviewed kinetic studies on FT synthesis over cobalt and found the following form to describe best the reaction:

$$-R_{CO} = \frac{a \cdot p_{CO} \cdot p_{H_2}}{(1 + b \cdot p_{CO})^2} \qquad \text{Equation 5}$$

with $-R_{CO}$ = Carbon monoxide consumption rate in mol/(s·kg_{cat})
p_{CO}, p_{H2} = partial pressure of CO or H_2, in bar
a, b = kinetic parameters, further defined in Table 4

The computer model calculates the reaction rate in the slurry reactor from the exit conditions. The rate in solid bed reactors is interpolated between rates at reactor entrance and exit. Kinetic parameters used for modelling in the present study are given in Table 4. Multiplying the CO consumption rate with the catalyst density gives the rate per reactor volume; for a desired conversion extent, the reactor volume follows. Table 4 is obtained from different literature sources, which introduces inaccuracy in estimating the reactor size.

Table 4. Kinetic modelling parameters1).

a	(mol/(s·kg_{cat}·bar^2))	$k_0 \cdot \exp\left(\frac{-E_{A,reaction}}{R \cdot T}\right)$	
b	(1/bar)	$k_1 \cdot \exp\left(\frac{-\Delta H_{ads}}{R \cdot T}\right)$	
R	(J/(K·mol))	8.31	
T	(K)		
		Solid bed reactor	**Slurry reactor**
$-E_A$	(kJ/mol)	-68 2)	-115 2)
k_0	(mol/(s·kg_{cat}·bar^2))	$1.2 \cdot 10^5$	$1.0 \cdot 10^{10}$
$-\Delta H_{ads}$	(kJ/mol)	192 3)	192 3)
k_1	(1/bar)	$3.5 \cdot 10^{-23}$	$3.5 \cdot 10^{-23}$
d	(kg_{cat}/$m^3_{reactor}$)	1200 4)	600 4)

1) Yates and Satterfield give values for the kinetic rate constant and adsorption rate constant in Equation 5, for Co catalysts in slurry at 220 and 240°C [32, Table V]. These values may not be used to calculate the E_A and ΔH_{ads}, because of the small temperature range. However, with known E_A and ΔH_{ads} (see notes 2 and 3), k_0 and k_1 can be calculated.

2) The apparent activation energy for the FT reaction depends strongly on diffusion limitation, which in turn depends on catalyst formulation and structure. When the catalyst particles are very small (slurry reactor), the reaction is not diffusion limited and the measured activation energy is about 100 kJ/mol for Co catalysts [32]. Under diffusion limitation (large pellets in solid reactor) the apparent activation energy is about half of the E_A without diffusion limitation (small pellets in slurry reactor). Values from Iglesia *et al.* [33, Table VII].

3) [30, Figure 2.1].

4) Pure catalyst weights 2 kg/l, solid bed has a porosity of ~40 vol %. Concentration in the slurry phase is assumed 30 vol % or 0.6 kg/l [10].

4.2 Economic calculations

Concurrent with the Aspen Plus technical performance calculations, an economic evaluation is carried out for the each concept. The FT Diesel production costs are calculated by dividing the total annual costs of a system by the produced amount of C_{5+} fuel (consisting of two fractions: $C_{5\text{-}9}$ and $C_{10\text{-}20}$). The total annual production costs consist of annual capital investments, operating and maintenance (including maintenance, consumables, labour, waste handling), biomass feedstock and electricity supply / demand (fixed power price). Relevant parameters to calculate these costs and the resulting FT fuel costs, are given in Table 5.

The total capital investment (TCI) is calculated using the *factored estimation* method [6; 9; 34], based on known costs for major equipment as found in literature or given by experts. The uncertainty range of such estimates is up to ± 30 %. Usually the TCI follows from multiplying the total purchased equipment costs by a factor to yield the Inside Battery Limit (ISBL) ‡, adding the costs of the Outside Battery Limit (OSBL) to yield the direct costs, adding indirect costs to yield the fixed capital investment, and finally adding working capital and start-up costs. However, the advised method cannot be followed entirely as base equipment costs in literature may be anywhere between f.o.b.§ and total

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‡ *Inside Battery Limits* only deals with the purchase and installation of process equipment, piping, instrumentation, controls, process buildings, etc. *Outside Battery Limits* includes utilities such as power distribution, steam plants, instrument air systems, sewers, waste water treatment, tankage, cooling towers, control buildings, land, etc.

§ *Free on board*: the price of a traded good after loading onto a ship but before shipping, thus *not* including transportation, insurance, and other costs needed to get a good from one country to another [35]; cost of equipment ready for shipment from supplier.

Table 5. Parameters for the economic evaluation[1)].

Interest rate	10 %
Economical lifetime	15 years
Technical lifetime	25 years
Annual operating costs[2)]	
Maintenance	3 % of Total Capital Investment
Labour	0.5 % at 400 MW_{HHV} input decreasing with scale (scale exponent = 0.25)
Dolomite	8.7 M€ at 400 MW_{HHV} input linear with scale
Gas cleaning	wet: 0.5 % of TCI, dry: 0 %
Insurance	0.1 % of TCI
Biomass[3)]	3 $€_{2002}/GJ_{HHV}$ (short term) - 2 $€_{2002}/GJ_{HHV}$ (long term)
Electricity	0.03 €/kWh
Annual load	8000 hours

[1)] Most of these parameters come from Hamelinck *et al.* [9].

[2)] A breakdown of operational costs for BIG/CC was applied by Faaij *et al.* [8]: Annual maintenance costs are 3 % of Total Capital Investment. Operation costs depend on labour [36], catalyst and chemicals consumption, residual streams disposal, and insurance. Labour costs decrease with scale (Equation 6) and are 0.5 % of the TCI for a 400 MWHHV input. Dolomite costs are 50 €/tonne, dolomite use is 0.3 kg/kg clean dry wood. At 400 MW HHV input the annual biomass throughput is 0.56 Mtonne. Wet cleaning costs are in waste water treatment and NaOH consumption, while dry residuals are assumed suitable for construction. Insurance is 1 % of annual depreciation.

[3)] Prices of delivered cultivated energy crops and forest thinnings in Western Europe amount currently 3-5 $€/GJ_{HHV}$, United States 2.3-3.3 $€/GJ_{HHV}$, and at some Latin American locations even lower costs are possible 1.2 - 2 $€/GJ_{HHV}$ [37; 38]. Improved crops and production systems are expected to bring the biomass price to the 1.6-2.1 $€/GJ_{HHV}$ range [United States, 39].

installed capital, and often a specific overall installation factor is given to yield the TCI of that piece of equipment.

Therefore, the capital investment is first calculated for each separate component, and later all components are added up to yield the TCI. The component's cost depends on its size (which follows from the Aspen Plus modelling), by scaling from a known scale and cost (see Table 6):

$$C_1 = C_0 \cdot \left(\frac{S_1}{S_0} \right)^R \qquad \text{Equation 6}$$

with S_0, S_1 = known scale, respectively the desired scale
C_0, C_1 = cost at the known scale, respectively at the desired scale
R = power scaling factor, or scale exponent

Various system components have a maximum size, above which multiple units will be placed in parallel. Hence the influence of economies of scale on the total system costs decreases. After scaling, the TCI of each component is found by multiplying the scaled base cost by an installation factor.

The given numbers must be used carefully, as the base equipment costs in literature are often ill defined. Also the applied percentages for additional costs differ greatly between studies. Of course these percentages depend on the specific location: OSBL costs in industrial areas may be lower since the necessary infrastructure is readily available, a Middle East location will differ greatly from a Western Europe location and OSBL are higher. All the factors to calculate TCI from purchased equipment are

Table 6. Costs of system components in M€$_{2002}$ 1).

Unit	Base cost	Scale Factor	Base Scale	Overall installation factor 25)	Maximum Size 26)
Pre-treatment 2)					
Conveyers 3)	0.41	0.8	33.5 wet tonne/hr	2.0	110
grinding 3)	0.48	0.6	33.5 wet tonne/hr	2.0	110
storage 3)	1.16	0.65	33.5 wet tonne/hr	2.0	110
dryer 3)	8.5	0.8	33.5 wet tonne/hr	2.0	110
iron removal 3)	0.43	0.7	33.5 wet tonne/hr	2.0	110
feeding system 3,4)	0.48	1	33.5 wet tonne/hr	2.0	110
Gasification System					
CFB gasifier 5)	44.3	0.7	68.8 dry tonne/hr	1.69	75
Air separation unit 99.5 % O_2 6)	27.9	0.75	576 tonne/day O_2	1.3	3200
Oxygen compressor 7)	18.1	0.85	13.2 MW$_e$	1.86	-
Gas Cleaning					
Tar cracker 3)	3.6	0.7	34.2 m^3/s gas	2.0	52
Oil scrubber 8)	1.64	0.7	14.7 m^3_{NTP}/s gas	1	-
Cyclones 3)	3.0	0.7	34.2 m^3/s gas	2.0	180
High-temperature heat exchanger 9)	8.1	0.6	138.1 MW$_{th}$	1.84	-
Particle filters 3)	1.9	0.65	12.1 m^3/s gas	2.0	64
Scrubbers 3)	3.0	0.7	12.1 m^3/s gas	2.0	64
Dry gas cleaning 10)	35.8	1.0	74.1 m^3/s gas	1.86	-
Guard beds (ZnO + active C) 11)	0.024	1.0	8.0 m^3_{NTP}/s gas	3	
Syngas Processing					
Compressor 12)	12.9	0.85	13.2 MW$_e$	1.86	-
Autothermal reformer 13)	31.1	0.6	100 m^3_{NTP}/s	2.3	-
Shift reactor 14)	12.2	0.65	8819 kmol CO+H_2/hr	1.81	-
PSA units A+B 15)	32.6	0.7	9600 kmol feed/hr	1.69	-
Ceramic membrane 16)	25.2	0.8	17 tonne H_2/hr	1	-
Selexol CO_2 removal 17)	63.0	0.7	9909 kmol CO_2/hr	1	-
FT Production					
Solid bed gas phase FT 60 bar 18)	25.3	1	208 m^3	1.30	-
Slurry phase FT 60 bar 19)	36.5	0.72	362 m^3	1	365
Product upgrading 20)	233	0.7	286 m^3_{FT}/hr	1	
Power Isle 21)					
Gas turbine + HRSG 3,22)	22.0	0.7	26.3 MW$_e$	2.0	-
Steam turbine + steam system 3,23)	5.9	0.7	10.3 MW$_e$	2.0	-
Expansion turbine 24)	5.0	0.7	10.3 MW$_e$	2.0	-

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1) Annual GDP deflation up to 1994 is determined from OECD [40] numbers. Average annual GDP deflation after 1994 is assumed to be 2.5 % for the US, 3.0 % for the EU. Cost numbers of Dutch origin are assumed to be dependent on the EU market, therefore EU GDP deflators are used. 1 €$_{2002}$ = 0.88 US\$$_{2002}$.

2) Total pre-treatment approximately sums up to a base cost of 11.46 M€$_{2002}$ at a base scale of 33.5 tonne wet/hour with an R factor of 0.79.

3) Based on first generation BIG/CC installations. Faaij *et al*. [41] evaluated a 29 MW$_e$ BIG/CC installation (input 9.30 kg dry wood/s, produces 10.55 m_{NTP}^3 fuel gas/s) using vendor quotes. When a range is given, the higher values are used [8]. The scale factors stem from Faaij *et al*. [8].

4) Two double screw feeders with rotary valves [41].

5) Direct costs are 29.74 MUS\$$_{1991}$ for a 1650 dry tonne/day input IGT gasifier, R = 0.7 [2]. Maximum input is 400 MW$_{th}$ HHV [3].

6) Various cost numbers with a large distribution are found in literature [2; 3; 16; 42; 43]. Here the cost number from Tijmensen [3] is used because it bests fits the real oxygen cost price [44], see discussion in [6] : Direct costs are 27.9 M€$_{2002}$ for a base scale of 576 tpd oxygen of 99.5 % purity, 1 bar, the overall installation factor is constant 30 %, R = 0.75. It is assumed that oxygen of 95 % purity is 5 % cheaper; oxygen of 80 % purity for the scale considered here, will always be mixed from 95% O_2 and air, and thus barely has cost advantages. The highest maximum scale found to date is 3200 tpd [Shell Bintulu, see 11]

7) Because of explosion risk, oxygen is compressed in a non-lubricated compressor, which is 1.4 times more expensive than lubricated compressors [45].

8) OLGA technology [20]. Cost numbers are assumed by the authors.

9) High temperature heat exchangers following the gasifier and (in some concepts) at other locations are modelled as HRSG's, raising steam of 86 bar/510 °C. A 39.2 kg steam/s unit (i.e. transferring 138.1 MW$_{th}$) costs 6.33 MUS\$$_{1997}$ f.o.b., overall

installation factor is 1.84 [42], which is assumed to consist of 22 % added direct costs (vary with scale) and 50 % added indirect costs (constant), see note 25.

10) Tijmensen [3] assumes the f.o.b. price for Hot Gas Cleaning equipment to be 30 $\text{MUS\$}_{2000}$ for a 400 MW$_{th}$ HHV input. This equals 74.1 m^3/s from a BCL gasifier (863 °C, 1.2 bar). There is no effect of scaling. The installation factor is taken from Faaij *et al*. [8] but for 400 MW$_{th}$ input scale (decreased added direct costs), see footnote 25.

11) For 427 MW$_{th}$ LHV biomass input concepts, Tijmensen [3] assumes that 1 wt% of the fuel S enters the ZnO bed, and two guard beds of 3 m^3 are required. These beds consist of 2300 kg steel, at a steel price 20 Dfl$_{2000}$/kg, so that each guard bed will cost 50.000 Dfl$_{2000}$ f.o.b., or 150.000 Dfl$_{2000}$ installed. Tijmensens 70 MW$_{th}$ LHV (13 dry tonne/hour biomass, assumed gas yield 100 kmol/dry tonne) corresponds to a local gas flow of 8.0 m$^3_{NTP}$/s.

12) Katofsky [21] assumes compressors to cost 700 $\text{US\$}_{1993}$ per required kW$_{mech}$, with an installation factor of 2.1. The relation used in the present study stems from the compressor manufacturer Sulzer quoted by Tijmensen [3]. At the indicated base-scale, total installed costs are about 15 % higher than assumed by Katofsky. Multiple compressors, for fuel gas, recycle streams, or hydrogen, are considered as separate units. The installation factor is taken from Faaij *et al*. [8] but for 400 MW$_{th}$ input scale (decreased added direct costs), see note 25.

13) Hamelinck [9] based on Katofsky [21] assumes 4.7 $\text{MUS\$}_{2001}$ with an overall installation factor of 2.3 for an 1390 kmol total/hour (8.8 m$^3_{NTP}$/s) unit, R=0.6, where Bechtel [46] gives a total capital investment of 20.52 $\text{MUS\$}_{1992}$ for an ATR unit of 364.7 MMscfd (120 m$^3_{NTP}$/s) gas feed, which is almost twice as costly. Here we assume the average between the two: 31.1 M€ total capital investment for a 100 m$^3_{NTP}$/s unit, R=0.6. The installation factor is assumed to consist of variable direct costs (53 % in base case) and fixed indirect costs (50 %), see note 25.

14) Cost numbers from Williams [2] are used: 9.02 $\text{MU\$}_{1995}$ for an 8819 kmol CO+H$_2$/hr reactor, overall installation factor is 1.81 and R = 0.65. Hendriks [47] writes that a reactor of 350.000 m$^3_{NTP}$/hr total gas (CO+H$_2$ is 93.3 %, this gives 14.6 Mmol/hr CO+H$_2$) costs 30 $\text{MUS\$}_{1994}$ installed (no R given) which corresponds with the line from Williams. Hendriks also gives an annual (sulphur tolerant) catalyst use of 0.7 $\text{MUS\$}_{1994}$, but considers this to be part of the operational costs. Numbers from Tijmensen [3] are considered too low: 0.95 MDfl$_{2000}$ for direct costs plus engineering (add 30 % to obtain total capital investment) for a 2400 kmol/hr unit, R = 0.6.

15) PSA units (excluding the recycle compressor) cost 23 $\text{MUS\$}_{1993}$ for a 9600 kmol feed/hour throughput, R=0.7 [21].

16) Membrane costs 68 $\text{US\$}_{1997}$/(kW/bar), but these costs are only 9 % of the total installed cost for a Hydrogen Separation Device. Investment costs stem from Parsons I&TG [48]. The economies of scale of the membrane surface are low because the required surface area is proportional to the throughput, this slightly influences the overall R factor of the complete HSD.

17) Costs for CO$_2$ removal through Selexol amounts 14.3 $\text{MUS\$}_{1993}$ fob (overall installation factor is 1.87) for an 810 kmol CO$_2$/hr unit, R=0.7 [21] up to 44 $\text{MUS\$}_{1994}$ installed for a 9909 kmol CO$_2$/hour unit [47]. The value from Hendriks is assumed to be right, since his research into CO$_2$ removal is comprehensive.

18) FT Gas phase reactor operating at 40 bar costs 35 MDfl$_{2000}$ (direct costs + engineering) for a production of 100 MW$_{FT}$ HHV (CO conversion = 150 mol/s, and $-R_{CO} = 6.03\cdot10^{-4}$ mol/s·kg$_{cat}$, thus reactor volume is 208 m^3 CNH), R = 1, there is no maximum scale, add 30 % to obtain total capital investment [3]. For operation at 60 bar and same volume, costs increase by 50 % (reactor with thin wall: thickness linear with pressure).

19) 19.995 $\text{MUS\$}_{1996}$ ISBL or 34.6 $\text{MUS\$}_{1996}$ "Total Costs" (includes offsites, HOService/Fee and contingency) here assumed to be total capital investment, for a FT slurry reactor train of *three* 362 m^3 reactors at 25.2 bar, each yielding 295 m$^3_{FT}$/day or 131 MW$_{FT}$ [16], the installation costs of one 362 m^3 reactor thus is 15.2 M€$_{2002}$. Operation at 60 bar makes the installation 2.4 times more expensive (see note 18).

20) Tijmensen [3] assumes that a hydrocracking unit has installed costs (=Total capital investment, CNH) of 17.05 MDfl$_{2000}$ (8.22 M€$_{2002}$) per 2000 bbl/day or 11352 GJ$_{C5+}$ per day (13.2 m^3/hr), but does not specify further, his source is confidential. According to Bechtel [16] a wax hydrocracking plant of 475 Mlbs/hr of FT diesel+gasoline product (286 m$_{FT}^3$/hr) costs 71.5 $\text{MUS\$}_{1996}$ total capital investment, furthermore a distillate hydrotreater (29.1 $\text{MUS\$}_{1996}$), a naphtha hydrotreater (9.8), a naphtha reforming plant (49.3), a C$_4$ isomerisation plant (7.2), a C$_5$/C$_6$ isomerisation plant (11.2), a C$_3$/C$_4$/C$_5$ alkylation plant (41.2), and a saturated gas plant (9.2) are necessary, thus summing up to 233 M€$_{2002}$. Thus upgrading costs in Bechtel's study are 1.3 - 3.3 (at R = 1 - 0.7) times Tijmensen's values. Here we assume that the Bechtel numbers are right.

21) For indication: A complete Combined Cycle amounts about 830 $\text{US\$}_{1997}$ per installed kW$_e$. Quoted from [49] by [50].

22) Scaled on Gas Turbine size.

23) Steam system consists of water and steam system, steam turbine, condenser and cooling. Scaled on Steam Turbine size.

24) Expansion turbine costs are assumed to be the same as steam turbine costs (without steam system).

25) Overall installation factor. Includes auxiliary equipment and installation labour, engineering and contingencies. Unless other values are given by literature, the overall installation factor is set 1.995 for a 70 MW$_{th}$ scale [8]. This value is based on 33% added *direct costs* to installed equipment (instrumentation and control 5%, buildings 1.5% grid connections 5%, site preparation 0.5%, civil works 10%, electronics 7%, and piping 4%) and 50 % added *indirect costs* to direct costs (engineering 15%, building interest 10%, project contingency 10%, fees/overheads/profits 10%, start-up costs 5%). The added *direct costs* part of the overall installation factor decreases with scale (average R-factor is 0.82).

26) Maximum sizes from Tijmensen *et al*. [3], unless indicated otherwise.

expected to be very dependent on scale, as ISBL relatively decreases with scale because of increasing process integration and OSBL decreases because certain off-sites have a fixed same scale for both small and large plants. However, no literature was found on this subject, therefore the influence of scale and/or location on the overall installation factor is not incorporated in the present study. Only where the composition of the installation factor (in direct, indirect, working capital and start-up) is known, the direct costs part of the overall installation factor is decreased with scale, assuming a scale exponent R = 0.82 [8].

5 System performance

5.1 Physical and economic performance

In Section 3 the most promising FT concepts were listed. Here we analyse the technical and economical performance of these concepts, in order to find the best configuration(s). Results for the first group of 7 once through concepts are given in Figure 4. These concepts apply a tar cracker and conventional wet gas cleaning, but no reforming. Obviously, the production of FT fuel becomes cheaper with increasing gasification pressure; the influence of oxygen purity seems negligible. This coheres with both a decreasing total capital investment (Figure 4) and an increasing plant efficiency (Figure 5). Capital costs decrease for a large part because of decreasing gas volume in the cleaning section. The extra costs for oxygen compression are more than outweighed by reduced compression costs downstream. The plant efficiency increases because at the higher partial reactant pressure, the selectivity for the desired product is higher (at same fixed conversion) even though at atmospheric and air gasification significantly more $CO+H_2$ is produced (Table 3). Moreover, the internal power needs decrease with higher gasification pressure and less nitrogen dilution. Slurry phase and solid bed reactors perform the same. Therefore, in the rest of this analysis we only discuss solid bed reactors.

