Resources, Conservation & Recycling 125 (2017) 218–232

Contents lists available at ScienceDirect

Resources, Conservation & Recycling

journal homepage: www.elsevier.com/locate/resconrec

Review

Towards a Multiple Input-Multiple Output paper mill: Opportunities for alternative raw materials and sidestream valorisation in the paper and board industry

Spyridon Bousios[a,b,*], Ernst Worrell[b]

[a] Centre of Competence Paper and Board, Ijsselburcht 3, 6825 BS, The Netherlands
[b] Copernicus Institute of Sustainable Development, Utrecht University, Heidelberglaan 2, 3584 CS, Utrecht, The Netherlands

ARTICLE INFO

Keywords:
Paper
Pulp
Non-wood fibers
Waste management

ABSTRACT

The paper and board industry (PBI) faces a series of challenges, ranging from shifts in the availability and quality of raw materials to the generation of large amounts of sidestreams whose disposal entails significant costs. The concept of the "Multiple Input-Multiple Output (MIMO) Paper Mill" is proposed here as an option for addressing these issues by introducing, on the one hand, flexibility regarding the types of fibre sources that can be used as raw materials and, on the other, a full utilisation of all fractions of the raw materials, including those that were so far considered to be sidestreams of papermaking. With regard to raw material flexibility, researchers have implemented various pretreatment and pulping methods on potential alternative, non-wood, fibre sources for the PBI, which can be found primarily in agro-industrial residues and plants specially cultivated for this purpose. Research on the conversion of various types of papermaking sidestreams into energy and material products has also been extensive, with the new products aimed at (re)use within both the PBI itself and other sectors. Given that technical aspects have gained the most attention so far, more focus should now be placed also on the economic and organisational sides of the concept. It is also crucial to start evaluating integrated MIMO cases, taking into account the interconnected effects that new raw materials have on the papermaking process and its sidestreams, instead of looking into isolated MI and MO examples.

1. Introduction

The paper and board industry (PBI) is becoming increasingly aware of the need for changes in its long-established modus operandi due to the increasing competition for natural resources and the pressure on all sectors of the economy to reduce their environmental impact. The implementation of the biorefinery concept has been proposed as such a change, aiming at the more efficient and complete use of biobased raw materials and sidestreams. A traditional definition of biorefinery refers to the "sustainable processing of biomass into a spectrum of marketable products and energy" (Cherubini, 2010), or "a facility integrating biomass extraction and conversion processes and equipment to produce fuels, power, heat and value-added chemicals" (Rafione et al., 2014). A PBI biorefinery can, however, encompass additional characteristics in the form of raw material reclamation from papermaking sidestreams for (re)use by the same facility or the cascading of unusable fractions from one facility's processes as feedstock for another of the same or a different sector. This vision comes thus also close to the Chertow definition of industrial symbiosis (Chertow, 2007): a physical exchange of materials, energy and by-products among traditionally separate industries for realising a competitive advantage.

In order to illustrate this possible future of the PBI we can introduce a new concept, the "Multiple Input-Multiple Output (MIMO) Paper Mill". This is a facility that can convert a variety of raw materials, including – but not limited to- wood cellulose and paper for recycling (PfR), into a wide range of end products and intermediates, including – but not limited to- paper and board products. The conversion of existing paper and board mills into MIMO mills could be seen as a necessary step for the PBI in order to overcome several of the challenges that it faces today. It entails, on the one hand, a measure of flexibility regarding the raw materials from which its products can be made and, on the other, the full utilisation of all fractions of the incoming raw materials, including those that have so far been considered as sidestreams of the papermaking process. This conversion into MIMO mills requires increased cooperation with other sectors in the economy (e.g. agriculture, chemical industry).

Several factors motivate the partial substitution of traditional PBI fibre sources – virgin wood fibres and PfR- by alternative raw materials

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[*] Corresponding author at: Centre of Competence Paper Board, Ijsselburcht 3, Arnhem 6825 BS, The Netherlands.
E-mail address: s.bousios@bumaga.nl (S. Bousios).

http://dx.doi.org/10.1016/j.resconrec.2017.06.020
Received 3 February 2017; Received in revised form 15 June 2017; Accepted 20 June 2017
0921-3449/

in the MI side of the MIMO concept. These may be related to raw material supply, including the costs thereof, or the quality of the end product. Starting with supply-related factors, the foremost concern is an increasing competition for biomass. Studying the impact of waste-to-energy on the demand/supply of recycled fibre indicates that a direct competition between the uses of PfR as fibre source and fuel could arise in the near future under most scenarios evaluated, leading to higher prices (Ristola, 2012). Competition for wood could also increase; the renewable energy targets of the European Union can create a worrisome mismatch between fibre demand and supply, with significantly increased prices for both wood and PfR as a result (McKinsey and Company Inc, 2007). Another important factor is the limits of paper recycling and what happens when these are approached. Some European countries are approaching the theoretical maximum paper collection rates (ca. 90%), which means that easy to reach and high-quality sources (industrial, commercial) have already been tapped, leaving growth potential only in more disperse, lower-quality sources (households) (Blanco et al., 2013). The influence of this on PfR quality is clear. In Spain, where collection rates increased by 10% between 2005 and 2008 with a strong contribution from households, an increase of unusable material content in PfR by 57% and of moisture by 25% was recorded during the same period. This practically meant that a typical newsprint mill would face additional annual costs for raw material and waste disposal exceeding 1.3 M€ (Miranda et al., 2011). Commingled collection systems for recyclable materials, where applied, can also create serious PfR quality issues (Blanco et al., 2013; Miranda et al., 2013). Potential solutions to such challenges could come in the form of improved PfR sorting (Blanco et al., 2013; Bobu et al., 2010), e.g. via increased automation, but this would require large investments. PfR quality has also been showing signs of deterioration in terms of higher ash contents and worse dewatering behaviour, with subsequent impact on the papermaking process and end product characteristics. Finally, recovered paper trade poses extra challenges; demand in Asia has by far surpassed local paper recovery, leading to massive imports from North America and Europe. A study of trade patterns (Arminen et al., 2015) has indicated that high-income countries could have very little control over demand for their own PfR, since the trade is driven by import demand, and that low transportation costs favour the export of PfR to Asia.

Moving to product quality-related factors, a development that could promote the use of alternative fibre sources is the lately problematic image of PfR as raw material for the production of food packaging. Mineral oils in particular have been in the spotlight with regard to their possible migration to foodstuff packaged in paperboard produced out of PfR (Biedermann and Grob, 2010; Lorenzini et al., 2010; Biedermann et al., 2011). Their origin is traced to printing inks and, depending on various conditions, they could migrate to the packed foodstuff in concentrations that far exceed the accepted limits by means of evaporation from the packaging and condensation on the content thereof. Given that the selection of only specific PfR types as raw material can be of limited value (Biedermann et al., 2011), the remaining solutions involve either introducing functional barriers in paper packaging or moving away from PfR for certain types of foodstuff packaging. Mineral oils are, in any case, one among several potential issues: a list of 157 hazardous chemical substances −49 of which were mineral oils- found in paper products and PfR has been compiled as a basis for a priority list of chemicals to be monitored (Pivnenko et al., 2015). 51 of these substances tend to remain in the solid matrix during paper recycling and can therefore end up in the new product, while 24 of these are classified as persistent and potentially bio-accumulating.

The management of sidestreams generated by the papermaking process −primarily during stock preparation and wastewater treatment- constitutes an important cost factor for the PBI, making technologies that could reduce sidestream management costs, or even make them profitable, very interesting. Reliable statistics about sidestream generation by the PBI are difficult to come by; in 2005 some 11 million tonnes of solid waste were generated in Europe (including from pulp production) and roughly 70% (7.7 million tonnes) thereof originated from using PfR as raw material (Monte et al., 2009). According to the same source, the utilisation of PfR results in 50–100 kg of dry solid waste per tonne of packaging paper production, 170–190 kg per tonne of newsprint production, 450–550 kg per tonne of graphic paper production and 500–600 kg per tonne of tissue production. Different paper mills, however, produce different amounts of sidestreams of varying compositions. Information about process water is even more scarce; as an indication, more than 70,000 dry tonnes of COD were contained in the process water of the Dutch PBI in the year 2008, when the sector's production volume was some 3 million tonnes, 80% of which was based on the utilisation of PfR.

The two main outlets of these sidestreams have historically been landfilling and incineration, although the significance of the former has been gradually decreasing owing to regulatory limitations in several European countries. In any case, both options entail significant costs for the sector, with recent information from Germany and the Netherlands indicating that disposing of solid sidestreams costs up to, or even more than, 100 €/t. Reducing these costs, or even turning them into profits, depends on the ability of the sector to utilise valuable components in the sidestreams by (re)using them internally or converting them to intermediates or products for other parties on the MO side of the MIMO concept.