Total Capital Investment for the 400 MW_{HHV} input plant (or 168 $MW_{FT,HHV}$ output) is about 286 M€. This is about 1.7 times as much as reported by Bechtel [26], the difference especially resides in the gasifier (our choice for pressurised CFB vs. Bechtel's atmospheric indirect) and the air separation unit required by the present concepts. Bechtel further has a reduced pretreatment (only drying) and their TCI does not include the combined cycle. Without the pre-treatment and gasification section and with a reduced gas cleaning section, the TCI would be 93 M€ or 0.55 k€/kW_{FT}. This can be compared with the TCI for grass roots natural gas fed FT plants, reported to amount 300 $MUS\$_{1997}$ for a capacity of 10,000 bbl/day (740 $MW_{FT,HHV}$) or 521 M€/GW_{FT} [51], and 750 $MUS\$_{2001}$ for a 2.22 $GW_{FT,HHV}$ plant or 394 M€/GW_{FT} [52]. Taking into account the scale disadvantage, the TCI found thus complies with literature values.

The capital build up for the extremes of these configurations is given in Figure 6. The gas cleaning costs, which make out a large part in atmospheric air fired concepts, are strongly reduced in pressurised oxygen fired concepts, where – naturally – the costs for oxygen production and compression play an important role. The shares of costs for pre-treatment, gasifier and power generation grow, but remain equally distributed.

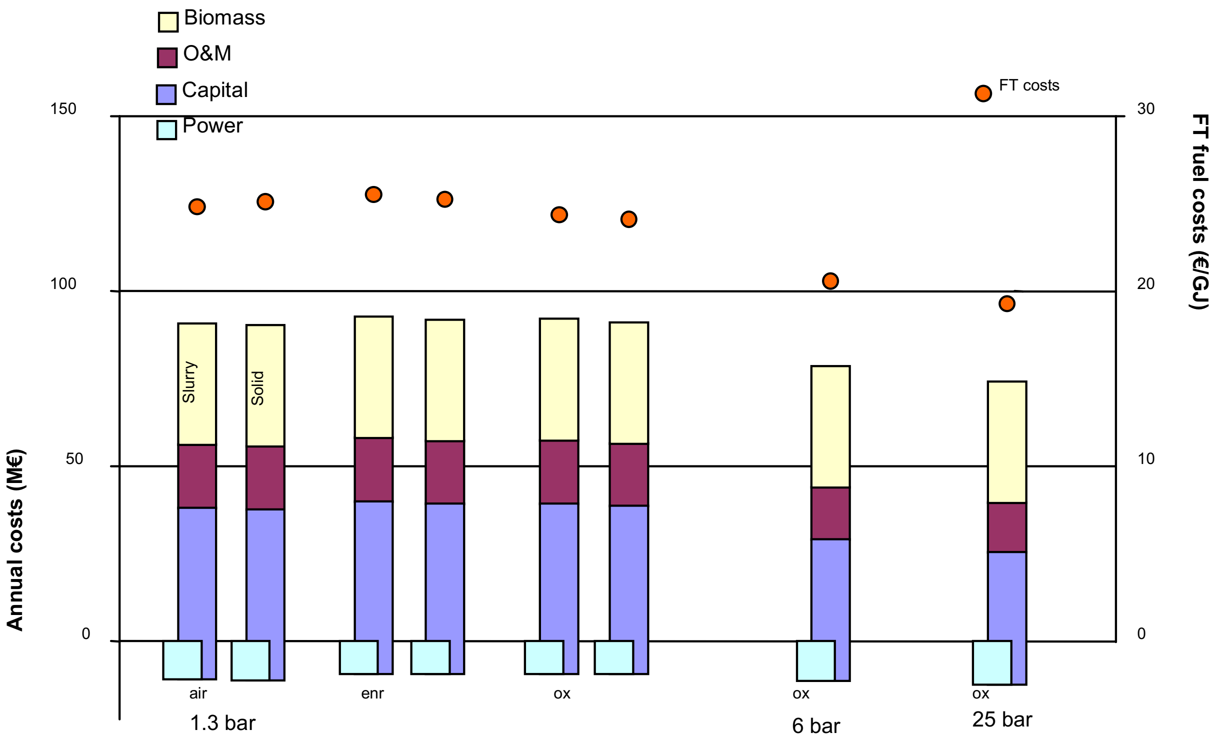

Figure 4. Annual costs (bars) and resulting FT liquids production costs (bullets), for different gasification configurations (air, enr and ox stand for air blown, oxygen enriched air blown and oxygen blown). Fixed settings: 400 MW_{HHV} input scale, tar cracker, wet gas cleaning, no reforming, once through FT, 70 % conversion.

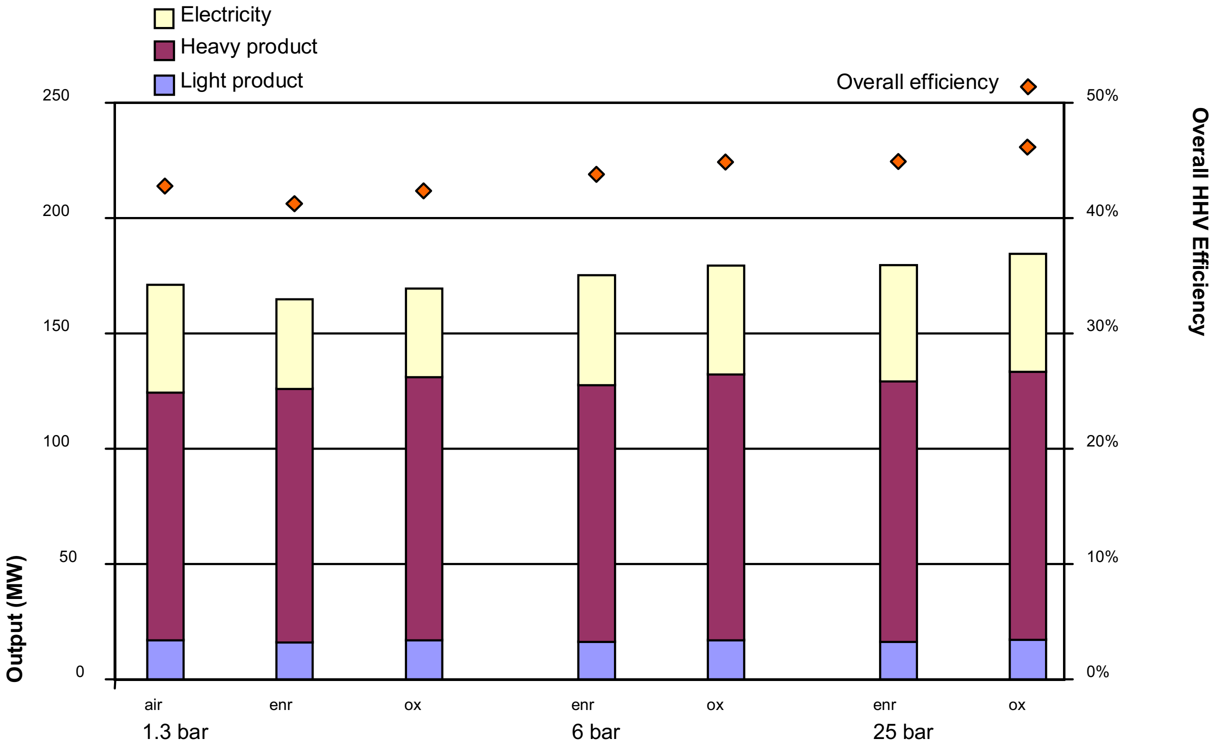

Figure 5. Product selectivity (bars) at 400 MW_{HHV} input, and resulting overall plant fuel+electricity efficiency (diamonds) for different gasifier configurations. Fixed settings: tar cracker, wet gas cleaning, no reforming, once through FT, 70 % conversion.

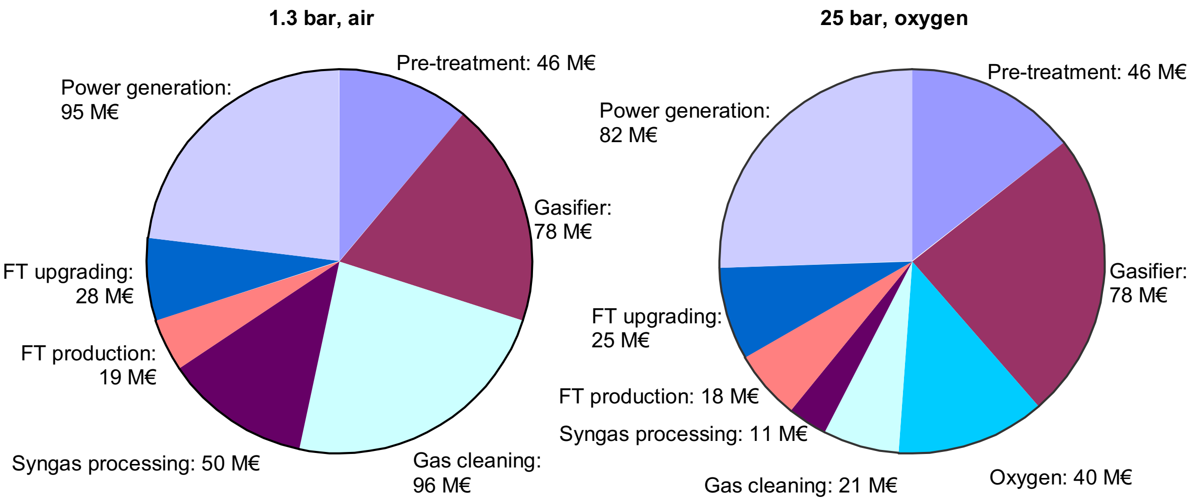

Figure 6. Capital break down for two once through concepts: based on air fired atmospheric gasifier (left) and based on oxygen fired 25 bar gasifier (right). All costs are in M€. Fixed settings: 400 MW_{HHV} input, tar cracker, wet gas cleaning, no reforming, solid bed FT, 70 % conversion.

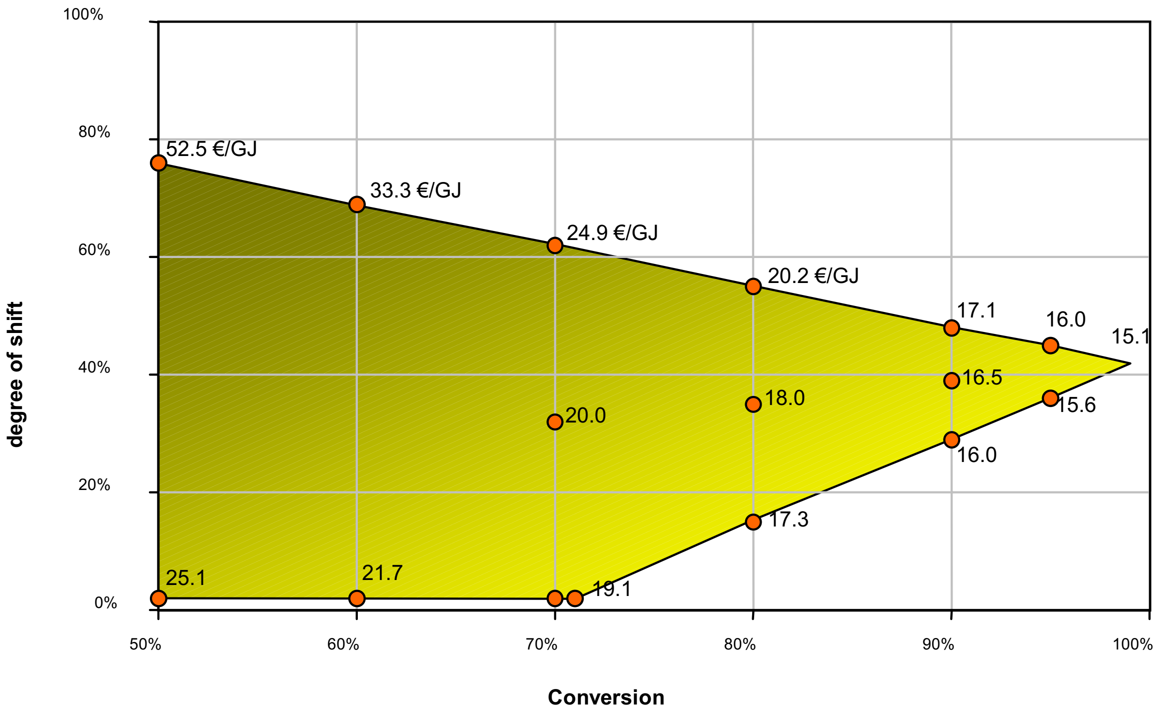

Figure 7. FT liquids production costs (numbers near bullets) as a function of conversion extent and degree of shift for 25 bar oxygen gasification at 400 MW_{HHV} input. Below area is hydrogen shortage, above area is carbon monoxide shortage. Fixed settings: tar cracker, wet gas cleaning, no reforming, once through FT.

So far, the $CO+H_2$ conversion in the FT reactor was set to 70 %. A higher conversion can be realised by a larger reactor, leading to higher capital costs. Since the capital costs of the FT reactor are not crucial for the total capital investment, a higher conversion leads to lower FT diesel production costs, see Figure 7.

According to the graph, the lowest costs are reached at total conversion. This stems from the assumption that reaction rate is constant (the average between entrance and exit reaction speed) over the length of the reactor. Of course, as CO and H_2 disappear during the conversion, the reaction rate decreases rapidly when approaching full conversion, leading to exponential reactor size increase for the last converted reactants. We assume that, at 90 % conversion, the entrance-exit-averaged reaction rate is just still representative for the volume-averaged reaction speed. A 400 MW_{HHV} input system applying conventional technologies can thus produce FT diesel at 16 €/GJ, which compares to the 15 €/GJ

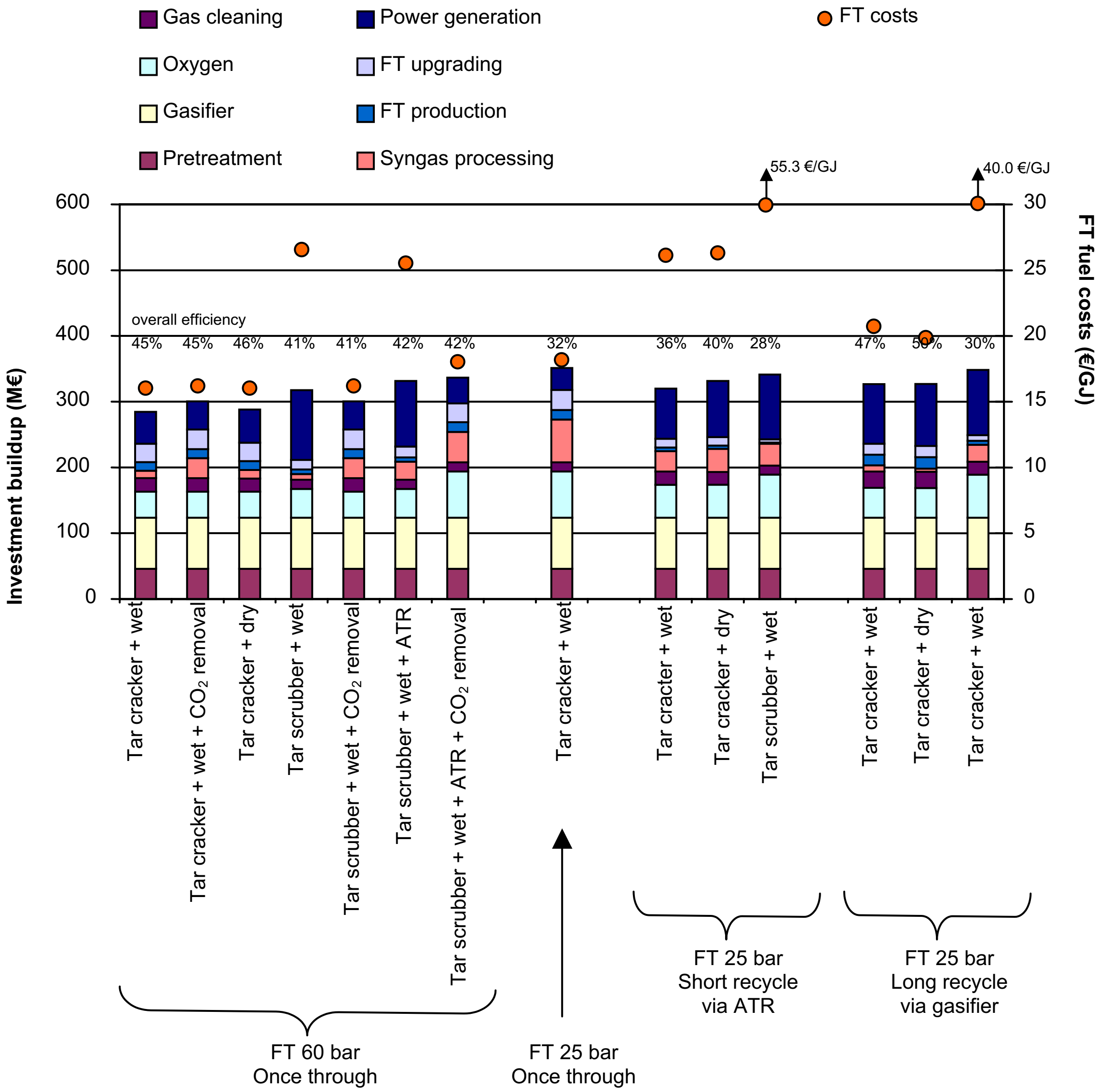

Figure 8. Variations on the key concept (25 bar oxygen gasification and 90 % $CO+H_2$ conversion), applying other gas cleaning trains, reforming and/or CO_2 removal, once through or recycle mode. The overall (fuel+electrical) HHV efficiency is indicated.

found by Tijmensen *et al.* [3] for a similar system at the same scale.

The CO_2 volume fraction before the FT reactor is about 30 %. Removing this fraction improves both selectivity and efficiency, but due to the accompanying increase in investment this does not result in lower product costs (see Figure 8). The efficiency for dry gas cleaned concepts is slightly higher than wet cleaned concepts, because the concurrent applied ceramic membrane (refer to Table 2) shifts more effectively (since product is taken away) than the traditional shift reactor. This is outweighed by a slightly higher capital investment, such that the resulting fuel price is the same. The oil scrubber effectively returns BTX and tars to the gasifier where they are cracked, but much energy remains in methane, ethane and ethane, so that the FT yield is low. The technical performance of concepts with oil scrubber improves when adding a reformer. However, this turns out a more expensive combination than a single tar cracker. Overall efficiencies for the economic best performing systems are 40 – 45 % on HHV basis.

The recycle is operated at 70 % conversion of $CO+H_2$ per pass, 50 % of the off-gas is recycled so that overall conversion is about 90 %, like in the once through concepts. The FT pressure is decreased to 25 bar to avoid very large pressure leaps. Nevertheless the continuous temperature leaps and product leaps (syngas partly to $C_{1\text{-}4}$ product, and than back to syngas) lead to a both low selectivity and low overall efficiency, and the capital costs are high. The FT 25 bar once through concept is included to show that the lower operation pressure contributes only marginally to the increased FT liquids production costs.

The influence of scale (see Figure 9) is examined for both wet and dry cleaned concepts. It is found that at large scale, FT diesel production costs can drop below 14 €/GJ. The resulting overall scale factor for both concepts is 0.78 (Tijmensen *et al.* assumed 0.91 [3]). The efficiency advantage of dry cleaned concepts does not lead to lower costs at larger scales, compared to wet cleaned concepts.

The uncertainty of this economic evaluation mainly follows from the ± 30 % uncertainty on capital investment, which also influences the Operating and Maintenance costs. Figure 10 shows that this results in ± 18 % uncertainty in the FT fuel costs. The biomass price (± 30 % uncertainty) brings ± 13 % uncertainty in the FT fuel cost. The calculated efficiency has a much lower uncertainty (assumed to be ± 5 %) acting on the produced amount of FT fuel and electricity. Its influence on the FT fuel cost is small. Also uncertainties in the electricity price (± 10 %) have little influence on the FT product.

5.2 Towards the ultimate concept

In the previous paragraph it was shown that, at present, FT diesel could be produced at 16.1 €/GJ by applying conventional technologies, at the reasonable scale of 400 MW_{th} input. This is still about four times the production costs of low sulphur fossil diesel, which are about 3.7 – 7.6 €/GJ [53]. However, as fuel production costs are only about one-third of the sales price, FT diesel from biomass can become competitive with fossil diesel when the fuel is partly exempted from excise duty and value added tax (11.6 €/GJ and 3.5 €/GJ, respectively, in the Netherlands), to value the environmental benefits.

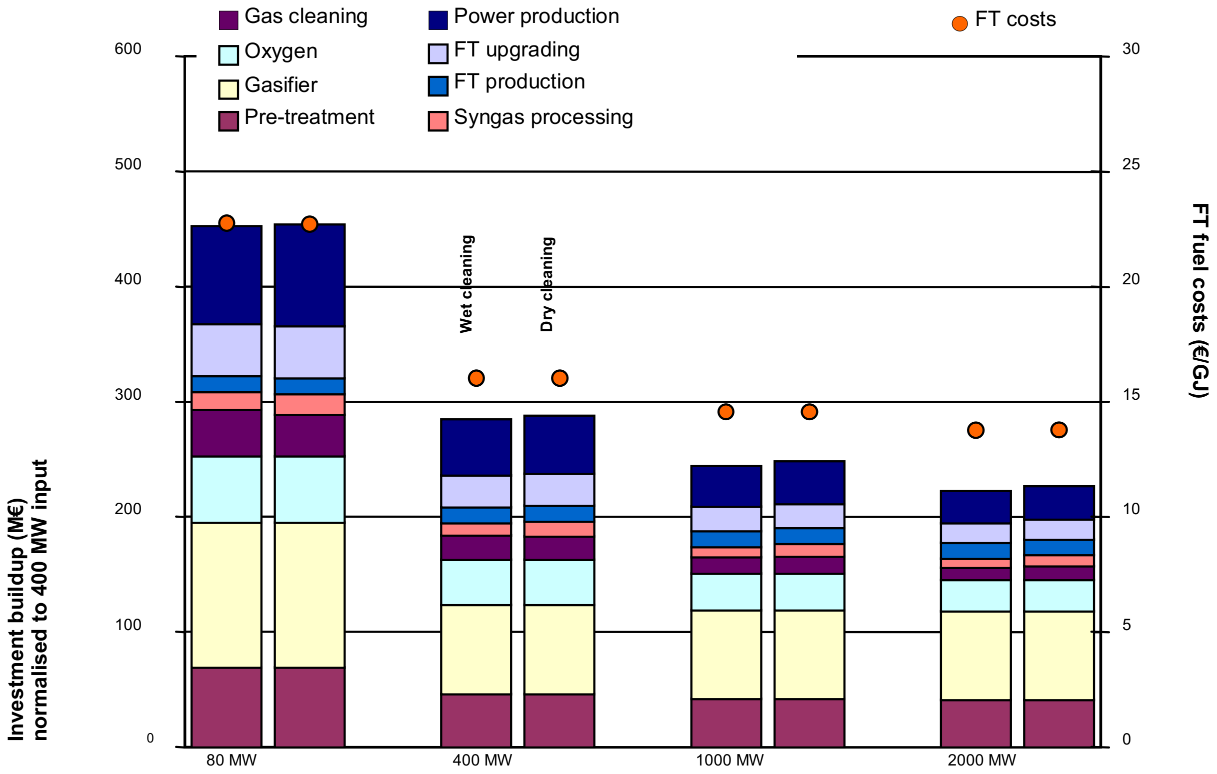

Figure 9. Influence of scale on relative investment costs (bars, left axis), and resulting FT liquids production costs (bullets, right axis) for wet and dry cleaned concepts. To compare the investments of 80, 1000 and 2000 MW with 400 MW, they have been multiplied by 5, 0.4 and 0.2 respectively. Fixed: 25 bar oxygen fired gasification, Tar cracker, no reforming, once through 90 % conversion.

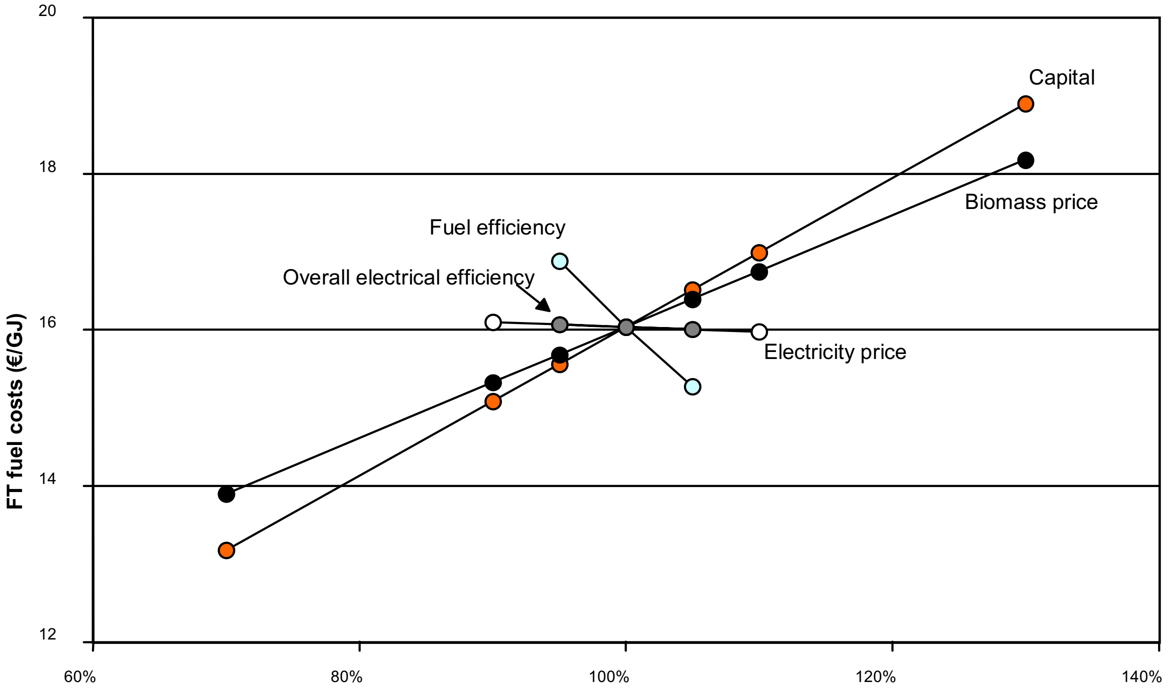

Figure 10. Influence of variation of uncertain parameters on FT liquid production costs. Fixed: 400 MW_{HHV} input system, 25 bar oxygen fired gasification, Tar cracker, wet cleaning, no reforming, once through 90 % conversion.

Looking at the longer term (~15 years), improvements are foreseen. Production at a larger scale may bring the FT fuel costs to below 14 €/GJ (recall Figure 9). Furthermore the biomass price may drop to 2 €/GJ, decreasing the FT production costs to 14 €/GJ (at 400 MW_{th} input) or even 11.5 €/GJ (2000 MW_{th}). Many technical developments may also improve the overall economic system performance, several of these are summarised in Table 7. Technological learning could reduce capital costs with 15 % for the third plant built [54], which reduces the FT costs with about 9 %. A major breakthrough would be the development of catalysts that would selectively produce a desired product fraction, *e.g.* the diesel fraction, avoiding the large amount of longer chain hydrocarbons that have to be cracked down again. If this pure 'diesel' selectivity could be realised, the upgrading section could be much cheaper or may even be omitted, implying another 5 – 10 % total capital cost reduction. Development of such catalysts is speculative. The collective effect of large scale, technological learning, and selective catalyst may bring the FT diesel costs down to 9 €/GJ.

It is concluded that green FT diesel at present is only competitive with fossil diesel when produced at a scale of at least 100 – 200 MW_{th} input, and when exempted from taxes. The competitiveness of future large scale FT production strongly depends on the evolvement of an international biomass market, with

Table 7. Options that may improve the technical and economical performance.

	Near term	Long term perspective	Impact
Oxygen production		Cryogenic separation more efficient, energy use could decrease from 300 kWh_e/tonne to theoretical minimum of 30 kWh_e/tonne [55]	Limited
		Gas turbine driven membrane separation cheaper1), however, availability of this technology is unknown	Significant
Gasification	High carbon conversion or recover energy from char in ash		2-3 %-points on gasification efficiency
Tar removal		Catalytic tar cracker operating at lower temperature: 900 °C instead of 1300 °C	High
		Physical/chemical instead of thermal tar removal technologies.	High
FT catalyst	α Slightly higher		Limited
		Selectivity towards desired product C_{10-20}, e.g. by sterical hindered catalysts, thus decreasing upgrading costs	Very high, but uncertain
Gas turbine		General efficiency increases slightly	Limited
Steam cycle		Higher steam pressures	Limited
Process integration	Improve heat integration		Limited
	Find the optimum between energy consumption in biomass drying and gain in gasification step		Limited
	Avoid compressor intercooling by balancing compression before and after gasification		Unknown
Scale		Larger scale decreases capital, labour and may be feedstock costs	Limited
Technological learning		More experience with large scale gasification, logistics, more automation	High

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1) Costs may eventually be 0.038 $€_{2002}/m_{NTP}^3$, while cryogenic separation costs about 0.05 $€_{2002}/m_{ntp}^3$ [55].

large quantities of biomass against low prices, and ongoing catalyst development. The technology roadmap towards such future is quite straightforward, as on the longer term the technology will basically be similar to that applied in this study. It can be expected that single devices may improve their efficiency. Also there may be continuing competition between dry gas cleaning and wet gas cleaning, and between solid bed FT reactors and slurry reactors.

Two other biofuels, methanol and hydrogen, were previously studied by Hamelinck and Faaij [9]. Produced at similar scale and biomass price (400 MW_{th}, available technologies, 3 $US\$_{2001}/GJ$) these fuels cost 12 and 11 €/GJ respectively. On the longer term, at large scale (2000 MW_{th}), with advanced technologies, reckoning with technological learning and at a biomass price of 2 US$/GJ, these costs decrease to 8.3 and 7.5 €/GJ. Although the recalled study cannot directly be compared with the present one, it seems that FT diesel is more expensive both on the short and long term. Besides the energy costs of the different alternative transportation fuels, mineral oil price and cost reductions in conventional diesel production, other factors determine whether FT diesel becomes a success or not, such as its applicability in existing fuel distribution systems, improvements in ICEV (internal combustion engine vehicle) technology, the availability of the FCV and realisation of its potentially highly efficient fuel use.

6 Conclusion and recommendation

The technological and economic performances of all promising biomass to Fischer-Tropsch liquids conversion concepts have been evaluated using a dynamic Aspen Plus flowsheet. The influence of each parameter or device, on investment costs, liquid product and electricity efficiency and resulting FT diesel costs have been made visible. Compared with our previous study [3], the present study offers flexibility to explore these influences. Moreover, the performance of system components is modelled more precise, heat, hydrogen and oxygen are integrated in the flowsheet if applicable, and the economic analysis is done with greater detail.

Total Capital Investment or TCI for a 400 MW_{HHV} input plant (or 168 $MW_{FT,HHV}$), consisting of a 25 bar oxygen fired gasifier, followed by a tar cracker and wet gas cleaning and a solid bed FT reactor with 70 % once through conversion, is about 286 M€. A high conversion (90 % of $CO+H_2$) can be realised in once through mode by application of a large reactor. This results in higher capital costs, but since the capital costs of the FT reactor are not crucial for the total capital investment, higher conversion leads to lower FT diesel production costs. Overall efficiencies for the best performing systems are 40 – 45 % on HHV basis. With such systems FT liquids can be produced at 16 €/GJ.

System variations to the above named concept give the following insights:

- Removing the 30 volume % CO_2 fraction prior to the FT reactor improves both selectivity and efficiency, but due to the accompanying increase in investment this does not result in lower product costs.

- The efficiency for the dry gas cleaned concepts is slightly higher than wet cleaned concepts as less steam is needed. This is outweighed by a slightly higher capital investment, such that the resulting fuel price is the same.
- The oil scrubber effectively returns BTX and tars to the gasifier where they are cracked, but much energy remains in methane, ethane and ethane, resulting in lower FT yields. The performance of concepts with oil scrubber improves when adding a reformer, however, this turns out to be a more expensive combination than a single tar cracker.
- Application of a recycle loop instead of once through does not decrease the production costs: The continuous temperature leaps and product leaps lead to both a low selectivity and a low overall efficiency. Furthermore, the capital costs are high.

The FT liquids production costs for the short term are still about two to four times the production costs for fossil diesel. Fossil diesel costs strongly depend on the oil price, and could go up. The biomass FT diesel could partly be exempted from excise duty and tax to value its environmental benefits. The combined effect may make FT diesel from biomass competitive with fossil diesel.

In the longer term (~15 years), more cost improvements are foreseen. The combined effects of larger production scale (2000 MW_{th}) and cheaper biomass (2 €/GJ) may bring the FT fuel production costs to 11.5 €/GJ. The technical and economical performance may be improved by for example developments in oxygen production and gasification, application of catalytic tar cracking instead of thermal, and increased selectivity towards the desired product in FT synthesis. These technical developments combined with technological learning (15 % cost reduction in third plant) bring the FT diesel costs to 9 €/GJ. These values for short and long term are comparable with the costs found by Tijmensen *et al.* However, his short term biomass price assumed was 2 €/GJ (vs. 3 €/GJ in present study), and his long term scale was 1600 MW_{th} input (vs 2000 MW_{th}), so that effectively the short term FT costs here, are lower than found by Tijmensen *et al.*, and the long term costs somewhat higher.

The quality of the analysis depends partly on the model and partly on the economic parameters. The influence of the model is most visible in the FT reaction, where selectivity and conversion rate are defined as a function of reactant concentration, ratio, pressure and temperature. The relations for selectivity and reaction rate, as presented in Equations 2 and 5 and Table 4, are strong simplifications of reality. However, they make the influence of process conditions on performance quite visible and thus add an important improvement compared to previous work in this field. The availability of more public accessible data of recent experiments, would improve the model.

The factored estimation method yields investment costs with an uncertainty range of ± 30 %, resulting in ± 3 $€/GJ_{FT}$. For a few devices (esp. gasifier), insight in the factors that determine the investments (influence of pressure, oxygen use) could not be underpinned, this makes it difficult to discuss improvement options and scale-up possibilities. The poor availability of economic data in general, often leads to repeatedly quoting old quotes, and losing the meaning of the economic numbers.

It is recommended that further research focuses on the further development of oxygen blown pressurised biomass gasification, especially the larger scale applications, and that more insight is acquired in the technical maximum of such gasifiers and the accompanying costs. Furthermore, continuing fundamental catalyst development is necessary to couple a high selectivity (of only the desired product) to a high conversion extent. And eventually the competitiveness of future large scale FT production strongly depends on the evolvement of an international biomass market, with large quantities of biomass against low prices.

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Paper 5

Prospects for ethanol from lignocellulosic biomass: Techno-economic performance as development progresses

Contents

Abstract **183**

1 Introduction **184**

2 Ethanol production **186**

2.1 Feedstock 186

2.2 Key conversion technology 187

2.3 Process integration 197

3 System selection **200**

4 Modelling **201**

4.1 Technical modelling 201

4.2 Economic analysis 203

5 Techno-economic performance **206**

6 Discussion and conclusions **211**

7 Acknowledgements **213**

References **213**

Prospects for ethanol from lignocellulosic biomass: Techno-economic performance as development progresses[*]

Abstract

Short and long term prospects of ethanol production from lignocellulosic biomass are evaluated. State of the art technology and technology currently under development are reviewed. Promising conversion concepts for short term and future are defined. Their technical performance was analysed, and results were used for economic evaluations. It is concluded that biomass to ethanol conversion efficiencies on short – long term may be 35 – 48 % HHV, electricity is co produced from the not fermentable lignin, so that overall efficiencies are 50 – 68 %. Capital investments represent about 40 % of the ethanol production costs. The ethanol producing part (pre-treatment, hydrolysis, fermentation and upgrading) accounts for about half of these investments; the rest is mainly in generation of steam and electricity. Development of more efficient pre-treatment technology, integration of several microbiological conversions into fewer reactors, and increasing ethanol production capacity may decrease specific investments for ethanol producing plants from currently 2.1 k€/kW$_{HHV}$ (at 400 MW$_{HHV}$ input) to ultimately 0.9 k€/kW ethanol production (2 GW). A combined effect of higher hydrolysis fermentation efficiency, lower specific capital investments, increase of scale and cheaper biomass feedstock costs (from 3 to 2 €/GJ), could bring the ethanol production costs from 22 €/GJ$_{HHV}$ at the short term (5 years) via 13 €/GJ (10-15 years) down to 8.7 €/GJ in 20 years or more, if further development of the technology would be stimulated.

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1 Introduction

Disadvantages of fossil fuel derived transportation fuels (greenhouse gas emissions, pollution, resource depletion, unbalanced supply-demand relations) are strongly reduced or even absent with bio transportation fuels [1; 2]. Of all biofuels, ethanol is already produced on a fair scale (about 14 - 26 Mtonne worldwide [3; 4]), and is easily applicable in present day internal combustion engine vehicles (ICEVs), as mixing with gasoline is possible. Ethanol is already commonly used in a 10 % ethanol / 90 % gasoline blend. Adapted ICEVs can use a blend of 85 % ethanol / 15 % gasoline (E85) or even 95 % ethanol (E95). Ethanol addition increases octane and reduce CO, VOC and particulate emissions of gasoline. And, via on board reforming to hydrogen, ethanol is also suitable for use in future fuel cell vehicles (FCVs). Those vehicles are supposed to have about double the current ICEV fuel efficiency [5]. About 90 % of all ethanol is derived from sugar or starch crops by fermentation; the rest is produced synthetically [4]. The bulk of the production and consumption is located in Brazil and the USA. Some 67 % of the production is used to fuel cars; the rest is used in food industry. Fermentation technologies for sugar and starch crops are very well developed, but have certain limits: These crops have a high value for food application, and their sugar yield per hectare is very low compared with the most prevalent forms of sugar in nature: cellulose and hemicellulose. Suitable processes for lignocellulosic biomass therefore have room for much further development: a bigger crop variety can be employed, a larger portion of these crops can be converted, and hence larger scales and lower costs are possible.

Lignocellulosic biomass can be converted to ethanol by hydrolysis and subsequent fermentation. Also thermo chemical processes can be used to produce ethanol: gasification followed either by fermentation, or by a catalysed reaction [6]; however, these are not considered here. Hydrolysis fermentation of lignocellulose is much more complicated than just fermentation of sugar. In hydrolysis the cellulosic part of the biomass is converted to sugars, and fermentation converts these sugars to ethanol. To increase the yield of hydrolysis, a pre-treatment step is needed that softens the biomass and breaks down cell structures to a large extent. Especially the pre-treatment and hydrolysis sections allow for many process configurations: Present pre-treatment processes are primarily chemically catalysed, but both economic and environmental arguments drive the development of physical pre-treatments. The pre-treatment technology chosen affects the yield of both pre-treatment and subsequent process steps. Acid reliant hydrolysis processes have been used for many decades, but have environmental consequences (esp. large amount of gypsum to dispose of). Enzymatic processes under development are supposed to have roughly equal costs today, but are more environmentally sound, and the costs can be decreased further. Therefore, most studies focus on enzymatic hydrolysis [5]. The fermentation step, on its turn, does not yet convert all sugars with equal success. Future overall performance depends strongly on development of cheaper and more efficient micro organisms and enzymes for fermentation. Newer micro organisms may also allow for combining more process steps in one vessel, such as fermentation of different sugars, and enzyme production [5]. Lastly, the biomass composition in (hemi) cellulose and sugar influences the ethanol yield.

The present production costs of ethanol show a broad range: Ethanol from sugar cane in Brazil costs 10 – 12 €/GJ[†] [7; 8], while in Europe and the USA sugar or starch derived ethanol cost 16.2 €/GJ$_{LHV}$ [9] to 23 €/GJ [10]. Projected present cellulosic ethanol production costs in Europe lie between 34 and 45 €/GJ [9; 11], and in the USA between 15 and 19 €/GJ [12; 13] (feedstock 1.8 €/GJ). Future costs are projected 4.5 - 10 €/GJ by Lynd *et al.* [14], 6 – 8 €/GJ by de Boer and den Uil, and 10 – 11 €/GJ (within 10 years) by Wooley *et al.* [15], approaching production cost of methanol (via biomass gasification) and fossil bulk fuels [5; 16; 17]. In order to obtain the projected economic performance, a number of technological breakthroughs is required. Cost reductions reside in improving individual process steps, far-reaching process integration, enzyme cost reduction, and using the remaining lignin to generate electricity.

Part of the studies mentioned above is old and need to be updated to include recent developments. Wooley *et al.* [15] present a very detailed analysis on one (currently) feasible configuration, but do not give indications for future performance. Lynd [5] and Lynd *et al.* [14] give an excellent overview of 1996 state of the art technology, assess the many different process parts and the merge of conversion steps into fewer reactors (consolidation) that may exist over time, and especially focus on the micro organism development [18]. But the technological and economic implications are only indicated qualitatively, and the (reported) detail in system calculation is limited.

In the present study we aim to give new insight in the development pathway of producing ethanol from lignocellulosic biomass: When could specific conversion configurations be realised, what are the key uncertainties in technology progress, and what should the RD&D strategy be? This is done by modelling and comparing concepts and drawing a route for the research, development and implementation of large-scale conversion processes. Improvement options for both individual process steps and the whole plant (integration, scale up) are assessed, which leads to key configurations that may come available in time, as development progresses. These configurations are analysed for their technical and economic performance. Development and implementation speed, and uncertainties in materialising the eventual goals are indicated. Our research consists of several steps:

- An inventory is made of process components, their stage of development, and their applicability in different process configurations. Experts were consulted to identify the potential barriers, uncertainties, and development time. The assessment includes technologies that are not yet commercially available (Section 2).
- Promising system configurations for present and future, are selected (Section 3).
- These configurations are analysed using Excel and Aspen Plus. The calculations have an approximate nature, especially to indicate the impacts of component choice, system configuration, scale, innovation, and process integration.
- Results on the energy and mass balances are used for the subsequent economic analysis. The method and assumptions for both technical and economic calculations are discussed in Section 4.

[†] Unless indicated different, all costs are in €$_{2003}$, and all GJ are on HHV basis. Costs data from previous years have been recalculated to 2003 by applying OECD deflation factors up to 1994, subsequent annual EU deflation is 3 %, annual US deflation is 2.5 %. Euros and dollars are treated equivalent (1 €$_{2003}$ = 1 US\$$_{2003}$), to ignore short-term currency fluctuations.

The results are presented and discussed in Section 5, and their sensitivity is analysed towards feedstock cost figures and scale.