This paper aims to review developments relevant for the transformation of the paper mill into a MIMO mill and to identify promising alternative inputs and outputs. The current level of knowledge regarding their technical and economic potential is to be examined, so as to provide a basis for further research. In the first part (MI) we are, therefore, looking into alternative sources of cellulose fibres for papermaking, while in the second part (MO) our attention turns to potential new products or intermediates, the production of which could utilise current papermaking sidestreams. MO possibilities only for paper and board mills will be examined, while opportunities for pulp mills, where the situation is completely different (e.g. availability and valorisation of lignin), fall beyond the scope of this work.

2. Multiple inputs opportunities

The potential alternative (i.e. non-wood) fibre sources for the PBI can be divided for the purposes of this article into two categories:

- Residues of the agro-industrial sector, including the food industry
- Plants cultivated as fibre sources

Exceptions beyond these categories are also possible, with an example being the production in the Netherlands of moulded fibre packaging (egg cartons) with grass from nature conservation areas partially substituting PfR (Anon, 2017a). In any case, regardless of this categorisation, alternative fibre sources have some common characteristics. Compared to softwood and hardwood, their contents of ash (silicate) appear to be higher, those of lignin lower, while cellulose contents are comparable (Judt, 1993). Lower levels of lignin indicate that their pulping may be easier and cheaper, while pulp mechanical strength is directly proportional to cellulose content (Ververis et al., 2004). Another factor in favour of such sources is the multitude of possible applications (green biorefinery, utilisation of agricultural residues after food production). This could lead to attractive business cases for the PBI, with low and stable prices for alternative fibres supported by the valorisation of all plant components. A common disadvantage, on the other hand, is the seasonal availability of such −mostly annual- plants, which means that ways of ensuring a steady, year-round fibre supply are necessary. Transportation issues may, further, arise due to the high volume and low density of non-wood fibre sources compared to wood or PfR (Ashori, 2006).

Fig. 1 summarises the multiple input opportunities for a MIMO mill.

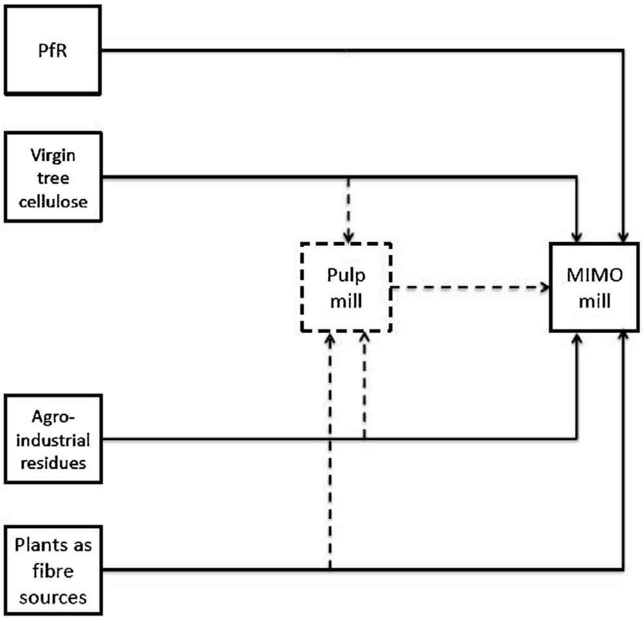

Fig. 1. Multiple inputs opportunities for a paper mill.

PfR and virgin cellulose from trees are currently the predominantly used raw materials, with PfR pulped on the site of the paper mill, while virgin cellulose pulp can be either produced by the paper mill itself or acquired from a pulp mill. To these possibilities are added the aforementioned agro-industrial residues and specially cultivated plants; in both these cases pulp can be produced at the site of the paper mill itself or centrally at a pulp mill serving more production locations.

2.1. *Residues of the agricultural sector*

2.1.1. *Wheat straw*

The most common varieties of wheat (*Triticum* spp.) have an average straw yield of 1.3 kg/kg grain (Iskalieva et al., 2012). Common applications of wheat straw (WS) include its uses as animal bedding and livestock fodder, soil improver, and mulch in the cultivation of vegetables and mushrooms. WS contains abundant holocellulose, little lignin, and a considerable amount of α-cellulose (Jiménez and López, 1993), a composition that, along with its availability, makes it interesting for papermaking purposes. Its outermost layer contains silica, is highly lignified and has predominantly short fibres or fines; its mechanical removal before pulping can improve pulp yields, bleaching, drainage, and decrease the silica content in black liquor. The depithing of WS can also improve pulp properties, especially drainage (Deniz et al., 2004). Mechanical fractionation into a chip fraction for papermaking and a meal fraction for animal feed or energy production is also possible (Papatheofanous et al., 1995); this also can improve delignification and pulp strength properties. The use of WS in papermaking is expected to produce environmental benefits. A Canadian study (Kissinger et al., 2007) found that the ecological footprint of WS chemical pulp was smaller than that of aspen or spruce pulps. WS use could further reduce harvest pressure on forest lands, with 0.6 ha of agricultural land relieving 5.5 ha of spruce forest or 2.3 ha of aspen forest.

A wide range of WS pulping techniques and papermaking applications has been examined. Organosolv processes have received considerable attention, including phenol (Jiménez et al., 1997), acetone (Jiménez et al., 1998), formaldehyde (Jiménez et al., 2000), ethanol (Jiménez et al., 2002a), ethanol-acetone (Jiménez et al., 2002b) and monoethanolamine (Salehi et al., 2014) pulping. A comparison of ethanol and formaldehyde WS pulping with conventional processes (kraft-anthraquinone [AQ], soda-AQ) demonstrated that organosolv pulps had lower strength and were difficult to bleach (Ates et al., 2008). Soda pulping of WS can produce a pulp that when mixed (10–50%) with PfR can match, or even improve, the strength properties of 100% PfR pulp (Guadalix et al., 1996). The enzymatic pretreatment of WS before soda pulping can significantly improve the burst and tear indices (Berrocal et al., 2004), while it can also reduce chlorine use in the Elemental Chlorine Free bleaching of soda-AQ pulp (Ates et al., 2015). The use of surfactants can improve soda pulping yields and pulp brightness (Mollabashi et al., 2011). A comparison of unbleached soda-AQ WS pulp with kraft eucalyptus pulp showed that although their average fibre lengths are similar, WS contains a considerably larger amount of fines. Pulp fractionation for fines removal improved strength and optical properties, as well as drainage, and made WS an interesting option for eucalyptus fibre substitution (Guo et al., 2009). Kraft pulping of WS has also been examined (Deniz et al., 2004; Ates et al., 2008; Levit et al., 2013). Partial hardwood substitution (10–20%) by WS in kraft pulp slightly increased pulp yield and significantly increased the tensile strength of unbleached pulp; in the case of bleached pulp, whose brightness can allow use in fine papers, tensile, burst and tear strength were all improved by WS addition. Enzymatic pretreatment was beneficial also for kraft pulping, reducing the kappa number, promoting internal fibrillation and improving bleaching characteristics. Small amounts (5–10%) of bleached chemical WS pulp can act as an inexpensive additive for high-yield pulps (e.g. hardwood bleached chemi-thermomechanical pulp) that have desired optical properties for printing and writing papers but lack in strength (Zhang et al., 2011). WS pulps have, finally, been produced with some unconventional methods. Cavitation pulping (Iskalieva et al., 2012; Badve et al., 2014) can be performed at room temperatures, thus saving energy, and produce fibres of sufficient quality for newsprint or packaging grades. Hot water pulping (Leponiemi, 2011), on the other hand, can be combined with peroxide bleaching for the production of writing and printing papers or boards that do not require a high brightness, or with mechanical refining for use in multi-ply board, moulded fibre packaging or fluting. Dissolved solids from the hot water treatment and fines from the fractionation of the mechanical pulp can, furthermore, be used for energy production.

2.1.2. *Rapeseed straw*

Rapeseed (*Brassica napus*) is widely cultivated as one of the most important sources of vegetable oil. Its main products are biodiesel, edible oil and animal feed. Rapeseed straw (RS) has a yield of 2.8–4.5 t/ha (Potůček et al., 2014) and no valuable applications, since it is too coarse for use as animal feed (Ahmadi et al., 2010). The fibre length of RS is similar to that of WS, while its overall fibre morphology resembles that of bagasse but with a shorter length. Its holocellulose content is higher than that of WS – containing, however, also more hemicellulose – and the two types of straw have comparable lignin contents; in terms of ash content, RS has the advantage (Hosseinpour et al., 2010; Mazhari Mousavi et al., 2013). Its chemical composition is also comparable to that of hardwoods, with the exception of its higher ash content (González et al., 2013a).