2 Ethanol production

2.1 Feedstock

Biomass structure. Compact descriptions of the sugars contained in lignocellulosic biomass, and basic chemistry to extract these sugars are given by e.g. Shleser [19] and the US Department of Energy [6]. Lignocellulosic or woody biomass is composed of carbohydrate polymers (cellulose and hemicellulose), lignin and a remaining smaller part (extractives, acids, salts and minerals). The cellulose and hemicellulose, which typically comprise two thirds of the dry mass, are polysaccharides that can be hydrolysed to sugars and eventually be fermented to ethanol. The lignin cannot be used for ethanol production.

Cellulose (40 – 60 % of the dry biomass) is a linear polymer of glucose; the orientation of the linkages and additional hydrogen bonding make the polymer rigid and difficult to break. In hydrolysis the polysaccharide is broken down to free sugar molecules by the addition of water. This is also called saccharification. The product, glucose, is a six-carbon sugar or hexose.

Hemicellulose (20 – 40 %) consists of short highly branched chains of various sugars: mainly xylose (five-carbon), and further arabinose (five-carbon), galactose, glucose and mannose (all six-carbon). It also contains smaller amounts of nonsugars such as acetyl groups [20]. Hemicellulose, because of its branched, amorphous nature, is relatively easy to hydrolyse.

Lignin (10 – 25 %) is present in all lignocellulosic biomass. Therefore, any ethanol production process will have lignin as a residue. It is a large complex polymer of phenylpropane and methoxy groups, a non-carbohydrate polyphenolic substance that encrusts the cell walls and cements the cells together. It is degradable by only few organisms, into higher value products such as organic acids, phenols and vanillin. Via chemical processes valuable fuel additives may be produced. Although these by-products can significantly enhance the competitiveness of ethanol technology [6], the present study deploys lignin only for power generation.

The combination of hemicellulose and lignin provides a protective sheath around the cellulose, which must be modified or removed before efficient hydrolysis of cellulose can occur, and the crystalline structure of cellulose makes it highly insoluble and resistant to attack. Therefore, to economically hydrolyse (hemi) cellulose, more advanced pre-treatment technologies are required than in processing sugar or starch crops. After the cellulose and hemicellulose have been saccharified, the remainder of the ethanol production process is similar to grain-ethanol. However, the different sugars require different enzymes for fermentation.

Feedstock choice. The costs of ethanol production are highly sensitive to the delivered feedstock cost and the operating scale. But, unlike for biofuels from gasified biomass, the biochemical biomass

Table 1. Typical lignocellulosic biomass compositions[1][2] (% dry basis).

		Hard wood			**Softwood**	**Grass**
Feedstock		**Black locust**	**Hybrid Poplar**	**Eucalyptus**	**Pine**	**Switch grass**
Cellulose		41.61	44.70	49.50	44.55	31.98
Glucan	6C	41.61	44.70	49.50	44.55	31.98
Hemicellulose		17.66	18.55	13.07	21.90	25.19
Xylan	5C	13.86	14.56	10.73	6.30	21.09
Arabinan	5C	0.94	0.82	0.31	1.60	2.84
Galactan	6C	0.93	0.97	0.76	2.56	0.95
Mannan	6C	1.92	2.20	1.27	11.43	0.30
Lignin[3]		26.70	26.44	27.71	27.67	18.13
Ash		2.15	1.71	1.26	0.32	5.95
Acids		4.57	1.48	4.19	2.67	1.21
Extractives[4]		7.31	7.12	4.27	2.88	17.54
Heating value[5] (GJ_{HHV}/$tonne_{dry}$)		19.5	19.6	19.5	19.6	18.6

[1] The exact biochemical composition of biomass depends on many different factors, such as, growth area, used fertilizers, time of harvesting and storage conditions [11]. Softwood hemicellulose yields more 6C sugars, whereas hardwood yields more 5C sugars [6].

[2] From database at US DOE Biofuels website [6]. The fractions from the source data have been corrected to yield 100 % mass closure

[3] Bark and bark residues have a relatively higher lignin content. The empirical formula for lignin is $C_9H_{10}O_2(OCH_3)_n$, with n the ratio of MeO to C9 groups: n = 1.4, 0.94 and 1.18 for hardwood, softwood and grasses respectively.

[4] Low molecular weight organic materials (aromatics, terpenes, alcohols), some of which may be toxic to ethanol fermenting organisms, and cause deposits in some pre-treatments. Some compounds could be sold as chemicals (e.g., antioxidants) having a higher value than ethanol, but costs for purification are unknown [6].

[5] Values stem from literature [6]. However, a relation between biomass composition and heating values has been suggested by various authors: the higher heating value of lignin is 24.4± 1.2 GJ/$tonne_{dry}$, whereas the holocellulose plus the rest have a heating value of about 17 GJ/$tonne_{dry}$ [21].

composition plays a very important role in process performance, since the feedstock influences the ethanol yield via its (hemi) cellulose and sugar composition (see Table 1).

Lignocellulosic perennial crops (e.g. short rotation coppices and grasses) are promising feedstock because of high yields, low costs, good suitability for low quality land (which is more easily available for energy crops), and low environmental impact [1]. Most ethanol conversion systems encountered in literature, have been based on a single feedstock. But considering the hydrolysis fermentation process, it is possible to use multiple feedstock types. This may even be necessary to achieve the desirable large scale towards the future (see Section 3). Table 1 presents biochemical compositions for several suitable feedstock. Pine has the highest combined sugar content, implying the highest potential ethanol production. The lignin content for most feedstock is about 27 %, but grasses contain significantly less, and may thus co-produce less electricity. The base feedstock in this study is hybrid poplar, a representative hard wood. The impact of using other feedstock will be addressed in the sensitivity analysis.

2.2 Key conversion technology

A simplified generic configuration of the hydrolysis fermentation process is given in Figure 1. The key process steps will be discussed hereafter, following the here presented order. For each step, the possible

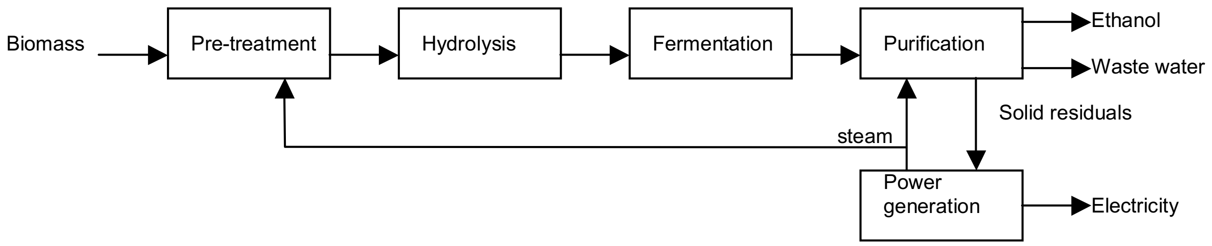

Figure 1. Generalised biomass to ethanol process.

variations at present, technological improvement options, and their research status (likelihood) will be assessed. The different configurations possible will be discussed in Section 2.3.

2.2.1 Mechanical pre-treatment

In biomass to ethanol processes, pre-treatment basically refers to the mechanical and physical actions to clean and size the biomass, and destroy its cell structure to make it more accessible to further chemical or biological treatment. The hemicellulose hydrolysis is often classed as pre-treatment, but will be discussed in the next paragraph. Each type of feedstock (whether softwoods, corn stover or bagasse) requires a particular pre-treatment method to minimize the degradation of the substrate, and to maximize the sugar yield. Cost-effective pre-treatment of cellulosic biomass is a major challenge of cellulose-ethanol technology research and development [22].

It may be necessary to clean the raw material by washing. Clean feedstock, like production wood, will in general not need this step. Subsequently, the raw material is sized: smaller chips give a larger surface area, so that transport of the catalysts, enzymes and steam to the fibres is easier and faster. This also allows the enzymes in the hydrolysis step to penetrate the fibres and to reach the sugar oligomers. Desired sizes in literature vary from a few centimetres [15] to 1 – 3 mm (before dilute acid pretreatment) [5]. Energy use - for the smallest particle sizes – can make up one third of the power requirements of the entire process [15]. Future cellulose-ethanol processes may reduce the need for costly and energy consuming mechanical pre-treatment, e.g. by accepting larger biomass chips. Extractives may be removed by steaming the chipped or milled biomass with low pressure steam (~ 160 °C) and subsequent soaking with ethanol or dilute acid [15].

2.2.2 Lignin removal, hemicellulose hydrolysis

In its function of making the cellulose feedstock more digestible by enzymes, this step is often classed as pre-treatment: the surrounding hemicellulose and/or lignin is removed, and the cellulose micro fibre structure is modified. By chemical, physical or biological treatment, lignin and all or part of the hemicellulose is solubilised. Subsequently, when water or steam is added, the free hemicellulose polymer is hydrolysed to monomeric and oligomeric sugars.

The soluble sugar products are primarily xylose, and further mannose, arabinose, and galactose. A small portion of the cellulose may already be converted to glucose. However, the cellulose bulk will be converted in a separate step. The product is filtered and pressed, solids (cellulose + lignin) go to the cellulose hydrolysis, and liquids (containing the sugars) go to a fermenting step.

During hydrolysis, hemicellulose sugars may be degraded to weak acids, furan derivates and phenolics. These compounds inhibit the later fermentation, leading to reduced ethanol yields. The production of these inhibitors increases when hydrolysis takes place at severer conditions: higher temperatures and higher acid concentrations. To raise the xylose yields, R&D is oriented at gaining new insights in chemical changes in xylose during chemical or biological treatment, at the influences of reaction conditions, and at understanding lignin-xylose interactions [6].

Chemical. Common chemical pre-treatment methods use dilute acid, alkaline, ammonia, organic solvent, sulphur dioxide, carbon dioxide or other chemicals. We discuss the most important approaches:

Acid catalyzed hydrolysis uses dilute sulphuric, hydrochloric, or nitric acids. Of all chemical pre-treatments, historically dilute sulphuric acid (0.5 to 1.5 %, T above 160 °C) has been most favoured for industrial application, because it achieves reasonably high sugar yields from hemicellulose: at least xylose yields of 75 to 90% [15; 23]. The acid will have to be removed/neutralised before fermentation, yielding a large amount of gypsum. This is usually done after the cellulose hydrolysis (see later). A concentrated acid based process also exists but is ranked to be very expensive [19].

Alkaline pre-treatment uses bases like sodium hydroxide or calcium hydroxide. All lignin and part of the hemicellulose are removed, and the reactivity of cellulose for later hydrolysis is sufficiently increased. Reactor costs are lower than those for acid technologies. However, the use of these – more expensive – salts in high concentrations raises environmental concerns and may lead to prohibitive recycling, wastewater treatment and residual handling costs. Alkaline-based methods are generally more effective at solubilising a greater fraction of lignin while leaving behind much of the hemicellulose in an insoluble, polymeric form [6].

Physical. Uncatalysed pre-treatment methods use steam explosion or Liquid Hot Water (LHW). *Steam explosion* is one of the most promising methods to make biomass more accessible to cellulase attack [24]. The material is heated using high-pressure steam (20-50 bar, 210-290ºC) for a few minutes; these reactions are then stopped by sudden decompression to atmospheric pressure. Most steam treatments yield high hemicellulose solubility and low lignin solubility. Studies conducted without added catalyst report xylose-sugars recoveries between 45 and 65%. To make it a viable option for the long term, the overall yield has to be increased and the costs have to be decreased.

The *Liquid Hot Water* process uses compressed, hot liquid water (at pressure above saturation point) to hydrolyse the hemicellulose. Xylose recovery is high (88 - 98 %), and no acid or chemical catalyst is needed in this process, which makes it economically interesting and environmentally attractive. Development of the LHW process is still in laboratory stage.

Biological. Biological pre-treatments use funghi to solubilize the lignin [22]. Biodelignification is the biological degradation of lignin by micro-organisms. It is mentioned in 1984 as possibly useful in the future [25], although at that time it was an expensive process, with low yields after long reaction time, and the micro-organisms were poisoned by lignin derivatives. Biological pre-treatment has the advantages of low energy use and mild environmental conditions. However, the very low hydrolysis rate is fatal for implementation [23]. Sometimes biological treatments are sometimes used in combination with chemical treatments [22]

Combinations. Several pre-treatment processes combine physical and chemical elements. Addition of dilute acid in steam explosion can effectively improve enzymatic hydrolysis, decrease the production of inhibitory compounds, and lead to more complete removal of hemicellulose. It is possible to recover around 70 % potential xylose as monomer. *Acid catalysed steam explosion* is one of the most cost-effective processes for hardwood and agricultural residues, but it is less effective for softwoods. Limitations include destruction of a portion of the xylan fraction, incomplete disruption of the biomass structure, and generation of compounds that may inhibit micro-organisms uses in downstream processes. The necessary water wash decreases the overall sugar yields [23].

Ammonia Fiber EXplosion (AFEX) involves liquid ammonia and steam-explosion. The process conserves the protein part; this high-value co-product is important for compensating the high process costs of the process. This is not interesting for the present research, in which low protein feedstock prevails. Also, although AFEX enhances hydrolysis of (hemi)cellulose from grass, the effect on biomass that contains more lignin (soft and hardwood) is meagre [23].

CO_2 explosion acts similar to steam and ammonia explosion. The glucose yields in the later enzymatic hydrolysis are low (75 %) compared to steam and ammonia explosion. Overall, however, CO_2 explosion is more cost effective than ammonia explosion and does not cause the formation of inhibitors as in steam explosion [23].

Comparison. Numerous pre-treatment methods or combinations of pre-treatment methods are thus available, all having their specific advantages and disadvantages. A comparison for the most promising of above discussed methods is made in Table 2. The choice for a pre-treatment technology heavily influences cost and performance in subsequent hydrolysis and fermentation [5]. The ideal pre-treatment process would produce reactive fibre; yield pentoses in nondegraded form; exhibit no significant inhibition of fermentation; require little or no feedstock size reduction; entail reactors of reasonable size (high solids loading), built of materials with a moderate cost; not produce solid residues; have a high degree of simplicity [5].

Of the promising pre-treatment options, dilute acid is as yet the most developed. Xylose yields are 75-90 %, which is much higher than when using steam-explosion (45-65 %). Dilute acid pre-treatment also produces less fermentation inhibitors, and significantly increases the later cellulose hydrolysis. However, the acid consumption is an expensive part of the method, gives a gypsum waste disposal problem and requires the use of expensive corrosion resistant materials. This may eventually tip the

Table 2. Comparison of various pre-treatment (lignin removal and hemicellulose hydrolysis) options.

Pre-treatment method	**Chemicals**	**Temperature / pressure**	**Reaction time**	**Xylose yield**	**Downstream enzymatic effect1)**	**Costs2)**	**Available**
Dilute acid hydrolysis3)	acid	>160 °C	2 – 10 min	75 – 90 %	< 85 %	+	now
Alkaline hydrolysis4)	base			60 – 75 %	55 %	++	now
Uncatalysed Steam explosion5)	-	160-260 °C	2 min	45 - 65 %	90 %	-	2 – 5 y
Acid catalysed steam explosion6)	acid	160-220 °C			88 % (2 steps)	-	2 – 5 y
Liquid Hot Water7)	none	190-230 °C $p > p_{sat}$	45 s to 4 min	88 – 98 %	> 90 %	-	5 – 10 y
Ammonia fiber explosion8)	ammonia	90 °C	30 min		50 – 90 % (2 steps)		
CO_2 explosion9)	CO_2	56.2 bar			75 % (2 steps)		

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1) This is the efficiency of the downstream enzymatic hydrolysis of cellulose to glucose, usually in 24 h. For comparison untreated poplar chips only hydrolyse 15 % in 24 h [23]. Effective pre-treatments in general approach or exceed in the subsequent enzymatic hydrolysis 80 % of theoretical cellulose conversion in five days [5].

2) + indicates that the effect is advantageous (less expensive). Steam explosion, AFEX and LHW do not require far-reaching particle size reduction, where dilute acid does [5]. On the other hand, steam explosion and LHW require large amounts of steam/water. Reith *et al.* [9] quantitatively rank the investment costs from best to worst: (1) alkaline extraction, (2) carbonic acid, (3) weak acid, and (4) strong acid and steam explosion.

3) Continuous flow process for low solids loading: 5 – 10 % substrate/total reaction mixture [23]. Dilute acid hydrolysis generally produces 80 % of the theoretical yield [5]. Requires fine grinded (1 – 3 mm) biomass [5]. The solids concentration in the reactor may be 30 %, acid concentration is 0.5 – 1 %, temperature is achieved by injection of 13 bar steam [15]. 0.3 kg dilute acid per kg feedstock is needed, or 0.12 when reckoning with recycling [15]. Compared to steam explosion, AFEX and LHW, dilute acid requires expensive construction materials (incoloy [15]), the costs are primarily a function of corrosivity and secondarily of the pressure [5]. Dilute acid hydrolysis of softwood in a two-stages process achieves yields of 89 percent for mannose, 82 percent for galactose and 50 percent for glucose [6]. Mild acid hydrolysis at 150 °C for several minutes results in a close to 100 % hemicellulose hydrolysis [11]. Enzymatic digestibility of cellulose remains under 85 % after dilute acid pre-treatment. When the dilute acid pretreated substrates are subjected to further acid hydrolysis, the cellulose susceptibility is higher than after steam explosion [26].

4) The digestibility of NaOH treated hard wood increased from 14 % to 55 % with the decrease of lignin content from 24-55 % to 20 %. However, no effect of dilute NaOH pre-treatment was observed for softwoods with lignin content greater than 26 % [23]. The yield of fermentable sugars in the pre-treatment step is qualified between steam explosion and acid hydrolysis by Reith *et al.* [9].

5) 90 % enzymatic efficiency in 24 h can be achieved in the downstream cellulose hydrolysis [23]. Particle size reduction is not required [5], however, effect and residence time in steam explosion are effected by chip size; steam explosion requires about 60 % of the energy of conventional mechanical methods to achieve the same size reduction [23]. Severe steam explosion leads to xylan and glucan losses [26].

6) Addition of H2SO4 in steam explosion improves the later enzymatic hydrolysis [23]. Water to solids ratio is 2 [23]. Total sugar production from bagasse (66.2 mass % holocellulose) has been reported 0.651 g/g bagasse, or 88 %. Water wash to remove the formed inhibiting degradation products also removes 20 – 25 % of the initial dry matter.

7) No particle size reduction required [5]. LHW both recovers most of the pentosans (over 90 %) and produces very reactive fibre (>90 % conversion). The hydrolysate barely inhibits in fermentation [27]. In a 15 g experiment, 60 – 80 litre water was used per kg$_{dry}$ biomass, for commercial applications, this amount should be decreased [28].

8) Also called AFEX. Consumes 1-2 kg ammonia/kg dry biomass. Sun [23] writes that the hemicellulose is not solubilised and the material composition after AFEX is barely changed from the original, where Lynd [5] reports a high pentose recovery. The hydrolysis of cellulose and hemicellulose in following steps varies between 50 % (aspen chips) and 90 % (grass) [23]. No particle size reduction is required [5].

9) 4 kg CO_2 kg fibre at 56.2 bar [23].

balance in favour of the less effective, but also less problematic and environmentally friendly steam explosion, also because additional research may lead to higher yields. The costs associated with steam explosion are as yet uncertain. The LHW process is still at the earliest laboratory stage and could come commercially available within 10 years, with yields projected around the 88 to 98%, higher than for dilute acid or steam-explosion. But the associated costs are uncertain (e.g. costs of the considerable water recycling).

2.2.3 Cellulose hydrolysis

Where lignin removal and hemicellulose hydrolysis are classed as pre-treatment, cellulose hydrolysis is abbreviated to hydrolysis: it is considered the major hydrolysis step. In hydrolysis, the cellulose is converted into glucose sugars ($(C_6H_{10}O_5)_n + nH_2O \rightarrow n\,C_6H_{12}O_6$). This reaction is catalysed by dilute acid, concentrated acid, or enzymes (cellulase). Hydrolysis without preceding pre-treatment yields typically < 20 %, whereas yields after pre-treatment often exceed 90 %

Acid hydrolysis. Acid hydrolysis is only applied in so-called two-stage acid processes, following acid pre-treatment. Both dilute and concentrated versions occur. The dilute acid process is the oldest technology for converting cellulose biomass to ethanol (first commercial plant in 1898). The first stage is essentially the hemicellulose hydrolysis as discussed above. If the reaction would continue, the sugars produced would convert into other chemicals – typically furfural. The sugar degradation not only reduces the sugar yield, but the furfural and other by-products can also inhibit the fermentation process. Therefore, the first stage is conducted under mild process conditions (e.g. 0.7 % sulphuric acid, 190 °C) to recover the 5-carbon sugars, while in the second stage only the remaining solids with the more resistant cellulose undergo harsher conditions (215 °C, but a milder 0.4 % acid) to recover the 6-carbon sugars. Both stages have a 3-minute residence time. Yields are 89 % for mannose, 82 % for galactose, but only 50 % for glucose. The hydrolysed solutions are recovered from both stages and fermented to alcohol [6; 22].

The concentrated acid process has a very high sugar yield (90 %), can handle diverse feedstock, is relatively rapid (10 – 12 h in total), and gives little degradation. Critical for the economical viability of this process is to minimize the amount of acid, by cost effectively separating the acid for recycling. Early (1948) membrane separation already achieved 80 % acid recovery. Continuous ion exchange (see § 2.2.4) now recovers over 97 % of the acid; 2 % of the sugar is lost. Furthermore the required equipment is more expensive than for dilute acid [6; 22].

Enzymatic hydrolysis. In the South Pacific during World War II, a fungus broke down cotton clothing and tents. This fungus, *Trichoderma Reesei*, in fact produced cellulase enzymes, which hydrolyses cellulose [6]. The first application of these enzymes for wood hydrolysis in an ethanol process was to simply replace the cellulose acid hydrolysis step with a cellulase enzyme hydrolysis step. This has several advantages: the very mild process conditions give potentially high yields, and the maintenance costs are low compared to acid or alkaline hydrolysis (no corrosion problem). The process is compatible with many pre-treatment options, although purely physical methods are typically not adequate [22; 23]. Many experts see enzymatic hydrolysis as key to cost-effective ethanol production in the long run [6].