The use of RS for papermaking has so far received less attention than WS. Soda pulping has been studied (Potůček et al., 2014; Mazhari Mousavi et al., 2013; Potůček and Milichovský, 2011) and it was found that soda-AQ pulping delivers better results with regard to the kappa number and requires lower amounts of chemicals. Soda-AQ pulp from RS can have better strength properties (tensile, tear) than kraft pulps from poplar or willow. RS has also been subjected to chemithermomechanical (CTMP) processes (Hosseinpour et al., 2010; González et al., 2013a), with the strength properties of such RS pulp being much superior to those of pine thermomechanical pulp (TMP). These strength properties can be further developed by means of refining, but with accompanying drainability issues. RS CTMP after beating has strength properties that are adequate for the production of fluting or liner. Neutral sulphite semi-chemical (NSSC) pulping of RS (Ahmadi et al.,

2010) has been proven suitable for the production of pulp for unbleached paper grades.

2.1.3. *Sunflower stalks*

Sunflower (*Helianthus annuus*) is widely cultivated for its edible oil and seeds, but also used as bird food, livestock forage, and for biodiesel production. Its stalks, which have received some attention as a possible fibre source for papermaking, are pithy (25–30%), with the pith consisting mainly of problematic small parenchyma cells. Depithing is, therefore, necessary, while it also reduces the ash content of the stalk. The average fibre length of depithed stalks is within the range of hardwoods. Soda pulping of depithed stalks demonstrated that depithing also improves pulp yields. AQ addition leads to further yield improvements and better strength properties, producing pulp suitable as a mixing component in the production of printing papers (Khristova et al., 1998). Hydrothermal treatment of sunflower stalks has also been studied, aimed at removing substances that hinder delignification during pulping and converting hemicelluloses to a liquid sugars fraction next to a solid fraction to be subjected to pulping (Caparrós et al., 2008). The combination of hydrothermal treatment with organosolv (ethanol) pulping has led to lower pulp yields compared to soda processes; the pulp contained more cellulose and less lignin, but also had somewhat lower strength. Although the yield and pulp properties from soda pulping are better, the combination of hydrothermal treatment and organosolv pulping is more environmentally benign and has the added benefit of the usable sugars fraction.

2.1.4. *Vine shoots*

Vine shoots (VS) that are made available via the canopy management of vines (*Vitis* spp.) in viticulture have also been considered as a potentially important fibre source. 1.4–2 t/ha of VS are generated annually; the pulping of VS in Spain, for example, could produce an amount of pulp equivalent to ca. 30% of the annual Spanish pulp production (Jiménez et al., 2009). Compared to other agro-residues, such as WS, VS have a high holocellulose content (similar to that of pine (Jiménez et al., 2009)), but also more lignin and less α-cellulose (Jiménez and López, 1993). A comparison between kraft, soda and organosolv (ethylene-glycol and ethanol) pulping of VS has demonstrated that kraft pulping is more advantageous in terms of pulp composition and less in terms of yield. The lignin content of the pulp was consistently high, indicating that VS are not a good material for the production of bleached pulps (Jiménez et al., 2009). Soda pulp from VS, although containing fibres with relatively low aspect ratio, had acceptable strength properties. It exhibited furthermore a low amount of fines and good drainability, comparable to that of unrefined softwood pulps (Mansouri et al., 2012).

2.1.5. *Tree trimmings*

Trimmings of trees that are widely cultivated for their fruit, e.g. olive (*Olea europaea*) and orange (*Citrus sinensis*), are also being considered as alternative fibre sources for the PBI with millions of tonnes available annually in certain regions (e.g. the Mediterranean). The kraft pulping of olive tree trimmings has been studied (López et al., 2001; Díaz et al., 2005), while attention has also been paid to organosolv processes, such as ethanol and ethanolamine-soda pulping (Jiménez et al., 2001; Mutjè et al., 2005). Mixing, for example, organosolv olive trimmings pulp with kraft eucalyptus pulp can produce paper of acceptable quality, while reducing the energy requirements for refining. Biorefinery concepts have also been proposed, with the hydrothermal treatment of olive tree trimmings producing a cellulose- and lignin-rich fraction and a sugars-rich fraction; ethanol pulping of the former can deliver a cellulose pulp for papermaking and soluble lignin for other products. Furthermore, trimmings of smaller dimensions and leaves can be separated in advance and combusted as a cheap and sustainable alternative to fossil fuels (Requejo et al., 2012). This fractionation of trimmings into fractions for combustion and for pulp has also been studied in the case of orange trees, with the cellulose-rich fraction being subjected to soda-AQ pulping and producing pulps with physical properties equal to, or better than, those from other alternative fibre sources, such as WS (González et al., 2011, 2013b).

2.1.6. *Greenhouse waste*

Several Dutch projects have focussed on waste streams, i.e. stems and leaves, from the cultivation of tomatoes (*Solanum lycopersicum*) and bell peppers (*Capsicum annuum*) in greenhouses (Anon, 2016). Pulp for the partial substitution of PfR in packaging has been produced by means of mechanical processes (e.g. refiners) in industrial-scale trials. Aspects of these projects have included, next to the papermaking process itself, elements of the entire value chain, ranging from the removal of impurities from the raw material and its conservation for longer periods, to marketing questions regarding the introduction of such new packaging products, and the utilisation of plant juices that are a by-product of mechanical pulping.

2.2. *Plants cultivated as fibre sources*

2.2.1. *Switchgrass*

Switchgrass (*Panicum virgatum*), a perennial rhizomatous grass native to North America, is characterised by adaptability to a wide range of soil and climate conditions (including erosion-prone and marginal lands), limited requirements for fertilisation and weed control, and high crop yields (Goel et al., 1996; Lewandowski et al., 2003; Fox et al., 1999). In North America it is grown for soil conservation purposes, for fodder and as an energy crop (ethanol, electricity), while it is also found in South America and Africa as forage crop and worldwide as an ornamental plant. It is also possible to find switchgrass varieties that can adapt to most European regions and it appears that the plant may be grown further north in Europe than in North America (Anon, 2017b). Switchgrass has been considered an attractive alternative fibre source for the PBI due to its low lignin content, relatively low ash content and favourable fibre morphology; its chemical composition is similar to deciduous woods and its fibre length similar to that of typical hardwoods (Goel et al., 1996).

Kraft, soda, soda-sulphite and soda-AQ pulping have produced pulps with strength properties at the low end of typical hardwood kraft pulps, but better than softwood mechanical pulps, lower freeness than hardwood pulps and easy to bleach without loss of fibre properties (Goel et al., 1996; Law et al., 2001). Washing before pulping can reduce ash contents and thus improve process economics for chemical pulping (Madakadze et al., 1999). It has been proposed that chemical switchgrass pulp could substitute 15–20% of hardwood kraft pulp in fine papermaking (Goel et al., 1996), or be used for the reinforcement of paper made out of mechanical pulp (e.g. newsprint) (Law et al., 2001). Mechanical switchgrass pulp could also partially replace mechanical hardwood pulp in printing papers with a much more limited energy demand for its production (Anon, 2017b).

2.2.2. *Miscanthus*

Miscanthus (*Miscanthus* spp.), another perennial rhizomatous grass, has its origins in East Asia and is characterised by its adaptability, resistance to low temperatures and efficient use of water and nutrients (Iglesias et al., 1996). It has a high growth potential under European climate conditions (Central and Southern Europe) and can provide an annual harvest after reaching full development in the third year since its establishment. Establishment costs are a disadvantage for miscanthus. *Miscanthus x giganteus*, primarily considered for cultivation in Europe, is a sterile hybrid that does not form seeds; this means that establishment is performed by micro-propagation or rhizome cutting. Although machinery that can perform harvesting and immediate planting of rhizomes is being developed, development of seeds will be a more preferable option for the future (Lewandowski et al., 2003). After drying on the field over the winter the moisture content of miscanthus is reduced from 70% to 10–20%, with levels below 15% allowing long-term storage for year-round delivery. Compacting of the chopped plant would increase its density before further processing, while baling on the field is another way of reducing its volume for transportation (Venturi et al., 1998). 30–40% of miscanthus biomass is comprised of components such as epithelial and parenchyma cells that result in high fines contents in pulp and disintegrate even further during refining. This leads to higher drainage resistance and more frequent web breaks on the paper machine (Cappelletto et al., 2000; Thykesson et al., 1998). Mechanical pretreatment has been proposed for removing foreign matter (dust, sand, pebbles) and unusable material (leaves, pith, epithelial and parenchyma cells). Fractionation of beaten pulps could also reduce fines contents.