Although acid processes are technically more mature, enzymatic processes have comparable projected costs and the potential of cost reductions as technology improves [20].

Hydrolysis is negatively influenced by structural features such as crystallinity, degree of cellulose polymerisation, and lignin content, and positively by surface area [23]. A low substrate concentration gives low yield and rate, and a high cellulase dosage may increase the costs disproportional. However, the substrate/enzyme ratio should not be too high (inhibition). Hydrolysis can be enhanced by adding certain surfactants (to facilitate desorption of cellulase after reaction), by using mixes of cellulase from different organisms, and by adding other enzymes (e.g. pectinase). Nearly complete saccharification of steam-exploded chips is possible [23].

To improve the yield and rate of the enzymatic hydrolysis, research focuses both on enhancing enzyme activity in distinctive hydrolysis and fermentation process steps [23], as well as combining the different steps in less reactors (discussed in § 2.3). Intermediate and end products of the hydrolysis, cellobiose and glucose, inhibit the cellulase activity. This can be avoided by supplying extra enzymes during the reaction, or by taking away the product by ultrafiltration or by simultaneous fermentation in the same reactor (see § 2.3). Enzymes can be recovered and recycled, so that the enzyme concentration can be higher against lower enzyme cost, although the enzyme quality decreases gradually [23].

Where chemical pre-treatment precedes enzymatic hydrolysis, poisonous materials to the enzymes need to be removed; this will be discussed in §2.2.4.

Cellulase supply. The cellulase enzyme is really a complex mix of enzymes that work together synergistically to attack typical parts of the cellulose fibre [6; 23]. Although the understanding of how cellulase acts has improved, there is still much to learn before enzyme cocktails with increased activity can efficiently be developed.

Cellulase enzymes are produced by organisms that live on cellulosic material; they may be produced in a separate reactor, or bought from industrial suppliers. In the long run cellulase production may take place in the same reactor as the hydrolysis and fermentation, which may ultimately be most efficient and more economic (§ 2.3). Both bacteria and fungi can produce cellulase enzymes, but fungi get the most research attention because of their aerobic growth conditions and fair production rate [23]. However, currently the cellulase production (optimal at 28 °C) is difficult to combine with the hydrolysis (preferably 70 °C).

Most of the present commercial applications for cellulase (esp. clothes bleaching) require only a very low cellulose hydrolysis rate, where for ethanol production still near complete hydrolysis is required. Moreover, those applications represent higher value markets, making cellulase at present an expensive product. Cellulase use is a costly part of ethanol production (now, cellulase is bought from industrial suppliers at 3.4 – 5.6 €/GJ_{HHV} ethanol, [6]). Through improved thermal stability, improved cellulose binding, reduced lignin binding, and improved active sites, cellulase enzyme performance is expected to be improved by a three-fold of 1999 in 2005, and a ten-fold in 2010 [15], so that cellulose conversion to glucose can increase, while at the same time the cost performance improves.

Comparison. Acid hydrolysis has been practised and understood for half a century and analyses of R&D-driven improvements project only modest cost improvements. The dilute acid process has a low sugar yield (50 – 70 % of the theoretical maximum). The enzymatic hydrolysis has currently high yields (75 – 85 %) and improvements are still projected (85 – 95 %), as the research field is only a decade young. Moreover, refraining from using acid may be better for the economics (cheaper construction materials, cutting operational costs), and the environment (no gypsum disposal).

Table 3. Comparison of process conditions and performance of three cellulose hydrolysis processes.

	Consumables	Temperature	Time	Glucose yield	Available
Dilute acid[1)]	< 1 % H_2SO_4	215 °C	3 min	50 – 70 %	Now
Concentrated acid[2)]	30 – 70 % H_2SO_4	40 °C	2 – 6 h	90 %	Now
Enzymatic[3)]	Cellulase	70 °C	1.5 days	75 % → 95 %	Now → 2020

[1)] Second stage of a two stage dilute acid process (0.4% sulphuric acid, 215°C, and a 3-minute residence time) yields 50 % of the theoretical yield of glucose [6]. At higher temperatures, even shorter reaction times are possible: 6 to 10 seconds at 240°C. High glucose yields (around 70%) are achieved only in highly dilute sugar streams [6], yields above 70 % are normally unattainable because glucose then is decomposed [29].

[2)] The concentrated acid process uses relatively mild temperatures, and the only pressures involved are those created by pumping materials from vessel to vessel. Reaction times are typically much longer than for dilute acid [22]. Conditions and yields from Graf and Koehler [22].

[3)] Yields are for the short to long-term systems, the high yield will come available in the CBP reactor (Table 4, note 2). The rate of biomass hydrolysis doubles for every 20 °C increase [13], to minimise reactor volume, the preferred temperature is 70 °C. At present, fermentation cannot take place at temperatures above 30 °C (and will therefore take place in a separate reactor).

2.2.4 Cleaning of substrate and liquid flows

During pre-treatment, degradation products of C5 and C6 sugars, primarily furfural and hydroxy methyl furfural (HMF) are formed, and acid is released. There may also be the acid from acid pre-treatment and hydrolysis. These components are toxic or inhibitory to the enzymatic hydrolysis and fermenting organisms and must be removed or neutralised prior to the fermentation; otherwise larger amounts of fermenting micro-organisms should be applied in fermentation.

To remove the undesired compounds, the slurry of pretreated biomass is first separated in a liquid and solid fraction. The pressed solid fraction is then washed with water to move more of the inhibitory materials to the liquid fraction. Eventually, the liquid fraction undergoes continuous ion exchange and overliming with calcium hydroxide while forming gypsum [30].

In continuous ion exchange ammonia replaces the acid (applied ratio is 1.1:1), and acids are recovered. This yields a flow of recovered acids, and a flow of purified sugar solution. For acid pre-treatment and hydrolysis situations, the acid flow is re-concentrated via multiple effect evaporators and recycled [22]. Acid recycling is necessary for good economic performance of concentrated acid processes. However, in processes using dilute acid for pre-treatment only, recovery may be too expensive compared to simple neutralization and disposal [31].

The remaining low concentrated acid is neutralised by adding lime. Overliming is the addition of $Ca(OH)_2$ to decrease acidity. Hydrated gypsum, $CaSO_4 \cdot 2H_2O$, is formed and precipitates; it is easily filtered from the sugar solution, although its inertness would also allow it to pass harmlessly through fermentation and distillation [5]. The gypsum may have some value as an agricultural soil conditioner

[32], but can also mean a waste problem. About 0.02 kg gypsum per kg feedstock needs to be disposed of, this can barely be improved [15]. Without acid recycling, the gypsum disposal from acid catalysed hydrolysis may even be 0.6 – 0.9 kg/kg feedstock [33]. After ion exchange and overliming, the hydrolysate liquid can be recombined with the solids, depending on the process configuration.

2.2.5 Fermentation

A variety of micro-organisms, generally either bacteria, yeast, or fungi, ferment carbohydrates to ethanol under oxygen-free conditions [5]. They do so to obtain energy and to grow. According to the reactions, the theoretical maximum yield is 0.51 kg ethanol and 0.49 kg carbon dioxide per kg sugar:

$$3C_5H_{10}O_5 \quad \rightarrow \quad 5C_2H_5OH + 5CO_2 \qquad \text{Equation 1}$$

$$C_6H_{12}O_6 \quad \rightarrow \quad 2C_2H_5OH + 2CO_2 \qquad \text{Equation 2}$$

Methods for C6 sugar fermentation were already known (at least) 6,000 years ago, when Sumerians, Babylonians and Egyptians began to perfect and describe the process of making beer from grain (starch). After it became possible to free the C6 sugars in lignocellulosic crops (end 19th century), conversion of the C5 sugars became interesting. They represent a high percentage of the available sugars, the ability to recover and ferment them into ethanol is important for the efficiency and economics of the process. Only in the 1980s research on xylose fermentation began to bear fruit, when a number of wild type yeast were identified that could convert xylose to ethanol [6]. Bacteria have drawn special attention from researchers because of their speed of fermentation. In general, bacteria can ferment in minutes as compared to hours for yeast.

All micro-organisms have limitations: either in the inability to process both pentoses and hexoses, the low yields of ethanol, or the co-production of cell mass at the cost of ethanol. Furthermore, the oxygen free condition of fermentation slowly exterminates the micro-organism population [5]. Therefore, in early processes, the different sugars were fermented in different sequential reactors. There is a tendency towards combining reaction steps in fewer reactors. When hydrolysis and fermentation reactions are connected directly, intermediate inhibitive products are avoided, and the yield is potentially higher. Also, genetic engineering and new screening technologies will bring bacteria and yeast that are capable of fermenting both glucose and xylose [6], although fermentation of xylose and arabinose remains problematic [34]. Near-term fermentation using genetically engineered yeast or bacteria may even utilize all five of the major biomass sugars – glucose, xylose, mannose, galactose and arabinose. Mid- to long-term technology will improve the fermentation efficiency of the organism (yielding more ethanol in less time), as well as its resistance, requiring less detoxification of the hydrolysate [15; 22].

The fermenting bacteria and yeast are grown in series of aerated seed reactors. These consume a side-streamed carbohydrate fraction (9 % of the cleaned hydrolysate [15]), and some protein nutrients. The consolidation of conversions in fewer reactors has impact on the total process integration, and is therefore discussed in §2.3.

2.2.6 Product recovery

The product stream from fermentation, also called "beer", is a mixture of ethanol, cell mass and water. In this flow, ethanol from cellulosic biomass has likely lower product concentrations (≤ 5 wt%) than in ethanol from corn. The maximum concentration of ethanol tolerated by the micro-organisms is about 10 wt% at 30 °C but decreases with increasing temperature. To maximize cellulase activity, the operation is rather at maximum temperature (37 °C), since the cost impact of cellulase production is high relative to distillation [5]. On the processing side, slurries become difficult to handle when containing over 15 wt % solids, which also corresponds to 5 % ethanol (two thirds carbohydrates, and < 50 wt% conversion) [5].

The first step is to recover the ethanol in a distillation or beer column, where most of the water remains with the solids part. The product (37 % ethanol) is then concentrated in a rectifying column to a concentration just below the azeotrope (95 %) [15]. Hydrated ethanol can be employed in E95 ICEVs [12], or in FCVs (requires onboard reforming), but for mixtures with gasoline water-free (anhydrous) ethanol is required. One can further distillate in the presence of an entrainer (e.g. benzene), dry by desiccants (e.g. corn grits), or use pervaporation or membranes [5]. By recycling between distillation and dehydration, eventually 99.9 % of the ethanol in the beer is retained in the dry product [15].

2.2.7 Residual solids / power production / wastewater treatment

The main solid residual from the process is lignin. Its amount and quality differs with feedstock and the applied process. Production of co-products from lignin, such as high-octane hydrocarbon fuel additives, may be important to the competitiveness of the process [6]. Lignin can replace phenol in the widely used phenol formaldehyde resins. Both production costs and market value of these products are complex. In corn based ethanol plants the stillage (20 % protein) is very valuable as animal feed.

In this study all residual solids (lignin, residual holocellulose compounds, and cell mass) are assumed to be deployed for production of heat and electricity. The solids come available at ~ 60 % moisture and are dried (using steam) to 15 %. To generate electricity and heat, at small scale (< 30 MW_e) probably a boiler with steam turbine would be applied. At larger scale a gasifier combined cycle (higher efficiency) may become attractive. In choosing the optimal power production means, two trends counteract each other: Future hydrolysis fermentation processes will demand less electrical energy whereas BIG/CC conversion efficiencies improve, so that the net electricity produced would increase. On the other hand future pre-treatment methods may require more steam. Besides, the consolidated processes convert more of the feedstock to ethanol, and consequently less boiler fuel will be available after the process. It may even be necessary to supply biomass feedstock as fuel directly to the boiler [5]. In that case the process can be balanced such that no excess electricity is generated.

After boiler or gasifier, ash and flue gas cleaning material remain. The amount of ash generated will depend strongly on whether acid is used in pre-treatment/hydrolysis (yielding gypsum in neutralisation). None of the solid wastes is hazardous [5]. Unfermented sugars in the liquid effluent form a non-negligible energy source and are partly recycled (40 % [15]), and partly dried and fired (as syrup) in the boiler. The residual water contains significant amounts of organic compounds such as Corn Steep Liquor (CSL), ammonium acetate, acetic acid, furfural and HMF, and needs processing before its disposal into the environment. Wooley [15] describes a treatment with two serial digesters, of which the first (anaerobic) produces biogas that is fired in the boiler (8 % of the total boiler load in that study). After the second (aerobic) digester, 99 % of the organic material is eliminated. Any sludge formed is also processed in the boiler.

2.3 Process integration

Where enzymatic hydrolysis is applied, different levels of process integration are possible (Figure 2). In all cases, pre-treatment of the biomass is required to make the cellulose more accessible to the enzymes, and to hydrolyse the hemicellulose.

The first application of enzymes to wood hydrolysis in an ethanol process was to simply replace the cellulose acid hydrolysis step with a cellulase enzyme hydrolysis step. This is called Separate (or Sequential) Hydrolysis and Fermentation (SHF). In the SHF configuration the joint liquid flow from both hydrolysis reactors first enters the glucose fermentation reactor. The mixture is then distilled to remove the ethanol leaving the unconverted xylose behind. In a second reactor, xylose is fermented to ethanol, and the ethanol is again distilled [6; 35]. The cellulose hydrolysis and glucose fermentation may also be located parallel to the xylose fermentation.

Simultaneous Saccharification and Fermentation (SSF) consolidates hydrolyses of cellulose with the direct fermentation of the produced glucose. This reduces the number of reactors involved by eliminating the separate hydrolysis reactor and, more important, avoiding the problem of product inhibition associated with enzymes: the presence of glucose inhibits the hydrolysis [6]. In SSF there is a trade-off between the cost of cellulase production and the cost of hydrolysis / fermentation. Short hydrolysis reaction times involve higher cellulase and lower hydrolysis fermentation costs than longer reaction times. The optimum is constrained by the cost of cellulase, and is about 3-4 days. SSF is assumed state of the art by Lynd in 1996 [5].

Lynd announced the co fermentation of hexoses and pentoses sugars (SSCF) as a focus for near-term development (in 1996), which meanwhile is being tested on pilot scale [6].

In Consolidated BioProcessing (CBP) ethanol and all required enzymes are produced by a single micro-organisms community, in a single reactor [5]. CBP seems the logical endpoint in the evolution of biomass conversion technology. Application of CBP implies no capital or operating costs for dedicated enzyme production (or purchase), reduced diversion of substrate for enzyme production, and compatible enzyme and fermentation systems [5]. As yet, there are no organisms or compatible combinations of micro organisms available that both produce cellulase and other enzymes at the required high levels and also produce ethanol at the required high concentrations and yields [5; 22], although various organisms already combine multiple functions. Approached pathways in the

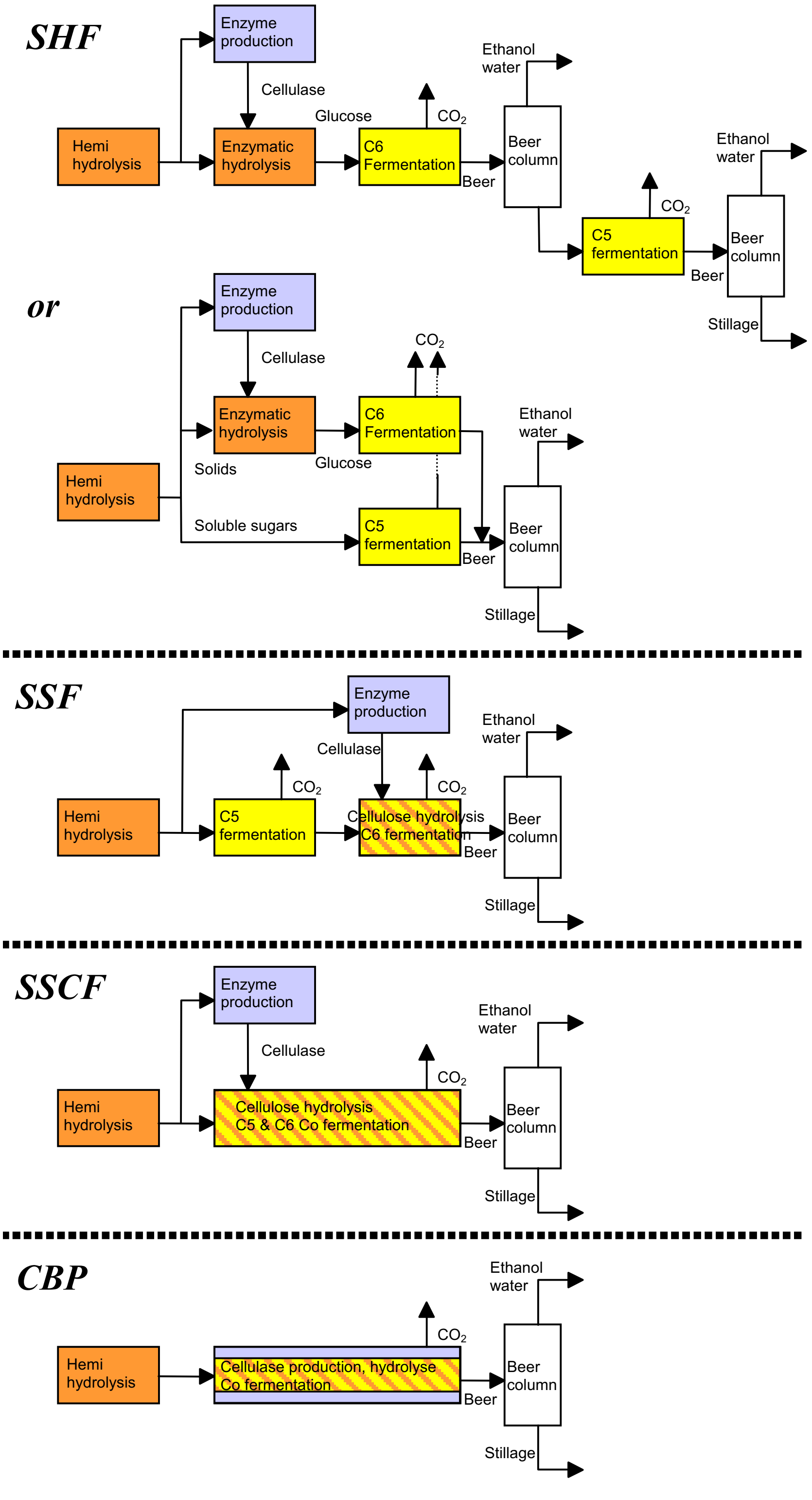

Figure 2. Levels of consolidation in enzymatic hydrolysis, fermentation, and enzyme production.

Table 4. Comparison of enzymatic process configurations.

	Micro-organism	**Consumed / lost**1)	**Conversion efficiency**			
			Cellulose2) **→ Glucose**	**Glucose → Ethanol**	**Xylose → Ethanol**	**Mannose Galactose Arabinose → Ethanol**
SHF3)	*S. Cerevisiae* (recombinant)	Cellulose 6 %	75 %	← 85 – 90 % →		
SSF4)	*S. Cerevisiae* (recombinant)	Cellulose 6 %	80 %	92.5 %	← 80 – 92 % →	
SSCF5)	Cellulase6)	Cellulose 5 % Sugars 5 %	88 %			
	*Z. Mobilis*7)	Glucose 4 % Xylose 3 % CSL All sugars 7 %		92 %	85 %	90 %
CBP8)	As yet unknown	Cellulose 4 %	90 %	← 92 – 95 % →		

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1) Cellulase producing organisms consume (2-6 % of the) cellulose, (6 % of the “feedstock” [11]). Fermenting organisms (“ethanologen”) consume sugars and a small amount of nutrients. A certain fraction of the sugars may also be lost to other products (contamination) [15].

2) Enzymatic hydrolysis gives high yields, which will show a gradual increase from 75 – 85 % now (SHF) to 85 – 95 % in future (CBP). The effective yield also depends on the pre-treatment method. The presented numbers hold for dilute acid pre-treatment and will be slightly higher for steam explosion and liquid hot water.

3) The fermentation yield (85 – 90 % of the theoretical maximum) is low due to enzyme inhibition by the products of the hydrolysis and is comparable with the starch hydrolysis and fermentation process. Fermentation by *Saccharomyces Cerevisiae* may achieve 90 % of theoretical [6].

4) In SSF fermentation with *Saccharomyces Cerevisiae* (yeast), ethanol yields are 92.5 % for glucose and 80 – 92 % for xylose [11]. However, on the short term, xylose and arabinose fermentation may remain low, 59 % was achieved by Sonderegger and Sauer [34].

5) Wooley *et al*. consider 7 % of all fermentable sugars to be lost to contamination.

6) Glucose yield holds for dilute acid pre-treatment. The most common organism used to produce cellulase industrially is *Trichoderma Reesei* (fungus) [24]. It grows in aerobic conditions and consumes both soluble sugars and holocellulose. 5 % of the clean hydrolyzate is directed to the cellulase reactor, where 100 % of the sugars is consumed, 0 % of the sugar oligomers, 0 % of the CSL, 100 % of the cellulose, 0 % of the hemicellulose, 0 % of the lignin. The total of cellulase (77 %) and biomass (23 %) produced equals 26 % of the cellulose and sugar consumed [15], furthermore CO_2 is produced at 107 % of the mass of cellulose and sugar consumed. Cellulase productivity on hardwood is significantly higher than on softwood [24]. Productivity increases with increasing cellulose concentration, but the yield per gram cellulose decreases due to mass transfer limitation [24]. Reaction time is 5 – 7 days [18]. The costs associated with dedicated cellulase production are 0.5 US$/gal ethanol, or 50 US$/tonne$_{\text{dry}}$ biomass hydrolysed [18]. Through time, less cellulase will be necessary, because of increased specific enzyme activity: threefold in 2005, and tenfold in 2010 [15].