Encouraging results regarding the PBI potential of miscanthus are found in the literature. Pulp produced with the neutral sulphite-AQ process had strength properties comparable to fast-growing hybrid poplar, with a more limited use of chemicals for cooking and within about half the cooking time (Kordsachia et al., 1993). Soda pulping in moderate temperatures (80–100 °C) can produce high pulp yields (ca. 80%) (Iglesias et al., 1996; Marín et al., 2009), while relatively high pulp yields (55–60%) are also expected from organosolv pulping (Milox process) (Ligero et al., 2010). Alkaline and acidic miscanthus pulps have demonstrated a strength development upon beating similar to comparable birch pulps, unlike other alternative fibres (Thykesson et al., 1998). Interesting results have been produced when miscanthus is used for the partial substitution of PfR. Bleached miscanthus CTMP has had a positive effect on strength properties when mixed with PfR, while miscanthus TMP did not result in considerable strength benefits or losses; optical properties remained more or less stable in both cases (Cappelletto et al., 2000). Refined miscanthus soda pulp has shown good potential for substituting PfR in packaging paper production, where it could help reduce the amount of starch used for strengthening PfR-based paper products (Marín et al., 2009). In the Netherlands there is already some commercial production of writing paper out of miscanthus, as well as plans for use in substituting PfR in tissue production.

2.2.3. Reed canary grass

Another perennial rhizomatous grass that is considered as a potential raw material for papermaking is reed canary grass (RCG). RCG (*Phalaris arundinacea*) is native to temperate regions of the Northern hemisphere, where it is sometimes used as forage crop. Being already adapted to short vegetation periods and low temperatures it is the only viable grass option for countries such as Sweden or Finland (Lewandowski et al., 2003). RCG can be grown on most soil types, including marginal lands, provides high biomass yields for at least 10–15 years after its establishment and can produce twice as much pulp annually than temperate hardwood (e.g. birch) (Finell, 2003). RCG harvesting, similar to switchgrass, can take place either in the autumn or spring, but delayed spring harvesting offers several benefits. The plant's moisture content then is 10–15%, simplifying storage, while pulp yields are higher, fines contents lower, fibre lengths higher and ash and silica contents lower (Pahkala et al., 1997; Pahkala and Pihala, 2000). As with other non-wood fibre sources, ash and silica contents tend to be higher than in wood, making some form of fractionation before pulping beneficial. Removing leaves, dust and dirt by means of air fractionation can reduce silica contents by 40% (Pahkala et al., 1997), while mechanical fractionation has been proposed for producing stem chips for pulping and leaf meal for energy generation (Finell et al., 2002). Dry fractionation can reduce ash and silica contents by 40%, reduce the fines content, improve pulp drainage, increase the average fibre length and result in a 15% higher pulp yield (Finell, 2003). It has also been combined with briquetting of the fractions, raising the chip fraction's density from 100 to about 350 kg/m^3, or from raw material for some 5 t of pulp per truck to material for some 18 t (Finell et al., 2002). The delayed harvesting/fractionation combination can minimise the variation of RCG's quality as a raw material for papermaking to industrially acceptable levels, while quality variation is further reduced when varieties suitable for the soil and climate of the growing location are selected (Finell and Nilsson, 2005).

RCG has been primarily considered as a short fibre source for replacing hardwood in papermaking. In fine paper hardwood fibres improve properties related to printability (e.g. opacity, surface smoothness), with softwood long fibres offering structure. Fine paper made out of 30% pine bleached long fibre and 70% RCG as short fibre demonstrated no negative influence on dewatering/drying, no critical changes in base paper properties and better smoothness and optical properties (Pahkala et al., 1997). Offset printing tests of such coated and surface-sized paper showed furthermore printing characteristics comparable to those of wood-based paper (Finell, 2003). Hardwood fibre is also applied in white-top liner for a nice printing surface; the partial substitution of expensive bleached hardwood pulp by bleached RCG pulp actually led to an improvement of optical properties (Finell, 2003). Refining RCG pulp does not lead to any fibre strength development (Thykesson et al., 1998).

2.2.4. Giant reed

The last of the perennial rhizomatous grasses to be considered as papermaking raw material, giant reed (*Arundo donax*) is a native species in the Mediterranean region and has become dispersed into all subtropical and warm-temperate regions (Lewandowski et al., 2003). It is adaptable to various soil and climatic conditions, resistant to drought, has a high biomass productivity and requires limited irrigation and nutrient inputs.

The fibre dimensions of giant reed are close to those of eucalyptus and the plant demonstrates low lignin and relatively high ash contents (Shatalov and Pereira, 2006). The low flexibility of giant reed fibres is expected to have a negative effect on paper tensile and burst strengths and on folding endurance; compared to hardwood pulps it has lower mechanical strength, with the exception of a better tearing resistance (Ververis et al., 2004). On the basis of these, giant reed is expected to provide pulp mainly for newsprint or (in low contents) for higher-quality printing and writing papers. A comparison between kraft pulping of giant reed and various organosolv processes has demonstrated that the latter are able to produce higher pulp yields, with better brightness, viscosity and bleaching results (Shatalov and Pereira, 2006). Strength properties of organosolv pulps could be further improved via limited refining but anything more than that will result in a dramatic increase of drainage resistance. Bisulfite pulping has failed to produce a pulp that could be sufficiently bleached for use in tissue production, while kraft pulping and bleaching produced a brighter pulp with good tear strength that is adequate for the production of lower-quality tissue paper (Williams and Biswas, 2010).

2.2.5. Cardoon

Cardoon (*Cynara cardunculus*) is a herbaceous perennial plant that can grow with high productivity in dry and hot regions and unproductive soils (Gominho et al., 2001, 2011). Its traditional applications include animal fodder, consumption as a vegetable and use as a source of natural rennet in cheese production, while industrially it has potential as an energy crop, raw material for paper pulp, source of edible seed oil (physicochemically comparable to sunflower oil) and as source of pharmacologically active compounds (Fernández et al., 2006; Benjelloun-Mlayah et al., 1997). The biomass production period starts a year after establishment and may last longer than 10 years. A considerable advantage is that the harvested biomass is practically dry, simplifying transportation and storage. The time of harvesting is important for papermaking purposes, since over-mature plants have higher lignin contents (Antunes et al., 2000).

Pulping cardoon stems together with leaves leads to lower yields and darker pulps (Benjelloun-Mlayah et al., 1997), while depithing the stems can also be advantageous in terms of pulp yield, kappa number, average fibre lengths, longer fibres/fines ratios and strength properties

Table 1
Summary of multiple inputs opportunities described in this paper.

Type of alternative fibre source	Advantages	Disadvantages	Comments
Residues of the agricultural sector			
Wheat straw	Abundant material, favourable composition, wide range of pulping methods already tested, partial substitution of virgin fibres and PfR possible in various paper products	Larger amount of fines than in virgin pulps	Depithing can improve pulp yields and properties
Rapeseed straw	Composition and fibre morphology comparable to those of other fibre sources, strength properties better than some virgin pulps		Refining can improve strength properties, but influences drainability
Sunflower stalks	Fibre length comparable to hardwoods	Depithing is necessary	Production of sugars along with fibres has been studied
Vine shoots	Abundant in certain regions, low amount of fines	Less favourable composition compared to other alternative fibre sources	
Tree trimmings	Abundant in certain regions		Biorefinery concepts have been studied
Greenhouse waste	Partial substitution of PfR in packaging products possible		
Plants cultivated as fibre sources			
Switchgrass	Adaptable and easy to cultivate, favourable composition and fibre morphology, partial substitution of various types of virgin pulps in various paper products possible		Washing before pulping can be advantageous for ash removal
Miscanthus	Adaptable, low moisture content at harvesting, better response to refining compared to other alternative fibre sources, partial substitution of PfR possible	High establishment costs, large amount of fines	Pretreatment/fractionation can address the fines issue
Reed canary grass	Adaptable, low moisture content at harvesting, good potential for hardwoods substitution		Pretreatment/fractionation can address the fines and silica issue and improve pulp yields and properties
Giant reed	Adaptable and easy to cultivate	Low fibre flexibility	
Cardoon	Low moisture content at harvesting, good strength properties		Depithing can improve pulp yields and properties

(Gominho et al., 2001; Gominho and Pereira, 2006). The mechanical depithing of the stem is considered to be easy. Cardoon fibres have similar cross-sectional dimensions to those of eucalyptus but are ca. 30% longer. Unbeaten kraft cardoon pulp has high bulk and good strength properties (e.g. better tensile strength, burst index and stretch than eucalyptus) that can be further developed via refining (Gominho et al., 2001; Abrantes et al., 2007). The good strength properties of cardoon pulps indicate that they could be used in the production of board and corrugated board grades. Blending kardoon kraft-AQ pulp with unbleached kraft pine pulp has improved both burst index and ring crush test (RCT), while the concora medium test (CMT) of this cardoon pulp was better than pulps out of PfR or hardwood. On the basis of these, it is suggested that cardoon can be used in liner production and for reinforcing PfR pulps (Abrantes et al., 2007). The organosolv (ethanol) pulping of depithed stems has produced lower yields than kraft pulping (Oliet et al., 2005) (Table 1).

3. Multiple outputs opportunities

The sidestreams generated by paper and board mills that are considered are defined as follows:

- Rejects (ragger, heavy, coarse, fine); produced during the utilisation of PfR, they can contain fibre lumps, plastics, metals, sand and glass
- Deinking sludge; produced during the deinking of PfR, it contains mostly short fibres/fines, inorganic fillers, as well as ink particles
- Primary sludge; produced during mechanical process water clarification, it contains mostly short fibres/fines and fillers
- Secondary sludge; produced during process water clarification by biological means
- Process water (often referred to as wastewater); a key component of papermaking, it is usually treated on-site for the removal of contaminants

Given that the amount and composition of rejects largely depend on the quality of the utilised PfR, what is found in them can differ widely between paper mills. Purely as an indication, reject samples from the production of newsprint have been found to contain 28% plastic foils/adhesive tapes, 27% fibres/flakes, 24% hard plastics, and 7% each of metallic compounds, wood and textiles (Hamm, 2000). Rejects from the production of packaging paper, on the other hand, contained 61% plastic foils/adhesive tapes, 13% each of hard plastics and fibres/flakes, 7% metallic compounds and 6% textiles. The same source also offers some information about the composition of examined deinking sludge samples. When generated during the production of wood-containing graphic paper, this contained 37% clay and other fillers, 29% fines and printing inks, 19% calcium carbonate, 8% extractable compounds (e.g. resins, fats, soluble printing inks and adhesives, deinking chemicals) and 7% fibres. In the case of deinking sludge from the production of hygienic paper, the composition was as follows: 40% fines and printing inks, 26% clay and other fillers, 20% calcium carbonate, 11% fibres and 3% extractable compounds.

Making use of Cherubini's definitions of biorefinery products (Cherubini, 2010), the processes for the utilisation of PBI sidestreams are divided into two categories for the purposes of this article:

- Production of energy products, which are used because of their energy content, providing heat, electricity or transportation services
- Production of material products, which are used for their physical or chemical properties

Although this classification offers a starting point, it is not without shortcomings, given that some products (e.g. pyrolysis oil) could be utilised as energy sources and for the production of chemicals, thus spanning both categories.

Fig. 2 summarises the multiple output possibilities for a MIMO mill, which next to its traditional paper or board products can generate energy and/or material products from the sidestreams generated by the production process. It is also possible (see Chapter 3.2.10) to have some form of internal reuse of the sidestreams, without any other processing thereof.

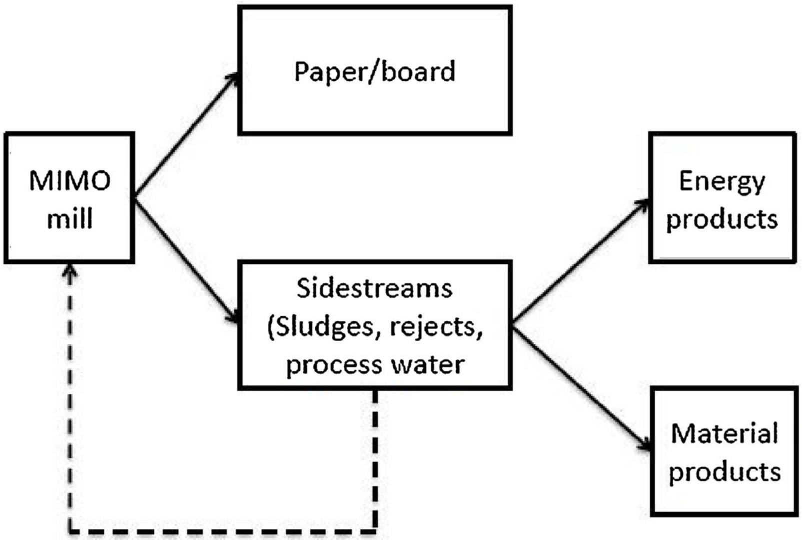

Fig. 2. Multiple outputs opportunities for a paper mill.

3.1. *Production of energy products*

3.1.1. *Incineration*

Incineration currently offers one of the main outlets for the management of PBI sidestreams. Its main advantages are the large reduction of the volume that needs to be disposed of and the potential for energy recovery, which can produce additional economic benefits in countries where "green" energy is subsidised. On the other hand, incineration requires large capital investments, the remaining ash must still find an outlet, measures for air emissions control are necessary, while the high moisture and inorganics contents of most sludges and rejects lower their heating value and make dewatering and/or drying prior to incineration necessary (Caputo and Pelagagge, 2001). Fluidised bed combustors have a higher efficiency for fuels with such high moisture contents, as well as lower emissions. The incineration of plastics-containing rejects requires taking also into account their chlorine content due to equipment corrosion and emissions considerations (Monte et al., 2009; Gavrilescu, 2008). Another possibility is the co-firing of PBI sludge, for example with other types of biomass or with coal (Tsai et al., 2002; Vamvuka et al., 2009; Coimbra et al., 2015).

3.1.2. *Gasification*

Gasification usually involves the partial oxidation of the feedstock by air, oxygen and/or steam for the production of synthesis gas, composed mainly of CO, CO_2, CH_4, H_2O and N_2, which is a more versatile energy carrier than heat and can also serve as a feedstock for the production of chemicals. This can be used in combustion engines or gas turbines, converted to liquid fuels, or reformed to hydrogen-rich gas. Considering that feedstocks with high moisture contents tend to produce gas with reduced calorific values and cannot sustain high-temperature thermal conversion processes, moisture must be first reduced to a maximum level of 15% by means of evaporative drying (Ouadi, 2012). Examples of research into PBI sidestreams' gasification are available in the literature (Cordiner et al., 2012; Ouadi et al., 2013a), with, for example, fixed bed gasification optimally applied in rejects of PfR-utilising mills, either with co-gasification of wood chips or with presorting of the rejects for the removal of some of the plastics content. The first commercial gasifier for the PBI will become operational in the Netherlands by 2016, processing 25,000 t of rejects annually in a fluidised bed installation; the produced syngas will be used for steam generation for the paper mill, reducing its natural gas consumption by 18 Mm^3/year (Elbersen et al., 2001).

A variation to the gasification theme is the supercritical water gasification (SCWG), which is actually a combination of thermal decomposition and hydrolysis. Next to converting organic material into synthesis gas, it also acts as a separation process, allowing the recovery of inorganic material. SCWG can be applied directly to wet sludge, eliminating the need for drying, and research into its applications for the PBI is ongoing (Zhang et al., 2010; Rönnlund et al., 2011).

3.1.3. *Pyrolysis*

Pyrolysis entails the thermal decomposition of organic matter in the complete absence of an oxidising agent, taking place in temperatures of 280–850 °C. Its three main types are slow, intermediate and fast pyrolysis, differing in heating rates and residence times, reaction temperatures and the relative yields of the solid (bio-char), gaseous (biogas) and liquid (bio-oil) products formed. As in gasification, pyrolysis also requires reduced feedstock moisture content (Ouadi, 2012). The pyrolysis of waste and biomass is a way of thermally upgrading to fuels with higher calorific values. Within the PBI the first installation for the pyrolysis of the plastic fraction of rejects from the recycling of beverage cartons as a fibre source was operational for a number of years in Spain, delivering next to the pyrolysis products also clean aluminium for recycling. Sludge pyrolysis, on the other hand, is still under development. Several examples of this can be found in the literature (Strezov and Evans, 2009; Lou et al., 2012; Ouadi et al., 2013b; Ridout et al., 2015); a way to integrate it to the energy system of a paper mill would be to use the oil as a fuel on-site and co-firing the gas and char for producing heat for drying the sludge and steam for electricity generation (Ouadi, 2012). Natural gas use for combusting the sludge would thus be eliminated. Pyrolysis oil from deinking sludge has some characteristics that could restrict its use in diesel engines, but these could be mitigated by blending with biodiesel (Yang et al., 2013). It could also be potentially converted to other liquid fuels or chemicals, but this may first require an improvement of its quality, for example by pretreating the sludge (Reckamp et al., 2014). The char could also find external applications in soil amendment or as fertiliser; in one example, char from deinking sludge has demonstrated environmental remediation potential on soil polluted with nickel (Méndez et al., 2014). The pyrolysis of sludge serves also as a separation technique, offering reclaimed inorganic fillers for reuse in the papermaking process (Lou et al., 2012), or by other users. Microwave pyrolysis, considered an improved fast pyrolysis method, has also been tested on PBI sludge (Jiang and Ma, 2011).