7) Wooley *et al*. [15] model hydrolysis, fermentation (*Zymomonas mobilis* bacterium) and seed production to take place in both seed reactor and SSCF reactor. For hydrolysis and fermentation, numbers are taken from the SSCF reactor. For sugar consumption, the number is taken from the seed reactor. SSCF fermentation of glucose yields ~ 93 % (of the theoretical maximum) [11]. Only conversion yields for glucose and xylose where found, the other sugars are assumed to convert at 90 % (average of other configurations). The nutrient Corn Steep Liquor or CSL (actually a 0.25 wt% dilution) is a necessary nitrogen source (7.7 % of the mass of entering cellulose) [15].

8) The fermentation yield of the bio ethanol production is 92 – 95 % (of the theoretical maximum) [36]. In aerobic situation, cellulase yield is much higher than in anaerobic situations. On the other hand, fermentation gives higher yields in anaerobic situation. In SHF, SSF and SSCF these reactions take place in separate reactors. In CBP, where cellulase production and fermentation are combined in one reactor, an anaerobic micro-organism culture will be used [18].

development of CBP organisms are described by Lynd *et al.* [18]. The different levels of consolidation are compared in Table 4.

Lynd *et al.* [14] expect significant cost reductions when progressing from improved SSF (10.5 €/GJ anhydrous ethanol; feedstock costed 2.1 €/GJ) via SSCF (9.8 €/GJ) to CBP (4.5 €/GJ). Others suggest that, as yet, cost reductions reside in optimising the separate reactors, because hydrolysis, growth and fermentation processes require very different temperature, pH, and oxygen level. Moreover, a configuration with separate reactors allows for better process control [33].

3 System selection

With the described technology, different configurations are possible. For further analysis, three systems are selected that could represent the short (5 years), middle (10 – 15 years) and long term (20 years or more). Based on status of the technology, the short-term system may only consist of components that are currently commercially available, or in pilot stage. The components of the middle term system are either in pilot stage or promising laboratory stage. The long-term system eventually inclines toward the theoretical possibilities of biomass to ethanol conversion; it incorporates components that are as yet being developed in laboratories. Furthermore, through time there will be a tendency towards application of large scale, increasing process integration and increasing yields, which will be incorporated in this analysis.

In Table 5 the three systems are characterised: The short term system applies dilute acid pre-treatment, which certainly has disadvantages in economic and environmental sense. Good alternatives are, however, not yet available. Enzymatic hydrolysis of cellulose is applied, already having comparable costs and higher yields than the (longer-known) dilute acid technology [33]. The necessary cellulase is produced from a 5 % split off stream after pre-treatment. The scale is set at 500 MW input, a boiler produces process steam and a steam turbine converts some steam to electricity.

For middle term pre-treatment steam explosion is assumed available. The direct xylose yield is lower than in dilute acid pre-treatment, but the downstream glucose yield in enzymatic hydrolysis increases from 80 to 90 %. Cellulase could also be produced in a separate reactor, but it may be cheaper to buy cellulase, especially as several companies are seriously aiming at producing cellulase a tenfold cheaper [6] Specific enzyme activity is three times higher than in the short term: while doubling the input

Table 5. The main differences in technology and step yields of the three selected systems.

	Short-term (5 yr)	**Mid-term (10 – 15 yr)**	**Long-term (> 20 yr)**
Biomass input HHV	400 MW	1000 MW	2000 MW
Pre-treatment	Milling and Dilute acid	Steam Explosion	LHW
Cellulase production	Separate reactor	Bought elsewhere	CBP reactor
Cellulose hydrolysis	Enzymatic	Enzymatic	Enzymatic
Process integration	SSF	SSCF	CBP
Power Generation	Boiler	BIG/CC	Boiler or BIG/CC
Present development stage	Available / pilot	Pilot / Laboratory	Laboratory / Sketch

capacity, less cellulase is needed [15]. The large scale (1000 MW input) allows for a first generation BIG/CC to produce steam and electricity.

The long-term scenario assumes that LHW directly yields high amounts of xylose, and downstream high amounts of glucose. The single CBP reactor and the larger scale make the concurrent cellulase production again cheaper than buying the enzymes. Because of the tenfold cellulase activity compared to the short term, its production consumes only a small fraction of the sugar and cellulose. The high overall biomass to ethanol conversion may leave no biomass for surplus electricity production. The plant scale doubles to 2000 MW input. Depending on the choice to either maximise fuel production, or efficiently co-produce electricity, a boiler or a BIG/CC is applied.

4 Modelling

4.1 Technical modelling

All main process data and assumptions for the chosen configurations and individual process steps (as discussed in the previous chapters) are summarised in Table 6. The modelling is done as follows: The production of ethanol is modelled in a spreadsheet; for each process step in the hydrolysis fermentation process, conversion percentages and losses are applied, to yield intermediate amounts of sugar and final amounts of ethanol and solid residuals. Steam and electrical power demand follow from solid biomass throughputs in the pre-treatment. The thermal conversion of the residual solids to steam and electricity is modelled in Aspen Plus. Internal steam use (pre-treatment, distillation, residue drying) and power use (air compressor, pumps and agitators for the reactor vessels, other auxiliaries) are subtracted from the amounts produced on the power island.

Cellulase production in the short term, in a separate reactor, consumes part of cellulose and xylose. In the long term cellulase production concurs with sugar production and fermentation. The cellulase production is modelled in two steps: (1) consuming part of the cellulose, and (2) when after hydrolysis all the sugars are available, part of all sugars is consumed. In other words, consumption precedes conversion. The continuous increasing specific enzyme activity is incorporated by decreasing the cellulase production or purchase for the middle and long term [15].

The solid residuals left are dried to 10 % mc and fired in either a boiler or a gasifier. This part is modelled in Aspen Plus, and requires data on heating value and composition of the materials involved. The higher heating value (dry basis) of the residual solids is calculated from the lignin and holocellulose fraction (note 5 under Table 1). The amounts of C, H and O are calculated from the amounts of polysaccharides and lignin left, accounting for the different lignin compositions in different feedstock (note 3 under Table 1). 40 % of the unconverted sugars in the liquid effluent are recycled; the rest is dried and fired in the boiler/gasifier [15]. The plant's waste water contains significant amounts of organic compounds, and undergoes anaerobic digestion. The amount of producible methane is derived from the amount of acetic acid extracted from biomass in the pre-treatment ([15] note 4 under Table 6). The boiler and gasifier are modelled as earlier described by Hamelinck and Faaij [17] and Faaij *et al.* [37].

Table 6. The main parameters and step yields for modelling of the three selected systems.

	Short-term **(5 yr)**	**Mid-term** **(10 – 15 yr)**	**Long-term** **(> 20 yr)**
Biomass input	400 MW$_{HHV}$	1000 MW$_{HHV}$	2000 MW$_{HHV}$
Pre-treatment	Dilute acid	Steam Explosion	LHW
Saccharification hemicellulose	75 – 90 %	45 – 65 %	88 – 98 %
Cellulase production	consumes 2 – 6 % of cellulose	bought	consumes 1 % of cellulose
Saccharification cellulose[1]	70 – 80 %	90 – 96 %	97 – 99 %
Cellulase production[2]	consumes per kg ethanol 63 g sugars 93 g cellulose	-	consumes per kg ethanol 6.3 g sugars 9.3 g cellulose
Seed fermenting and byproducts[3]	← consumes per kg ethanol → 4.0 g glucose 7.4 g xylose 130 g CSL		
Fermenting			
xylose	80 – 92 %	80 – 90 %	92 – 95 %
glucose	90 – 95 %	90 – 95 %	92 – 95 %
other sugars	80 – 92 %	90 %	92 – 95 %
Process integration	SSF	SSCF	CBP
Power Generation[4]	← Boiler or BIG/CC →		
Power use[5], kW$_e$ per MW$_{HHV}$ input			
chipper	5	1	1
air compressor	31	n/a	n/a
reactor vessels	27	13	7
auxiliaries	29 (sum: 92)	29 (sum: 43)	29 (sum: 37)
Steam use[6]			
pre-treatment (kg per kg dry feed)	0.2 (4 bar) 0.3 (11 bar)	0.2 (4 bar) 0.1 (25 bar)	0.8 (4 bar) 0.4 (25 bar)
drier (tonne/twe)	1.01 (11 bar)	1.01 (11 bar)	1.01 (11 bar)
distillation (kg per l ethanol)	2.57 (4 bar)	1.03 (4 bar)	1.03 (4 bar)

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[1] Glucose yield (after dilute acid pre-treatment) increases from 75 % via 88 % to 90 %, for SSF → SSCF → CBP (Table 4); but the relative yields after different pre-treatments also increase: dilute acid: ~ 85 %, steam explosion ~90 %, LHW ~ 93 %.

[2] Cellulase consumption in the short term is same as for the SSCF plant modelled by Wooley *et al.*: an SSCF reactor producing 17653 kg/h ethanol consumes 547 kg/h cellulase. To produce this 547 kg cellulase, 1120 kg sugar and 1636 kg cellulose is consumed. A threefold reduction (in cellulase consumption) is applied for the middle term, and tenfold for the long term, but in the middle term cellulase will be bought from dedicated cellulase producers.

[3] The production of seed and by-products consumes 70 kg glucose (calculated form the cellulose part hydrolysed in the seed reactor, 85 % of the glucose is directly converted to ethanol, and a small amount leaves the reactor), 130 kg xylose, and 2326 g Corn Steep Liquor (or CSL, a necessary nitrogen source nutrient).

[4] Three material flows are deployed for power generation: all solid residuals, 60 % of the unconverted sugar, and methane from the anaerobic wastewater treatment. The solid residuals contain 63 % moisture and have to be dried to ~ 10 %, the heat consumption is 2.8 MJ/twe (12 bar, 200 °C steam) and the electricity consumption is 70 kWh$_e$/twe [38]. Since the material is dried before the boiler, the boiler efficiency is assumed 90 % (this in contrast to the 50 % moisture feed applied by Wooley *et al.*, and their 62 % boiler efficiency). In Wooley *et al.* [15], 1137 kg methane and 1292 kg CO_2 are produced from wastewater. These products stem largely from 3606 kg ammonium acetate, formed in continuous ion exchange from acetic acid. The amount of acetic acid follows directly from its fraction in the biomass feedstock (Table 1).

[5] In the 450 MW$_{HHV}$ input SSCF plant in Wooley *et al.* [15], electricity is mainly used by the air compressor (14 MW$_e$, air for cellulase production), and the pumps and agitators of the reactor vessels (6 MW$_e$); other auxiliaries use another 13 MW$_e$. In CBP no air will be required (anaerobic cellulase production). Decreased or increased reactor integration will influence the energy used by pumps and agitators, this influence is assumed proportional to the total reactor capacity, or the number of reactor trains. Compared to SSCF, SSF is assumed to have twice as much reactor volume and CBP half. For dilute acid pre-treatment the feedstock is chipped from 50 to 2 mm, for steam explosion and LHW the feedstock may be 19 mm [15].

[6] All pre-treatment options require steam. Dilute acid uses 0.5 kg steam of 4 bar per l ethanol, and 0.7 kg/l of 11 bar. LHW requires steam of 4 bar (1.8. kg/l) and 25 bar (0.8 kg/l) [14]. Via the reported efficiencies (46.1 % and 54.1 % HHV respectively) and the original poplar feedstock HHV$_{dry}$, the steam requirement per tonne feedstock is calculated. From the difference in solids loading, 10 – 15 % for LHW and > 50 % for steam explosion [14; 27], it is deduced that steam explosion

requires about a quarter of the amount of steam of LHW, the desired steam pressures are similar. Distillation in all cases requires steam 2.57 kg of 4 bar per l ethanol, in the LHW case this is partly (60 %) rest heat from the pre-treatment [14], it is assumed that also steam explosion rest heat can be used. The drier uses 1.01 tonne steam of 11 bar per tonne water evaporated.

4.2 Economic analysis

Ethanol production costs are calculated by dividing the total annual costs of each system by the produced amount of ethanol. The total annual costs consist of annual capital requirements, operating and maintenance (including maintenance, consumables, labour, waste handling), biomass feedstock and electricity reimbursement (fixed power price). Relevant parameters to calculate these costs and the resulting ethanol costs are given in Table 7.

Most cost parameters are assumed to remain constant through time (excluding inflation). This may not be realistic, as some consumables may become more expensive if raw materials would run down, energy prices would rise, or environmental measures would be enacted. On the other hand, the reimbursement for the co-produced electricity could then increase.

Table 7. Parameters for the economic evaluation 1).

Interest rate		10 %
Economical lifetime		15 years
Technical lifetime		25 years
Investment path 2)		20 % in first year, 30 % in second and 50 % in last year
Operational costs 2)		
Fixed	Maintenance	3 % of TCI
Variable	Labour	0.5 % of TCI at 400 MW_{HHV} input decreasing with scale (R = 0.25)
	Gas cleaning	0.5 % of BIG/CC capital investment
	Insurance	0.1 % of TCI
	Consumables	
	Dilute acid	0.82 €/$tonne_{dry}$ biomass
	Lime	0.87 €/$tonne_{dry}$ biomass
	Cellulase	0.13 → 0.044 → 0.013 €/l ethanol (purchase)
	Ammonia	0.24 €/kg, consumption is 0.062 kg/l ethanol (cellulase production integrated)
	CSL	0.20 €/kg, consumption is 0.086 kg/l ethanol (cellulase production integrated)
	Dolomite	50 €/tonne, dolomite use is 0.3 kg/kg clean dry wood
Biomass 3)		3 $€_{2002}/GJ_{HHV}$ (short term), 2.5 $€_{2002}/GJ_{HHV}$ (medium), 2 $€_{2002}/GJ_{HHV}$ (long)
Electricity (reimbursement)		0.03 €/kWh
Annual load		8000 hours (91 % of time)

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1) From Hamelinck *et al.* [17], Faaij *et al* [37] and Wooley *et al.* [15]. Costs in $€_{2003}$ unless indicated otherwise.

2) From the investment path follows that the total capital requirement TCR is 118 % of the total capital investment TCI.

3) Operating and Maintenance costs consist of fixed and variable costs. Fixed O&M (labour, overhead, maintenance, insurance and taxes) are annually 3 – 3.5 % [13; 37] of the TCI. Variable O&M include consumables and disposal costs depending on mass balances in the plant. A breakdown of variable O&M for BIG/CC was applied by Faaij *et al.* [37]: Operation costs depend on labour [39], catalyst and chemicals consumption, residual streams disposal, and insurance. Labour costs decrease with scale and are 0.5 % of the TCI for a 400 MW HHV input. Insurance is 1 % of annual depreciation (Faaij *et al*). Costs for consumables typical for ethanol production (from Wooley *et al.*) were originally per $tonne_{52\%}$. Dilute acid and lime are only consumed in and after dilute acid pre-treatment. The costs associated with dedicated cellulase production are at present 0.5 US$/gal ethanol, or 50 US$/$tonne_{dry}$ biomass hydrolysed [18], but decrease by a factor 3 and 10 for the middle and long term. CSL and ammonia are major consumables in cellulase production when integrated in the ethanol plant; consumption decreases by a factor 10 in the long term (because less cellulase required).

4) Prices of delivered cultivated energy crops and forest thinnings in Western Europe amount currently 3-5 €/GJ_{HHV}, United States 2.3-3.3 €/GJ_{HHV}, and at some Latin American locations even lower costs are possible 1.2 - 2 €/GJ_{HHV} [40; 41]. Improved crops and production systems are expected to bring the biomass price to the 1.6-2.1 €/GJ_{HHV} range (United States [3]).

Table 8. Costs of system components in M€$_{2003}$$^{1)}$.

Component	Base investment costs$^{2)}$	Scale factor	Base scale	Installation factor$^{2)}$	Maximum size
Pre-treatment					
Mechanical$^{3)}$	4.44	0.67	83.3 tonne$_{dry}$/h (input)	2.0	83.3
Mill$^{4)}$	0.37	0.7	50 tonne$_{wet}$/h	1	
Dilute acid$^{5)}$	14.1	0.78	83.3 tonne$_{dry}$/h (input)	2.36	
Steam explosion$^{6)}$	1.41	0.78	83.3 tonne$_{dry}$/h (input)	2.36	
LHW$^{7)}$	5.62	0.78	83.3 tonne$_{dry}$/h (input)	2.36	
Ion exchange$^{5)}$	2.39	0.33	83.3 tonne$_{dry}$/h (input)	1.88	
Overliming$^{5)}$	0.77	0.46	83.3 tonne$_{dry}$/h (input)	2.04	
Hydrolysis + fermentation					
Cellulase production$^{8)}$ (SSF)	1.28	0.8	50 kg/h cellulase	2.03	50
Seed fermentors$^{9)}$ (SSF+SSCF)	0.26	0.6	3.53 tonne/h ethanol	2.20	3.53
C5 fermentation (SSF)$^{10)}$	0.67	0.8	1.04 tonne/h ethanol	1.88	1.04
Hydrolyse-fermentation (SSF)$^{11)}$	0.67	0.8	1.04 tonne/h ethanol	1.88	1.04
SSCF$^{12)}$	0.67	0.8	1.04 tonne/h ethanol	1.88	1.04
CBP$^{13)}$	0.67	0.8	1.04 tonne/h ethanol	1.88	1.04
Upgrading					
Distillation and purification$^{14)}$	{ 2.96	0.7	18466 kg/h ethanol	2.75	18466
	1.35	0.7	9233 kg/h ethanol	2.75	9233
Molecular sieve$^{14)}$	2.92	0.7	18466 kg/h ethanol		18466
Residuals					
Solids separation$^{15)}$	1.05	0.65	10.1 tonne$_{dry}$/h	2.20	10.1
(An)aerobic digestion$^{16)}$	1.54	0.6	43 tonne/h waste water	1.95	43
Drier$^{17)}$	7.98	0.8	33.5 tonne$_{wet}$/h	1.86	110
Power Island					
Boiler$^{18)}$	27.1	0.73	173 MW steam raised	2.20	
Gasifier$^{19)}$	40.0	0.7	68.8 tonne$_{dry}$/h	1.69	75
Gas turbine$^{19)}$	16.9	0.7	26.3 MW$_e$	1.86	
HRSG$^{19)}$	7.34	0.6	39.2 kg/s steam	1.84	
Steam system + turbine$^{19)}$	5.36	0.7	10.3 MW$_e$	1.86	

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1) Average annual GDP deflation (after 1994) is assumed to be 2.5 % for the US, 3.0 % for the EU, 1 €$_{2003}$ = 1 US$\$_{2003}$.

2) As the cost numbers stem from different literature sources, the base investment costs may be anywhere between free on board (f.o.b.) and total installed costs. To become the components capital investment, the base investment is scaled and multiplied with the installation factor [42]. Wooley *et al*. apply both an "installation factor" (different for each piece of equipment) and "additional cost factors" (together ~ 1.57). Their additional cost factors are in fact OSBL, Indirect Costs, Start-up and Working Capital, so that the "installation factor" must give the ISBL. To comply with the method followed here, Wooley *et al.*'s installation factors are increased by 6.4 % to yield the direct installation factor. The indirect installation factor (see text) therefore is 94 % of the "additional cost factors", 1.48. The direct costs part of the overall installation factor is decreased with scale (average R-factor is 0.82, derived from [37]).

3) Pre-treatment consists of feeders, conveyors, separators, hoppers, and a washer. Also a chipper (~ 20 mm) is included for all concepts. This pre-treatment [15] is much cheaper than before gasification [42], because the expensive dryer can be omitted. The *direct* installation factor is 1.28, so that the total installation factor is 2.0.

4) A mill (1-3 mm) is only used before dilute acid pre-treatment. Grinding 50 tonne$_{wet}$/h with a hammermill costs 0.37 M€$_{2003}$ (total installed), scale factor is 0.7 [43]. The disk refiner in Wooley *et al*. [15] costs about the same (0.38 MUS$\$_{1997}$ for 159 tonne$_{wet}$/h, scale factor 0.62, installation factor 1.3), but makes 20 mm chips. Grinding before gasification is much more expensive: 33.5 tonne$_{wet}$/h (plant input) costs 0.41 MUS$\$_{2001}$, scale factor 0.6 and installation factor 1.86 [17].

5) Wooley *et al*. gives costs for dilute acid pre-treatment, continuous ion exchange, and overliming (in their Appendix B second table). Some costs are difficult to allocate to one or another part of the process. Three main components/areas are discerned: The dilute acid pre-treatment reactor (12.46 MUS$\$_{1998}$, scale factor 0.78, direct installation factor 1.5, see note 2), the ISEP continuous ion exchange reactor (2.06 MUS$\$_{1997}$, scale factor 0.33, installation factor 1.2), overliming and neutralisation area (0.66 MUS\$1997, scale factor 0.46, installation factor 1.3). The residual costs from the table are equally divided over these components to increase the quoted installation factors with 21 %. The costs hold for the base plant scale of 83.3 tonne$_{dry}$/hr.

6) Van Hooijdonk [36] assumes the costs for the steam explosion reactor similar to dilute acid pre-treatment. However, Lasser *et al*. do experiments with LHW and steam explosion in essentially the same reactor. Steam explosion can have a fourfold larger solids loading compared to LHW (by mass).