3.1.4. *Anaerobic digestion*

Anaerobic digestion (AD) is a series of processes during which microorganisms break down organic matter in the absence of oxygen. AD is more widespread in the PBI as a wastewater treatment option, especially in mills utilising PfR, due to the high COD levels of their wastewater, the presence of well digestible starch and the low concentrations of toxic/inhibitory compounds. Under such conditions AD can result in 58–90% COD removal, with production of 0.24–0.4 m^3 CH_4/kg COD removed (Meyer and Edwards, 2014). Applying AD for converting 25% of the COD available globally in the wastewater of the pulp and paper industry into biogas can result to the generation of 1–100 TWh of electricity annually (Meyer and Edwards, 2014). Anaerobic wastewater treatment should be combined with aerobic post-treatment for releasing into surface water an effluent of sufficient quality, with this combination generating lower amounts of sludge and requiring less space than aerobic treatment alone.

AD is furthermore gaining interest as an option to treat biosolids (waste activated sludge [WAS]) from aerobic wastewater treatment in the PBI. The main bottleneck is the hydrolysis of complex organic matter and research is focussed into overcoming this via mechanical, thermal, chemical or biological processes (Meyer and Edwards, 2014; Elliott and Mahmood, 2007; Yunqin et al., 2009, 2010; Stephenson et al., 2012; Tyagi et al., 2014; Kinnunen et al., 2015). The AD of WAS can reduce solid waste by 30–70% (Elliott and Mahmood, 2007), while also producing CH_4. It can also improve WAS dewatering, thus limiting the use of chemicals for this purpose (Karlsson et al., 2011). In mills where primary and secondary sludges are mixed for dewatering (e.g. prior to incineration), diverting the WAS to AD can improve mechanical dewatering efficiency, as well as incineration efficiency (Hagelqvist, 2013). AD produces also nutrient-rich reject water that could be recirculated to the aerobic treatment, reducing the need for nutrients addition, and digestate that could be further processed to commercial fertilisers.

3.2. Production of material products

3.2.1. Land spreading

The spreading of PBI sludge on forest and agricultural lands is a practice that has a long history in certain countries, while banned in others. In the UK, for example, more than 700,000 t (wet) were spread on more than 10,000 ha of agricultural land in 2003 (Gibbs et al., 2005). Its advantages include soil nourishing and conditioning, breaking down of pesticides, water retention in fast-draining soils and neutralisation of acidic soils when sludge rich in ash is applied (Monte et al., 2009; Scott and Smith, 2017; Abdullah et al., 2015). The main factors that determine its contribution to plant growth are the C:N ratio and the N loading of the sludge. Soil improvement has been demonstrated in some studies (Phillips et al., 1997; Rato Nunes et al., 2008), although it has also been suggested that crops should not be planted directly after sludge application so as to allow for a period of “equilibration” (Norris and Titshall, 2011). Sludge from deinking and PfR-utilising paper mills tends to have higher heavy metals loadings, but these are similar to, or lower than, those of manure or other organic materials applied in agricultural land (Gibbs et al., 2005). Some odour problems may exist, especially in the first days after application. Land spreading takes place during certain periods and therefore requires adequate sludge storage capacity.

3.2.2. Soil remediation

An alternative form of land application for PBI sludge is its use in soil remediation. A restoration of ecosystem functions is required in degraded sites (e.g. due to mining or industrial activities) that can poorly support plant establishment due to improper physical, chemical or biological soil properties. PBI sludge has been found to be a tool for the revegetation of degraded surface mine soils, enhancing the establishment and growth of plant cover (Fierro et al., 1999; Filiatrault et al., 2006). Furthermore, its application on metal-polluted soils can chemically stabilise the metals and decrease their mobility, and thus also plant uptake (Calace et al., 2005; Battaglia et al., 2007).

3.2.3. Landfill cover

The use of PBI sludge as landfill cover has been one of the more traditional outlets; in the US, for example, more than 29 landfills were closed between 1990 and 2003 using paper sludge as a hydraulic barrier layer, while many more have been using it as daily cover (Carroll, 2008). The geotechnical properties of PBI sludges resemble those of bentonite and compacted clays used as hydraulic barrier layers in landfills, while material costs are much lower (Nutini and Kinman, 1991; Moo-Young and Zimmie, 1997; Zule et al., 2007). When subjected to landfill conditions, PBI sludges do not undergo (bio)chemical changes and thus have no negative environmental impacts (Zule et al., 2007).

3.2.4. Composting

Composting, the solid-phase decomposition of organic matter by microorganisms under controlled conditions, can offer an alternative to the land spreading of PBI sludge, producing a marketable material for agriculture/horticulture. Traditional composting techniques are, however, limited by their space requirements, while the concentration of heavy metals can also be a limiting factor. Compared to land spreading, composting can effect organic matter breakdown prior to application, providing a better soil amendment; it can also have benefits related to odour reduction, the reduction of mass, volume and moisture content (reducing transportation costs), and improved storage and handling characteristics (Tucker, 2005). Co-composting PBI sludge with N-rich organic waste (e.g. kitchen and catering waste) can be mutually beneficial (Tucker, 2005), while composting of deinking sludge (Gea et al., 2005), and co-composting of mixed wastewater treatment sludge with fly ash (Hackett et al., 1999) have also been successfully presented in the literature.

3.2.5. Building materials

The incorporation of PBI sidestreams into the production of building materials has already been applied on industrial scale in various countries. An example of this is the use of sludge in the production of bricks. Addition of up to ca. 15% PBI sludge is feasible and, according to industrial experience, can be beneficial in terms of raw material substitution, reduced water use due to the moisture content of the sludge, and a small energy contribution to the process (Cusidó et al., 2015). The fibre content of sludge can also improve brick flexibility and thus reduce cracking (Černec et al., 2005). PBI sludge could be furthermore applied as a pore-forming agent, with brick porosity playing an important role in reducing thermal conductivity. Low-conductivity bricks can promote energy savings in the built environment and also have industrial applications (insulating firebricks) (Sutcu and Akkurt, 2009; Sutcu et al., 2012).

Cement and cementitious materials are another field of current and potential application. PBI sludges are used as feedstock in cement kilns, with the organic content supplying its calorific value and the inorganic ashes contributing to the production of clinker (Monte et al., 2009). Another possibility examined is the production of pozzolanic metakaolin, which can serve as an additive in cement, by means of controlled calcination (Pera and Amrouz, 1998; Frías et al., 2008, 2015). It has even been found that blending 20% of such calcined sludge with cement can improve the cement’s performance against freezing and thawing (Vegas et al., 2009). Other possibilities have also been studied, such as the use of sludge (Agulló et al., 2006; Yan et al., 2011) or sludge incineration ash (Ferrándiz-Mas et al., 2014) in cementitious masonry products (plaster, mortar), or the substitution of kraft softwood fibres in fibre-cement products by sludge (Modolo et al., 2011). A more “exotic” application is the hydrothermal production of a tobermorite-containing material out of sludge incineration ash for the immobilisation of Cs^+, Sr^{2+} and Co^{2+} ions in metallurgical industry waste and in nuclear waste containment with cement (Coleman and Brassington, 2003; Coleman et al., 2006).

Some research has also focused on application in concrete. Studies have focussed on partially replacing cement or fillers by PBI sludge (Ahmadi and Al-Khaja, 2001; Balwaik and Raut, 2011), or using residual fibres in sludge for the reinforcement of concrete (Naik et al., 2004). An interesting development is the potential use of sludge incineration ash that has been converted into a super-hydrophobic powder through low-cost surface functionalisation as an admixture or surface coating in concrete (Wong et al., 2015). This material can improve concrete durability by decreasing water absorption, while not affecting the evaporation rate, and thus leading to a dryer interior of the concrete structure. This improves resistance against various deterioration mechanisms, e.g. water-borne aggressive agents, freezing or steel corrosion.

Panel materials, used in construction, but also elsewhere (e.g. in furniture), provide another possible outlet. The use of sludge and rejects for the substitution of wood fibres in particleboard and hardboard is possible, although limitations with regard to the mechanical properties or the visual appearance of the product have been observed (Taramian et al., 2007; Tikhonova et al., 2014). Application of deinking sludge has also been studied in hardboard, softboard, medium-density fibreboard (MDF) and cement-bonded fibreboard (Goroyias et al., 2004). Another possibility is alternative wood adhesives. Secondary PBI sludge contains proteins and carbohydrates that are sources of bio-based adhesives. Sludge has, therefore, been studied as a co-adhesive together with common urea-formaldehyde resin in particleboard and MDF, offering significant reductions of formaldehyde emissions from the panel material (Migneault et al., 2011; Xing et al., 2013); formaldehyde is a suspected carcinogenic. Instead of applying untreated sludge, extracting intracellular proteins from it for use as adhesive has also been proposed (Pervaiz and Sain, 2011; Pervaiz, 2012).