7) Direct capital costs for LHW could not be found, but are derived from Lynd *et al.* [14]. LHW specific capital costs in an advanced technology case are 3.8 €/tonne$_{dry}$ biomass processed, where dilute acid (including milling) has capital costs of 13.9 €/tonne$_{dry}$ biomass processed, the throughput for LHW was 4.2 times bigger and no milling is included. This implies that at same scale, the specific capital costs are 2.5 times smaller for LHW compared to dilute acid pre-treatment. At present, LHW would instead be much more expensive than dilute acid [36].

8) 547 kg/h Cellulase production [15, Appendix B Table 4, and PFD A400] takes place in 11 reactors. Equipment costs for this complete cellulase area are 12.05 MUS$_{1997}$ (scale factor 0.8, direct installation factor 1.29). For cellulase purchase (middle term) see Operational costs in Table 7.

9) 5 Seed fermentors produce the fermenting micro-organisms for eventual production of 17.65 tonne/h ethanol. The area costs are estimated from Wooley *et al.* [15, Appendix B Table 3]: 1.11 MUS$_{1997}$ (scale factor 0.6, direct installation factor 1.4).

10) Pentose fermentation (SSF) is assumed to take place in a range of reactors similar in size to that of SSCF fermentation (note 12).

11) Cellulose hydrolyse and hexose fermentation (SSF) are assumed to take place in a range of reactors similar in size to that of SSCF fermentation (note 12).

12) 17 SSCF fermentors are needed to eventually produce 17.65 tonne/h ethanol, the area costs are determined from Wooley *et al.* Appendix B Table 3 by subtracting the Seed fermentor costs (note 9) from the total A300 area costs: 9.87 MUS$_{1997}$ (scale factor 0.8, direct installation factor 1.2).

13) The CBP reactor train is assumed similar in size to that of SSCF fermentation (note 12).

14) By beer distillation and rectification, the hydrated product is obtained. The equipment for the total area A500 [15] costs 7.41 MUS$_{1997}$, but includes the molecular sieve (2.52 MUS$) for the anhydrous product, which is dealt with separately. The scale factor is 0.7, direct installation factor is 1.75. The capacity is 18.47 tonne/h ethanol, but a significant part of the major equipment (2.33 MUS$) has a maximum size half of this.

15) Solids separation after the beer distillation includes lignin separation, and from wastewater treatment it includes a sludge belt press and screw (part of A600, Wooley *et al.*). An installation delivering 30.3 tonne$_{dry}$/h solids costs 2.71 MUS$_{1997}$, scale factor 0.65, direct installation factor 1.4. At this capacity a major equipment part is needed in threefold.

16) Costs for Area 600 (Wooley *et al.*) except solids separation (note 15): 5.31 MU$_{1997}$, scale factor 0.6, direct installation factor 1.24. The scale is 173 tonne/h water (with a major item needed in fourfold). The present model does not yield the amount of water, but it is assumed that this amount relates quite direct to the amount of ethanol, because the micro-organisms have a limited ethanol tolerance (see § 2.2.6). In the described plant the largest part of the water is recycled before water treatment, eventually per kg ethanol 9.4 kg water is cleaned.

17) Wooley *et al.* present a drier (98 tonne$_{wet}$/h input) that dries from mc 62.5 → 51.2 %. In the boiler, biogas and natural gas are cofired to increase the performance. The present model applies a drier feedstock (~10 %, energy consumption is 2.8 MJ/twe, or 1.01 tonne steam/twe) for boiler or gasifier, so that no natural gas co firing is necessary. Data are taken from [17], the overall installation factor is 1.86.

18) The costs for the boiler (from Wooley *et al.*) include the fluidised bed combustion reactor, feeders, BFW preheater, steam drums, and superheater, but exclude the steam turbine. The total costs are 23.4 MUS$_{1997}$, scale factor 0.73, direct installation factor is 1.4, for a system that raises 235 tonne/h superheated steam of 510 °C 86 bar (from BFW 177 °C, 98 bar), or transfers 173 MW of heat.

19) The cost numbers presented for gasification, gas turbine, HRSG and steam turbine were previously discussed by Hamelinck and Faaij [17]. 85 % of the costs of gas turbine + HRSG are in the gas turbine.

The total capital investment, or TCI, is calculated by *factored estimation* [17; 44], based on known costs for major equipment as found in literature or given by experts. The uncertainty range of such estimates is up to ± 30 %. Usually the TCI follows from multiplying the total purchased equipment costs by a factor to yield the Inside Battery Limit (ISBL)‡, adding the costs of the Outside Battery Limit (OSBL) to yield the direct costs, adding indirect costs to yield the fixed capital investment, and finally adding working capital and start-up costs. However, the advised method cannot be followed entirely as base equipment costs in literature may be anywhere between f.o.b.§ and total installed capital, and often a specific overall installation factor is given to yield the TCI of that piece of equipment.

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‡ *Inside Battery Limits* only deals with the purchase and installation of process equipment, piping, instrumentation, controls, process buildings, etc. *Outside Battery Limits* includes utilities such as power distribution, steam plants, instrument air systems, sewers, waste water treatment, tankage, cooling towers, control buildings, land, etc.

§ *Free on board*: the price of a traded good after loading onto a ship but before shipping, thus *not* including transportation, insurance, and other costs needed to get a good from one country to another; cost of equipment ready for shipment from supplier.

Therefore the TCI is first calculated for each separate unit, and later all unit TCIs are added up. The unit TCI depends on the size of that unit (which follows from the spreadsheet and Aspen Plus modelling), by scaling from a known size (see Table 8). Various system components have a maximum size, above which multiple units will be placed in parallel. Hence the influence of economies of scale on the total system costs decreases. This aspect is dealt with by assuming that the base investment costs of multiple units are proportional to the cost of the maximum size: the base investment cost per size becomes constant. After scaling, the TCI of each component is found by multiplying the scaled base cost by an installation factor.

The individual equipment costs at base scale and with an installation factor as presented in Table 8, stem for a large part from a few sources: Wooley *et al.* [15] is used concerning hydrolysis, fermentation and upgrading. For the power island we used data we presented earlier [17]. For other configurations than the SSCF presented by Wooley *et al.*, we have assumed that more (SSF) or less (CBP) reactors of the same size are required per amount of ethanol produced, since less or more functions are combined per reactor. Specific arguments for the applied individual equipment costs are given in the footnotes under Table 8.

The given numbers must be used with reserve, as the base equipment costs in literature are often ill defined. Also the applied percentages for additional costs differ greatly between studies. Of course these percentages depend on the specific location: OSBL costs in industrial areas may be lower since much of the necessary infrastructure is readily available. However, no literature was found on this subject. Therefore, the influence of location on the overall installation factor is not incorporated in the present study. All the factors to calculate TCI from purchased equipment can be expected to be very dependent on scale. ISBL relatively decreases with scale because of increasing process integration and OSBL decreases because some offsite facilities (e.g. storage, harbour, sewage system) have a fixed same scale for both small and large plants. Where the composition of the installation factor (direct, indirect, working capital and start-up) is known, the direct costs part of the overall installation factor is decreased with scale (average R-factor is assumed to be 0.82, derived from [37]). Otherwise, the installation factor is kept constant between scales.

5 Techno-economic performance

In this section we present the results on technical modelling, process economics and the sensitivity towards several parameters. Poplar is used as the base feedstock. Figure 3 (left) shows the increasing ethanol yield from short and middle to long-term concepts, and the decrease of combustible co-products. The summed efficiency of product and residuals is very stable at 88 – 89 %. This is especially because each increase in (hemi)cellulose hydrolysis induces an equal loss in solid residuals.

Furthermore, 60 % of the non-fermented sugars are in the syrup fed to boiler, and the amount of digestion gas produced is independent of the process. The solid residual fraction of the middle term concept is slightly higher than the short and long term, as no sugar/cellulose is consumed for cellulase production. Washing out the extractives and acids accounts for the largest part of the 12 % energy loss.

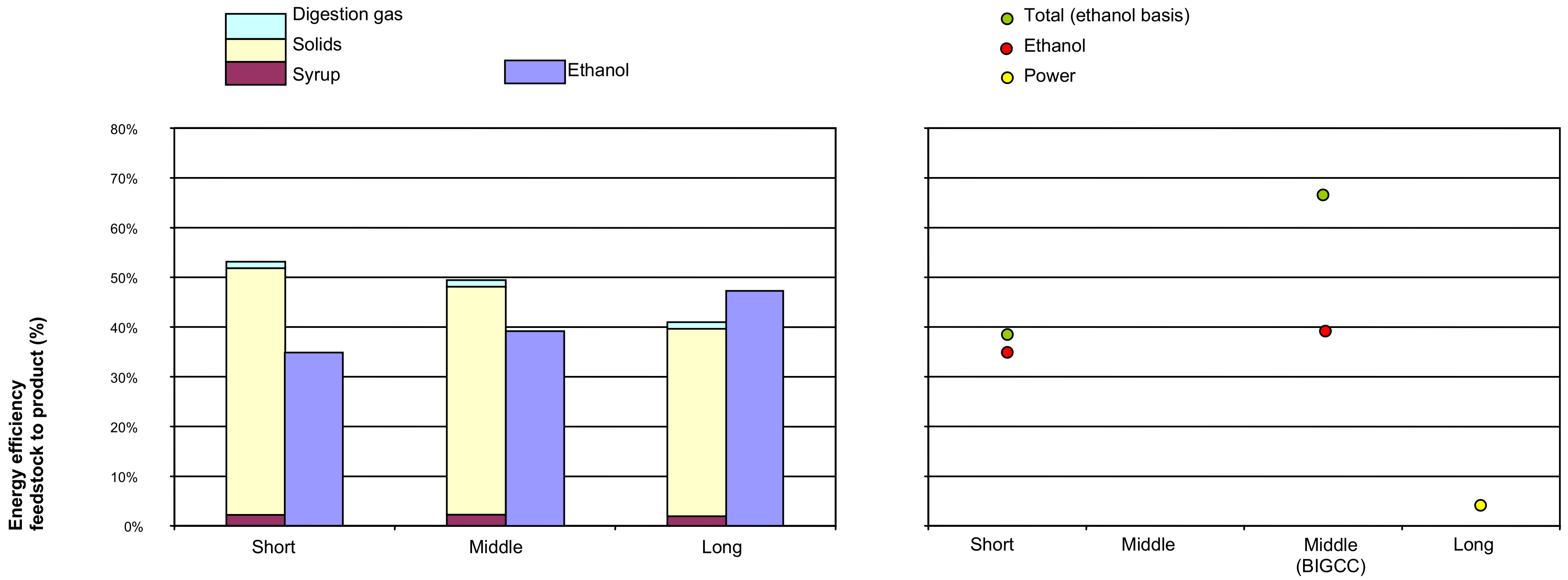

Figure 3. Energy efficiency from feedstock to ethanol and combustible rest streams (left) and to ethanol and electricity (right). Total conversion efficiency on ethanol basis assumes that the electricity was first produced from part of the biomass at $\eta = 45$ % and that all remaining biomass was converted to the ethanol. This total efficiency can be calculated by $\eta_T = \eta_{EtOH}/(1-\eta_E/\eta_{E0})$, where η_{EtOH} is the ethanol efficiency found, η_E is the electricity efficiency found, and η_{E0} is the assumed standard electricity efficiency, $\eta_{E0} = 45$ %.

The energy in the combustible co-products can only be recovered to a certain extent, by power generation. When electricity would be the desired product here, preferably a gasifier combined cycle set-up would be applied. However, several process units such as distillation in the short term and LHW hydrolysis (long term) require a large amount of steam, that can only be raised after a boiler. And the relative amount of residuals that can be used for steam raising decreases through time. Distillation in middle and long term concepts use less steam than distillation in short term concepts, because the steam explosion and LHW rest heat is deployed for part of the distillation. Therefore, only in the middle term concept, a BIG/CC configuration can be applied. The resulting efficiencies from biomass to ethanol and power are shown in Figure 3 (right). The total efficiency is expressed on ethanol basis, because the quality of energy in ethanol or electricity is not equal. This efficiency assumes that the electricity part could be produced from biomass at 45 % HHV in an advanced BIG/CC [37]. The total efficiency is about 38 % for the short term, and 52 % in the long term, the middle term concept that applies a BIG/CC for electricity generation performs best at 67 %. Lynd *et al* [14] calculated 50.3 % HHV efficiency (ethanol + electricity) using base case technology (SSF), 61.2 % for advanced case, and 69.3 % for best-parameter case. Their short term concept already produces ethanol against 46 %, which is about the same as our long term concept, and quite high considering that the theoretical maximum is about 52 %.

Figure 4 shows that, while efficiencies gradually increase from short to long term, the relative investments decrease. The specific investments are found to amount 2.1 k€/kW$_{HHV}$ ethanol produced for the SSF based plant on the short term, and 1.2 – 1.6 k€/kW$_{HHV}$ for the middle term. This compares with the value found by Wooley *et al.* [13] for a 2000 tonne$_{dry}$/day (about 450 MW$_{HHV}$ in, η = 32 %, 145 MW$_{HHV}$ ethanol) grassroots facility costing 234 MUS\$$_{1997}$ (TCI + 10 % other costs) or 1.87 k€/kW$_{HHV}$

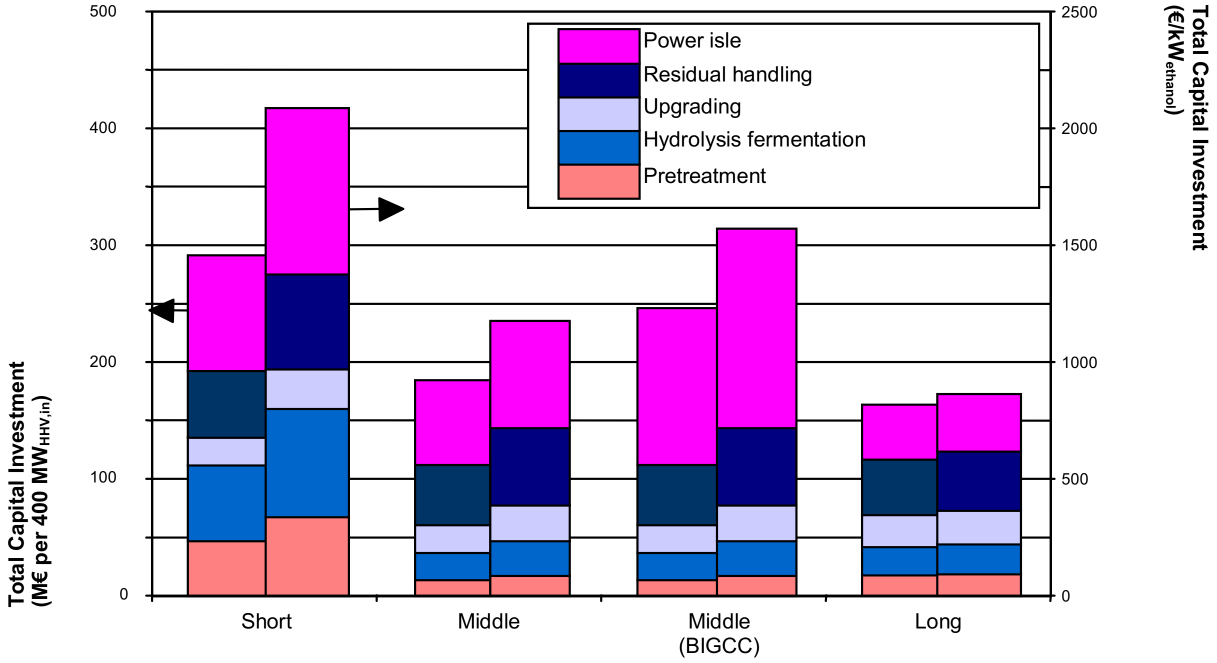

Figure 4. Total Capital Investment build-up. Left axis: normalised to 400 MW$_{HHV}$ input, but including scale effects for the larger concepts. Right axis, expressed in €/kW ethanol produced.

ethanol. Also from a technological viewpoint, the facility of Wooley *et al.* can be placed between the short and middle term concept of the present study. IEA/AFIS earlier estimated the investment to be 2.3 – 3.8 k€/kW$_{HHV}$ ethanol for weak acid hydrolysed processes, and about 1 k€/kW$_{HHV}$ for enzymatic hydrolysis [45]. The ultimate CBP plant studied here has specific investment costs of 0.9 k€/kW$_{HHV}$ ethanol produced.

While the efficiency of a BIG/CC equipped plant is much higher than a plant with boiler and steam cycle, the associated extra investment is also large. This results in higher ethanol product costs. Only when the co-produced electricity is valued higher than 0.05 €/kWh, the BIG/CC option becomes economically viable (middle term). For the assumed base case electricity reimbursement of 0.03 €/kWh, the ethanol product price in different stages of development is shown in Figure 5. The combined effect of more efficient and cheaper technology, larger scale, and cheaper feedstock, may decrease the ethanol production costs from 22 via 13 to 9 €/GJ. For the short term similar costs were found by IEA/AFIS [45], although their feedstock was cheaper (1.9 €/GJ$_{HHV}$). The cost levels are slightly higher than short and middle term values presented by Wooley *et al.* [15], but about twice as high as the ethanol cost path by Lynd *et al* [14], who presented production costs for SSF to amount 11 - 13 €/GJ, SSCF 7.6 – 9.8 €/GJ, and CBP 4.5 – 6.6 €/GJ. The difference may partly be explained by the high conversion efficiencies, and by more advantageous input parameters: the feedstock in Lynd *et al* costs 2.1 – 2.4 €/GJ, the input scale is 1.6 GW$_{HHV}$, and the electricity reimbursement 0.05 €/kWh. A large part of the remaining difference is caused by higher annual capital costs in the present study, and within the uncertainty of the factored estimation.

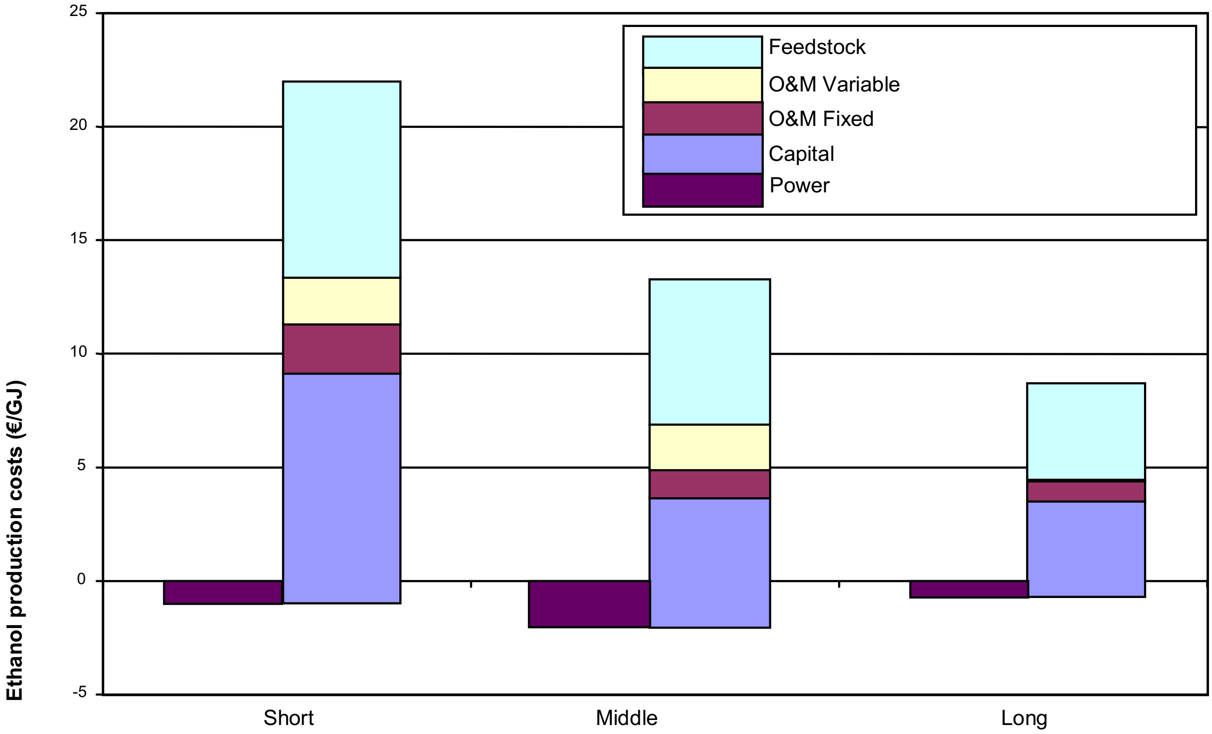

Figure 5. Break down of ethanol production costs. Electricity – co produced by a boiler / steam cycle – gives a negative contribution. Biomass feedstock costs decrease from 3 via 2.5 to 2 €/GJ$_{HHV}$, input scale increases from 400 via 1000 to 2000 MW$_{HHV}$.