3.2.6. *Natural fibre-plastic composites*

The plastics industry is commonly using fibres (e.g. fibreglass) and mineral fillers (e.g. clay, talc, calcium carbonate, silica) in order to adapt the mechanical properties of thermoplastic polymers or to decrease production costs. Natural fibres and inorganics found in PBI sidestreams can offer a lower-cost alternative to these materials and have thus gained the interest of researchers. Several studies (Son et al., 2001, 2004; Ismail et al., 2005; Girones et al., 2010) have demonstrated the feasibility of this application, with sludge sometimes even having a reinforcing effect. A special category of composite materials, Wood-Plastic Composites (WPC), offers further opportunities for the application of PBI sidestreams. WPC usually use wood flour as a fibre source, combined with a thermoplastic polymer matrix. Given that wood flour can be used for the production of fuel pellets, interest in substituting this material is substantial. A number of studies in this direction with encouraging results is available (Hamzeh et al., 2011; Huang et al., 2012; Soucy et al., 2014; Yang et al., 2015), although sidestream composition plays an important role in the impact of this substitution on the properties of the end product.

3.2.7. *Sorbents*

PBI sidestreams are being evaluated as a source of low-cost sorbents for the removal of organic and inorganic substances from aqueous solutions or in the gas phase, substituting more expensive options, such as activated carbon. Especially on the field of wastewater treatment, PBI sludge (or sludge ash) subjected to various forms of treatment (e.g. calcination, pyrolysis, physical activation), or even untreated, has been found to offer an alternative for the removal of metals (Calace et al., 2003; Méndez et al., 2009; Wajima and Munakata, 2011; Wajima, 2014), phenols (Calace et al., 2002), ammonium and phosphate (Wajima and Munakata, 2011; Wajima et al., 2007; Hojamberdiev et al., 2008; Wajima and Rakovan, 2013), and pharmaceuticals (Calisto et al., 2014; Jaria et al., 2015). Furthermore, PBI sludge can be the precursor for effective adsorbents for the removal of NO_2 or CO_2 (e.g. for biogas cleaning) in the gas phase (Hofman and Pietrzak, 2012; Espejel-Ayala et al., 2014). Next to the substitution of activated carbon, however, success has also been recorded in producing it out of PBI sludge (Khalili et al., 2000; Khalili et al., 2002; Li et al., 2011). PBI sludge can, finally, serve as absorbent for the removal of oil and other hydrophobic liquids from hard and water surfaces (Likon et al., 2011; Likon and Trebše, 2012) with such products being already on the market in the USA.

3.2.8. *Enzymatic hydrolysis and fermentation products*

PBI sludges have attracted interest as feedstock for the production of fuels and chemicals via hydrolysis and fermentation due to their significant carbohydrates content. In the development of 2nd generation lignocellulosic biofuels they have some considerable advantages compared to other biomass types (e.g. agricultural sidestreams). These include their currently low/negative value, availability in significant volumes at fixed locations, extensive prior physical/chemical treatment and low lignin contents that make them more accessible to enzymes without pretreatment, and availability of utilities and infrastructure in paper mill sites.

The production of ethanol has been widely studied. Simultaneous saccharification and fermentation (SSF) processes have been generally preferred over separate hydrolysis and fermentation (SHF), with SSF promising higher overall yields, shorter residence times and process integration with one reactor for both steps (Marques et al., 2008a); SHF offers optimal conditions for both steps, since they take place in separate reactors (Peng and Chen, 2011). Some examples of work with SSF can be found in (Lark et al., 1997; Fan et al., 2003; Yamashita et al., 2008; Boshoff et al., 2016), with SHF in (Peng and Chen, 2011; Sebastião et al., 2016), while (Marques et al., 2008a) offers a comparison of both on the same feedstock. Recovery of the mineral fillers in sludge for reuse can also be performed in the SSF residue (Fan and Lynd, 2007). A “quick and dirty” SSF has been proposed for converting the more easily accessible polysaccharides with a short residence time, while the residue is used for biogas production (Kemppainen et al., 2012). Although sludge pretreatment is not necessary, some research has demonstrated the benefits of chemical swelling and mechanical grinding (Yamashita et al., 2010), or of cationic polyelectrolytes addition and pretreatment by H_2O_2 (Gurram et al., 2015), on yields and production costs. Sludge fractionation for the removal of inorganic fillers could also be beneficial, since reactor sizes can then be reduced and enzyme adsorption on the fillers instead of on cellulose can be avoided (Chen et al., 2014a, 2014b; Robus et al., 2016). The production of ethanol can also deliver hydrogen and cellulases as co-products when a consolidated bioprocessing process is implemented (Moreau et al., 2015), while butanol can also be produced by SSF of PBI sludge (Guan et al., 2016).

Producing lactic acid (LA) has also been considered (Marques et al., 2008b; Budhavaram and Fan, 2009), with LA production –which is currently mostly based on the fermentation of starch-derived sucrose and glucose- expected to jump from 86 kt in 2001 to 1 Mt in 2020; producing such a speciality chemical may be more economically feasible than biofuels for the sludge quantities normally present at a single paper mill (Shi et al., 2015). Contrary to ethanol production, the presence of minerals in the sludge may be beneficial for LA fermentation, since these are typically added as buffering reagents (Budhavaram and Fan, 2009).

3.2.9. *Polyhydroxyalkanoates*

Polyhydroxyalkanoates (PHAs) are a group of polyesters that can be produced by certain microorganisms, where they serve as intracellular carbon and energy storage units. Due to industrially relevant properties comparable to those of commodity plastics, their biodegradability, and their production from renewable sources, they have been gaining increasing attention as a promising bioplastic material (Laycock et al., 2014). Their currently limited competitiveness is owed to high production costs due to the use of specific substrates and genetically modified microorganisms under sterile process conditions. The

Table 2
Summary of multiple outputs opportunities described in this paper.

Type of sidestream utilisation	Advantages	Disadvantages	Comments
Production of energy products			
Incineration	Sidestream volume reduction, energy recovery	Large investment, low calorific value of sidestreams	Availability of subsidies for "green" energy makes incineration more attractive, widespread practice
Gasification	Energy recovery, synthesis gas is a versatile product	Large investment, high-moisture sidestreams require drying	Applied in practice
Pyrolysis	Can serve also as a separation technique, energy recovery	High-moisture sidestreams require drying	Applied in practice
Anaerobic digestion	Energy recovery, AD of WAS reduces its volume	WAS must be pretreated before AD	Widespread wastewater treatment practice
Production of material products			
Land spreading	Can be beneficial for agricultural land	Odour problems, sludge storage necessary, banned in several countries	Widespread practice in some countries
Soil remediation	Can support revegetation of degraded soils		
Landfill cover	Similar performance –at a lower cost- compared to traditional materials		Applied in practice
Composting	Overall improvement over land spreading	Space requirements	
Building materials	Wide range of possible applications within various materials		Applied in practice for some building materials
Natural fibre-plastic composites	Possible to offer lower-cost alternatives to currently used raw materials		Sidestream composition plays an important role in determining what is possible
Sorbents	Wide range of possible applications for the removal of various substances at a lower cost than conventional solutions		Some examples of application in practice exist
Enzymatic hydrolysis and fermentation	Sidestreams more accessible to enzymes compared to other biomass types	Not clear whether amounts available at one paper mill are enough for economically attractive production of biofuels	Removal of inorganic fillers can be beneficial for product yields and process economics
Polyhydroxyalkanoates	Combining wastewater treatment with the production of a new product, possibility to have some influence on the characteristics of the end product, product with possible applications in papermaking		
Reuse	Partial substitution of papermaking raw materials	Possible negative influence on paper strength, drainage, biological activity in the system etc.	Some examples of application in practice exist

alternative of microbial community engineering, however, selects PHAs-producing microorganisms from the natural environment (Jiang et al., 2012) and much research is being directed in its application for the utilisation of industrial sidestreams as raw materials for low-cost PHAs production. PBI wastewater is one of these possible feedstocks (Jiang et al., 2012; Bengtsson et al., 2008; Jarpa et al., 2012), with the process aiming at efficient wastewater treatment next to PHAs production. Anaerobic fermentation of the wastewater can serve as pretreatment for converting organic matter into volatile fatty acids (VFAs) as substrate. The other steps of the process include the selection of PHAs-producing microorganisms and the accumulation of PHAs within them (Bengtsson et al., 2008). The composition of the VFAs influences the properties of the produced polymer, offering possibilities for regulating end product characteristics (Bengtsson et al., 2008). Downstream processing of the product begins with the extraction of the PHAs from the microorganisms, for which various physical or chemical methods are being considered. It is interesting to note that some early attempts are being made to utilise PHAs also as a raw material in papermaking, particularly as a surface sizing/coating agent (Laycock et al., 2014).