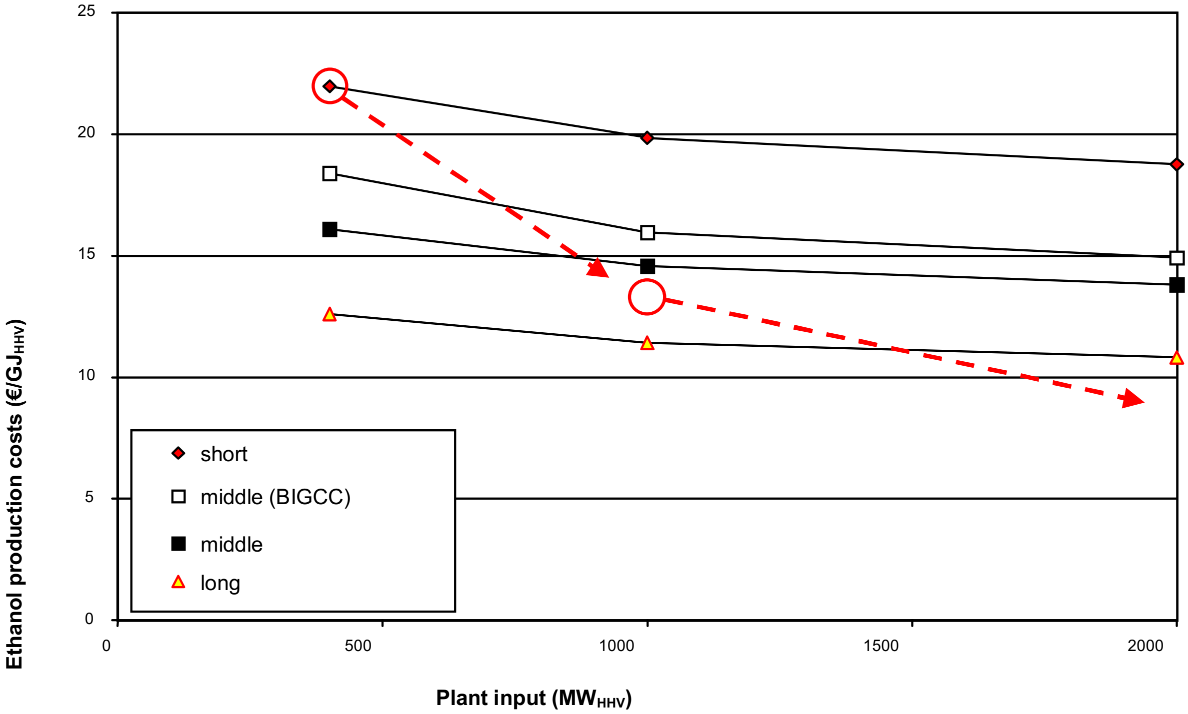

Figure 7. Sensitivity to scale (biomass costs are kept 3 €/GJ). The circles correspond to the production costs of Figure 5 (decreasing biomass costs 3 → 2.5 → 2 €/GJ).

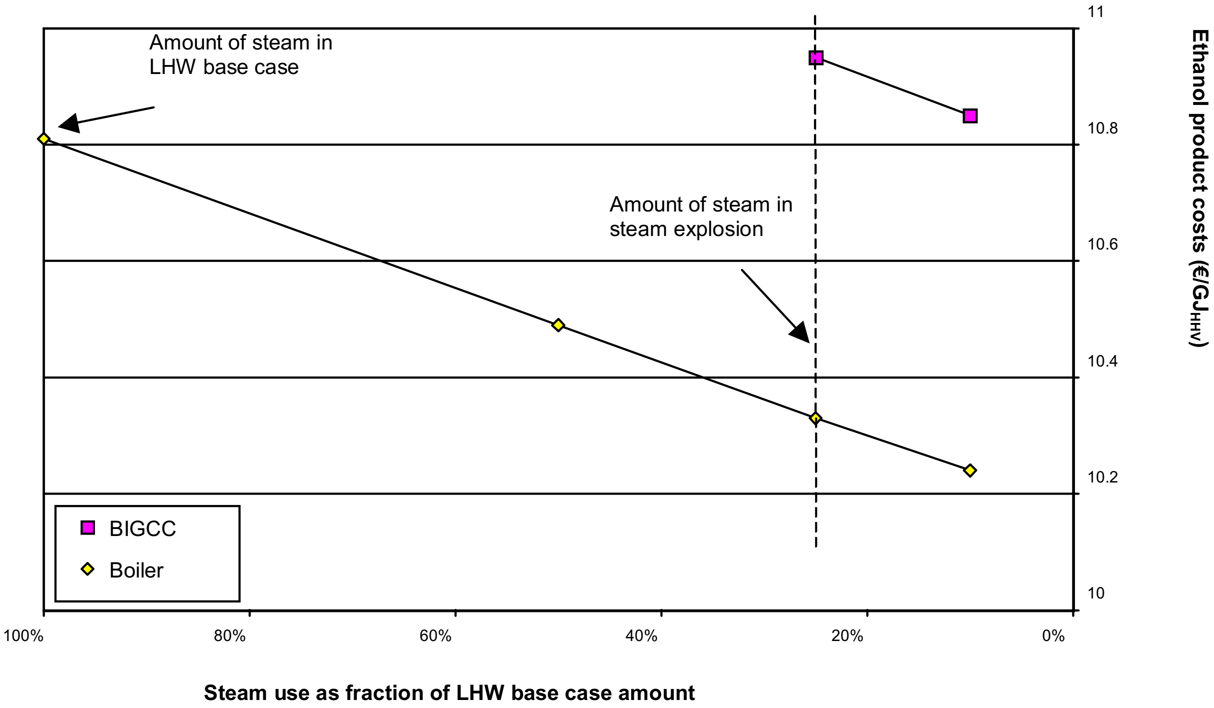

Figure 6. Influence of amount of steam used in the LHW process (long term), on the ethanol production costs.

The production costs decrease through time because of a number of reasons. Process improvement (higher efficiency, cheaper installation) is masked by other factors such as the larger scale, and the cheaper biomass feedstock. In Figure 7, the ethanol production costs for constant feedstock costs (3 €/GJ) is given, with the 400 – 2000 MW input scales applied on all concepts. Bare process improvement reduces the cost price with 27 and 22 % when proceeding respectively from short to middle and from middle to long term. The circles and dotted arrows indicate the combined effect as in Figure 7. Per 0.5 €/GJ increase in feedstock costs, the ethanol production costs increase with 1 €/GJ.

LHW in the long-term concept uses a large amount of steam compared to steam explosion in the middle term concept. This amount of LHW steam was not directly found in literature, but deducted from the difference in solids loading between both types of hemicellulose hydrolysis. If less steam is needed for the LHW process, then the amount of co-produced electricity will increase and the ethanol product price decreases. If just as much steam is needed as for steam explosion, then BIG/CC again becomes an option. Although this concerns a very large difference in steam use, the effect on the product costs are small (Figure 6).

When other feedstock than poplar is used, the amount of holocellulose and thus the ethanol yield will be different. The co produced amount of electricity depends on the amounts of solid residuals, syrup, and digestion gas as discussed earlier. In the pre-treatment, part of the feedstock energy is lost with washing out extractives and acids. In this perspective, switch grass, containing a large portion of extractives and acids, has a significantly lower (fuel+electricity) efficiency, which may increase the product costs by about 0.5 €/GJ. The differences between the other researched fuels are small. On the longer term the (hemi)cellulosic fraction may be increased by (genetic) plant cultivation. The price and available amount of biomass types are as yet of bigger importance to the feedstock choice.

6 Discussion and conclusions

The production of ethanol from cellulosic biomass has been evaluated for three stages of technological development. On short term (time indication 5 yr) a system may be realised that applies the well known dilute acid pre-treatment and where the different microbiological conversions all take place in different reactors. The middle term (10 - 15 yr) system may comprise steam explosion, which enables a better subsequent cellulose hydrolysis and a much smaller gypsum waste stream. Several conversions are then combined into fewer reactors. The ultimate (> 20 yr) facility may adopt Liquid Hot Water, allowing for higher yields of both hemicellulose and cellulose sugars, and all microbiological conversions take place in one reactor.

The total capital investments per kW_{HHV} installed ethanol production may decrease from 2.1 k€ (short term) via 1.2 – 1.6 k€ (middle) to 0.9 k€. This development includes both the cost advantages of the described technology development and an increase of scale. With the concurrent increasing biomass to ethanol conversion efficiency, the production costs are 22, 15 and eventually 11 €/GJ (biomass feedstock costs constant 3 €/GJ). Capital represents about 40 % of the ethanol production costs. The ethanol producing part (pre-treatment, hydrolysis, fermentation and upgrading) contributes about half of the total capital investment for the short and long term processes. However, the cost reduction (per installed kW_{HHV} product) for this section chiefly takes place when progressing from SSF to SSCF, and the further progress to CBP is of less importance to capital cost reduction. Biomass feedstock also represents about 40 % of the ethanol production costs. A 1 €/GJ cheaper feedstock results in 2.9 or 2.1 €/GJ cheaper ethanol on the short or long term. With 2 or even 1.5 €/GJ feedstock, the long term ethanol production price may thus decrease to 8.7 or 7.9 €/GJ.

The developments of pre-treatment methods and the ongoing reactor integration are independent trends. Steam explosion is situated between laboratory and pilot stage, and will most likely be commercially available on the middle term. Development of LHW is less evident: depending on technological breakthroughs and stimulation from the market, it may come commercially available earlier or later, than 20 years from now. This would mean either a more attractive ethanol product cost on the middle term, or a less attractive cost on the long term.

The integration of more conversions within fewer reactors is a more gradual development than suggested by the choice of concepts evaluated. The continuous development of (new) micro organisms improves the performance per reactor and may enable the combination of increasingly more functions within less reactors, but always as small steps in the progress from SSF via SSCF to CBP. These steps may often be possible using the existing capital assets. Furthermore the micro organism development itself may be approached via different paths by different players in the ethanol production field.

The results indicate that cellulosic ethanol on short term may be competitive with sugar/starch ethanol, but certainly not with sugar cane ethanol. Ethanol production, market penetration and cost reduction could go hand in hand with technology development and transition from traditional to cellulosic crops. The 13 and 8.7 €/GJ for the middle/long term are higher than values previously found by others. However, the difference is especially caused by the difference in electricity reimbursement and feedstock costs assumed. In any case, the ethanol produced unlikely reaches a cost level competitive with current fossil derived gasoline (production costs before tax 4 - 6 €/GJ [46]) or some other renewable motor fuels such as methanol from biomass (8 – 12 to eventually 5 – 7 €/GJ [17], bio-methanol has lower investment and higher efficiency, especially on the short term). Next to economic viability, the prospects for biofuel ethanol depend e.g. on its CO_2 reduction cost effectiveness and its ease of implementation, compared to other biofuels. Though ethanol is (together with FT) a more expensive option than methanol and hydrogen, it certainly has implementation advantages, as demonstrated in Brazil.

In further research the energy use and costs of the pre-treatment section (size reduction) needs more attention. More research is necessary to reduce the disposal of gypsum, or to determine the effect of waste disposal gypsum to the fuel price of an ethanol plant with dilute acid pre-treatment. The investment costs for steam explosion and LHW need to be assessed more exact. Also, more modelling work need to be done for the LHW reactor. The steam explosion pre-treatment is promising, but will require more development before sufficient conversion yields are guaranteed. The LHW conversion yields are high, but only determined on laboratory stage. Continuing development of new micro organisms is required to ensure fermentation of xylose and arabinose, and decrease the cellulase enzyme costs.

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Appendix

Abbreviations and glossary

bone dry - 0 % moisture
CNG - Compressed natural gas
DM - Direct methanol
EV - Electric vehicle
E10 - Blend of 10 volume % ethanol in gasoline
E100 - neat ethanol
FCV - Fuel cell vehicle
FFV - Flexible fuel vehicle
GHG - Greenhouse gas
HEV - Hybrid electric vehicle
HHV - Higher heating value
ICEV - Internal combustion engine vehicle (sometimes ICV)
ICEV-CI - Compression ignition internal combustion engine vehicle
ICEV-SI - Spark ignition internal combustion engine vehicle
LHV - Lower heating value
tC - tonne carbon emitted
VOC - Volatile Organic Compound
ZEV - Zero emission vehicle
DME - Dimethyl ether
MTBE - Methyl tertiary butyl ether
M85 - Blend of 85 volume % methanol in gasoline
ETBE - Ethyl tertiary butyl ether
oven dry - 10 % moisture
PEM - Proton Exchange Membrane aka Polymer Electrolyte Membrane
POX - Partial oxidation
RME - Rapeseed methyl ester
SNG - Synthetic natural gas
SR - Steam reforming
VAT - Value added tax

Resources and fuels characteristics

Table A 1. Resources and fuels characteristics, used in this dissertation [1-5].

	Density[1] **(kg/m^3)**	**Heating Value**[2] **(GJ$_{LHV}$/tonne$_{wet}$)**	**(GJ$_{HHV}$/tonne$_{dry}$)**	**Octane (R+M)/2**[3]	**Cetane**[4]	**CO$_2$ emission**[5] **(kg/kg$_{dry}$)**
Resources						
Coal[6]		21	24.5			2.2
Crude oil	845	42.7	45.4			3.1
Natural gas[7] USA	0.724 (gas)	47.8	53.1			2.7
Natural gas NL	0.781	40.5				
Biomass[8]		8.0	19.5			1.9
Fossil derived						
Gasoline[9]	740 – 750	43.2 – 43.7	47.3	90 - 100	5 - 20	3.2
Diesel[9]	810 – 860	41.9 – 43.1	45.2	n/a	40 – 55	3.2
LFO[10]	845	43.9	46.9			
HFO[11]	1013	40.0	42.1			
LPG[12]	525	46.1	48.0	102		3.0
Methane	0.658 (gas)	50.1	55.5			$^{44}/_{16}$
Biofuels						
Ethanol	791	26.4	29.8	100	8	$^{44}/_{23}$
Methanol	791	19.8	22.9	101	5	$^{44}/_{32}$
Hydrogen[13]	70.8 (liquid) 0.0848 (gas)	120	143	97		0
FT diesel	770	42.9	46.4			
Rapeseed oil[14]	910 – 922	37	39.4	n/a	40 – 51	3.2
Biodiesel RME[14]	880 – 920	37.3	39.8	n/a	51 – 58	2.9
Pyrolysis oil[15]	1110 – 1250	15.3	22.7	n/a	Unknown	2.0
HTU Bio-crude[16]		30 – 35	33 - 38			2.86
HTU Diesel[16]		44.4	47			3.19
DME[17]	665 (liquid)	28.2	29.8	n/a	55 – 60	$^{44}/_{23}$
DMM[18]	867	23.4	25.5	n/a	24	$^{33}/_{19}$
MTBE[19]	746	34.8	37.6	120+	n/a	$^{5}/_{2}$
ETBE[19]	745	36.2	39.0	120+	n/a	$^{44}/_{17}$

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[1] Density of the liquid form at standard conditions (1 atm, 15°C), unless stated otherwise.

[2] "The use of a Lower Heating Value (LHV) assumes that water vapour is not recovered but is emitted as a gaseous state in the flue gas, thus leaving the system. The alternative is the Higher Heating Value (HHV) which assumes that the water vapour, which originates from both fuel moisture and fuel hydrogen, is condensed and the heat of condensation is recovered." (Forsberg 1999). The following relations hold: $HHV_{wb} = HHV_{db} * (1 - W_{wb})$, $LHV_{wb} = LHV_{db} * (1 - W_{wb})$, $LHV_{wb} = HHV_{wb} - \Delta H_{H2O,evap} * (m_{H2O/H2} * H_{wb} + W_{wb})$, with wb = wet basis, db = dry basis, W = water content mass fraction, H = hydrogen content mass fraction, $\Delta H_{H2O,evap}$ = energy for evaporating water = 2.26 GJ/tonne, $m_{H2O/H2}$ = amount of water formed per hydrogen = 8.94 g/g.

[3] The octane number expresses the fuel's performance in a spark ignition engine, more precise the ability of the fuel to resist premature detonation and to burn evenly when exposed to heat and pressure. It is actually the maximum compression ratio at which a particular fuel can be utilized in an engine without knocking or self-ignition. The Research Octane Number (RON) simulates fuel performance under low severity engine operation. The Motor Octane Number (MON) simulates more severe operation that might be incurred at high speed or high load. In practice the octane of a gasoline is reported as the average of RON and MON or (R+M)/2, also called antiknock index (AKI) [2].

[4] The cetane number is a primary measure of fuel suitability for diesel engines (compression ignited). It essentially expresses the delay before ignition. The shorter the delay the better – and the higher the cetane number. A higher cetane number correlates with improved combustion, improved cold starting, reduced noise, white smoke, HC, CO and particulate emissions particularly during early warm-up phase. The minimum required is about 40.

[5] Only stoichiometric CO_2 emission in combustion, calculated from weight percentage C times 44/12. To calculate lifecycle emissions, energy use in production, transport and conversion should be accounted for.

[6] Average value (Greet/ANL). Bituminous/anthracite coal would be 27-30 GJ/tonne, and lignite/subbituminous coal would be 15-19 GJ/tonne.

7) Natural gas can be various gas mixes, depending on origin and time, it largely consists of methane (80 - 95 % by volume), further ethane, nitrogen, carbon dioxide and small amounts of ethane and carbon dioxide [2].

8) For indication only, more detailed data refer to biomass in the attached Papers. The (bulk) density is very dependent on the commodity (e.g. stem, pellet). The LHV is derived from the HHV (moisture content 50 % of total mass, hydrogen content 6 % of dry mass).

9) Composition and characteristics of gasoline and diesel differ per country.

10) Light Fuel Oil includes distillate fuel oils 0, 1 and 2, the data presented holds for fuel oil no.2.

11) Heavy Fuel Oil includes distillate fuel oil 4 and residual fuel oils 5 and 6, here data for fuel oil no.6 is used.

12) Liquefied petroleum gas is a collective name for different mixes of petroleum gases. Data presented hold for the mix 70 % propane / 30 % butane [1].

13) Liquid hydrogen at –253 °C. The difference between MON 63 and RON 130+ is big, the performance of hydrogen in ICEVs is thus very dependent on engine speed and load.

14) The HHV is calculated from the LHV (hydrogen content rapeseed oil is 11.7 % by weight, RME 12.5 %) [1]. [2] The physical properties of DME are very similar to the properties of LPG. Pressure is at least 5 bar to keep DME in liquid phase.

15) Pyrolysis liquids properties vary considerably depending on feedstock, process, and product recovery. Lower heating value for mc 26 % (11 % from the dry feed, rest from 10 % moisture in the feed). Organics yield 62.4 % of dry feed [6]. Hydrogen 6.6 %, carbon 55.5 % of dry product. Cetane number was not found, although many other characteristics were given by Gust et al. [7].

16) The organic “biocrude” readily separates from the water, therefore it is assumed that the crude contains zero water. At room temperature it is a solid, and it becomes a liquid at about 80 °C. LHV_{dry} is 30 – 35 $GJ/tonne_{dry}$, H content 12 – 20 wt% (as measured at the TNO MEPpilot plant, Apeldoorn the Netherlands [8]). Earlier it was reported that LHV may be up to 39 MJ/kg (presumably dry basis), water content 3 – 15 % [9]. After hydrotreating, hydrodesulfurisation and hydrocracking, 25 wt% of the product consists of gas oil that “can be expected to be a basis for premium diesel fuel” [8]. specific CO_2 emission, and diesel HHV derived from ADL [10], LHV diesel calculated by assuming H = 13.1 % by weight.

17) Some of the physical properties of dimethylether (DME: CH_3OCH_3) are very close to those of LPG. The vapour pressure is 5.1 bar at 20 °C [11].

18) Dimethoxymethane ($CH_3OCH_2OCH_3$) has many trivial names, most notably methylal. LHV calculated from HHV [12].

19) The HHV of methyl tert butyl ether (MTBE: $CH_3OC(CH_3)_3$) and Ethyl tert butyl ether (ETBE: $CH_3CH_2OC(CH_3)_3$) is calculated from LHV (H = 13.6 wt% and 13.7 wt% respectively).

Conversion factors

Table A 2. Unit conversions factors used in this dissertation [5].

Unit (trivial name)	Equals SI unit
1 gal (US liquid)	= $3.785 \cdot 10^{-3}$ m^3
1 scf	= $28.3 \cdot 10^{-3}$ m^3
1 barrel	= 42 gallon = 0.1589 m^3
1 lb mass (“pound avoirdupois”)	= 0.4535 kg
1 ton (short, = 2000 lb mass)	= 907 kg
1 tonne	= 1000 kg
1 BTU (International Steam Table)	= 1056 J
1 t.o.e. (“ton oil equivalent”)	= 41.9 GJ
1 barrel oil (= 42 gallon or 0.159 m^3)	= 6.12 GJ_{HHV}
1 gal gasoline equivalent	= 133.38 MJ_{HHV}
1 litre gasoline equivalent	= 35.24 MJ_{HHV}
1 MWh	= 3.6 GJ
1 BTU/gal	= 279 kJ/m^3
1 BTU/scf	= 37.3 kJ/m^3
1 mile	= 1.61 km
1 psi	= 0.069 bar
1 €/GJ_{HHV}	0.0352 €/l gasoline 0.0377 €/l diesel

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Curriculum Vitae,
Selection of publications,
Dankwoord

Curriculum Vitae

Carlo Hamelinck was born (February 25, 1975) and raised in Hulst, Zealand Flanders. Here, he attended secondary school (RK Jansenius SG), and obtained the "atheneum" certificate of secondary education in 1993. From 1993 till 1998 he studied Chemical Technology at the Eindhoven University of Technology. In these years he did a trainee ship in Paris, was representative in the student council and active for the international students reception committee. He graduated on an exergy analysis of sewage sludge gasification followed by methanol production. Thus he was put in touch with biofuels.

From 1998 till 2003 he was junior researcher at the Utrecht University Copernicus Institute's department of Science Technology and Society. Next to the biofuels research presented in this dissertation, he was also involved in a study on co-firing biomass in a pulverised coal power plant, and in a study exploring the Dutch possibilities for Enhanced Coal Bed Methane, a method of underground coal dioxide storage combined with methane recovery from coal seams.

Carlo currently works as bio energy consultant at Ecofys, a company that offers research and consultancy services and product development in the (renewable) energy field.

Selection of publications

Academic journals

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