3.2.10. *Reuse*

The reuse of certain sidestreams with a high fibre content, such as fine rejects or primary sludge, by the PBI itself for the partial substitution of incoming raw materials can be a great step towards their valorisation. Primary sludge recycling is, in fact, not uncommon, although it may not be always possible within the same mill that generates it due to its effect on product quality; products with low optical properties requirements are preferable and low-ash sludge can be a proper raw material there, while high-ash sludge can be seen as a fillers source (Ochoa de Alda, 2008). Biosludge reuse has also been studied. Due to its composition it should have a detrimental impact on paper mechanical properties, but this can be compensated for by surface sizing with starch (Huber et al., 2014). The value of this concept depends on various factors, including the costs of sludge disposal, raw material and starch, and product requirements; additional factors to be considered are biosludge influence on biological activity within the mill system and on paper drainage. A more advanced biosludge reuse concept proposes its hydrolysis, with the solid fraction applied in board production and the liquid hydrolysate in biogas production (Kaluža et al., 2014).

Sludge fractionation is another concept for sludge reuse and overall valorisation in the best possible way. Reclamation of the filler fraction has already been industrially applied in some cases (Jortama, 2003), while current developments among PBI suppliers are moving towards sludge fractionation in fibres, usable and unusable fines, and fillers, with each fraction being subsequently –based on its properties- either internally reused or externally valorised.

3.2.11. *Other options*

A number of other possibilities have been less widely studied. An option that has been already applied in the PBI is the use of sludge as animal bedding (Scott and Smith, 2017; Villagrá et al., 2011) or cat litter (Monte et al., 2009). PBI sludge has been found to be a good option for the partial substitution of wood chips in pallets production (Kim et al., 2009). Paper sludge ash appears to be promising as a hydraulic binder in roadworks for soil stabilisation (Segui et al., 2012) and for the production of super-hydrophobic powders for use, among others, as coating in civil engineering infrastructure (Spathi et al., 2015). PBI sludge and wastewater are being considered as substrates for oleaginous microorganisms, and thus as cheap feedstocks for biodiesel production out of the lipids accumulated within them (Upadhyaya

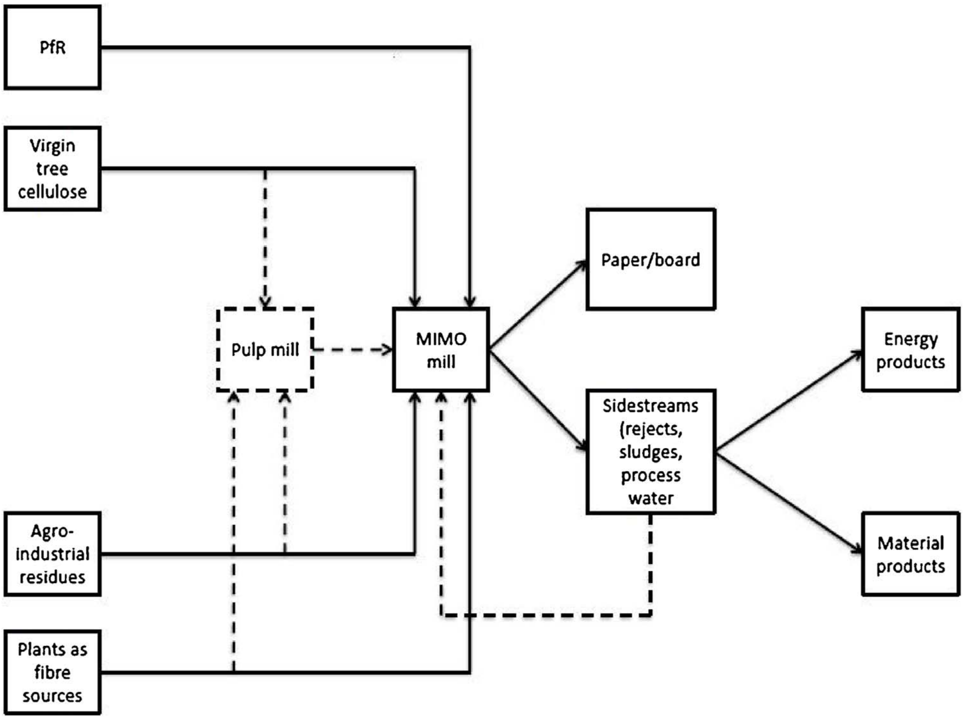

Fig. 3. Combined multiple inputs and multiple outputs opportunities for a paper mill.

et al., 2013; Zhang et al., 2014; Deeba et al., 2016). Production of hydrogen from PBI sludge has been proposed, either by means of enzymatic hydrolysis and fermentation (Kádár et al., 2003) or via AD (Valdez-Vazquez et al., 2005; Elsamadony and Tawfik, 2015). Acid hydrolysis of PBI sidestreams can be applied for the production of levulinic acid (Bozell et al., 2000; Schütt et al., 2013) or NanoCrystalline Cellulose (NCC). Carboxymethyl cellulose synthesis out of PBI sludge is another proposed route (He et al., 2009; Mastrantonio et al., 2015). In the field of wastewater treatment, an aerobic process based on granular, instead of floccular sludge, can −next to a reduced space and energy footprint of the treatment plant- produce a polysaccharide-based biomaterial (alginate-like exopolysaccharide) that could find an application in the surface sizing of paper (Lin et al., 2015) (Table 2).

4. Discussion

Combining the two sides of the MIMO mill as summarised in Figs. 1 and 2 Figure 1 allows us to see the concept in its full potential (Fig. 3). The paper- and/or board-producing mill utilises both traditional and new fibre sources, that are processed into pulp on its site or at centralised pulp mills supplying multiple locations, and makes its sidestreams available for conversion to energy and/or material products, with some internal reuse without any additional processing also being in some cases possible.

Several factors discussed in this paper (e.g. increasing competition for biomass, quality of PfR, costs of sidestreams disposal) make the MIMO mill concept a potentially important development for the future of the PBI. A large number of relevant possibilities for its practical implementation have received attention from researchers, as presented here. Many options regarding potential new raw materials for papermaking from plants specifically cultivated as fibre sources or from sidestreams of the agro-industrial sector, as well as new intermediate- or end products from the sidestreams generated by the PBI appear to be technically possible. Scanning the available literature, however, one is likely to come up with very limited indications about what could actually be feasible from an economic point of view. Kraft or soda pulping processes for producing alternative fibres on the MI side, for example, might be no obstacle for paper producers already applying these for wood pulping, but might constitute a prohibitively high investment for paper mills wishing to partially substitute PfR. This could, in turn, indicate that technical research should be re-oriented towards simpler (e.g. mechanical, mild chemical) processing methods for providing alternative fibres to certain segments of the PBI. Still missing economic analysis can also help establish which of the proposed ideas on the MO side could have real potential for application in practice, and under which circumstances. This is crucial information needed by PBI decision makers in order to help them select the most promising ideas to pursue. Another element necessary for the realisation of the MIMO concept in practice that will need to receive more attention, besides its technical and economic feasibility, is the establishment of value chains connecting actors within and across various sectors, who must also adapt their current business models based on the new opportunities and realities. Previous experience with such early-stage cooperative projects suggests that this point, although not as extensively discussed as the technical issues, is both challenging and crucial for the successful implementation of a promising idea. Finally, when looking at the MIMO concept in its full form, it becomes obvious that the utilisation of new raw materials by the PBI will also definitively have an impact on the amount and composition of its sidestreams. Using annual plants as a fibre source, for example, may significantly influence the quantity and quality of organic content in process water compared to using 100% PfR. Identifying what these interconnected changes may be and conducting integrated assessments of total MIMO cases will, therefore, also need to receive attention by researchers.

5. Conclusions

A transition of the PBI towards operating its paper mills as MIMO mills can help the sector address a variety of issues which it faces today, ranging from an increasing competition for biomass to the high costs associated with sidestream management. A MIMO mill has the advantages, compared to current standard practices, of being flexible with regard to the raw materials that it can utilise and of being able to direct all fractions of the incoming raw materials towards some form of valuable application. Research has been, as the present article demonstrates, extensive around both potential new raw materials for papermaking and new biobased products out of PBI sidestreams. In order for this – up to now primarily technical- research to become usable by decision makers in the sector, it needs to be complemented by insights into the economic and organisational aspects of the MIMO mill, which are currently scarce, as well as by research focusing on the MIMO mill as an integrated system, rather than simply on one possible new raw material or product at a time.

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