

13th International Conference on Greenhouse Gas Control Technologies, GHGT-13, 14-18
November 2016, Lausanne, Switzerland

Impact of fuel selection on techno-environmental performance of post-combustion calcium looping process applied to a cement plant

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Abstract

Calcium looping CO₂ capture is a promising technology to reduce CO₂ emissions from cement production. Coal is generally considered the fuel used to drive the calcium looping process as coal is already used as feedstock for cement production. This study assesses the impact of different fuels (coal, natural gas and woody biomass) on the technological and environmental performance of post-combustion calcium looping at a cement plant in North-western Europe. Process modelling is used to determine the impact of the different fuels on the mass and energy balance of the process. Life cycle assessment is carried out to evaluate the environmental performance of the different systems. Results indicate that firing natural gas or biomass instead of coal in an add-on calcium looping process can improve the efficiency of the process, as it decreases the fuel, limestone and electricity consumption. Consequently, while coal fired calcium looping can reduce life cycle climate change potential by 92%, the use of natural gas or biomass can make the process carbon neutral (reduction of 100%) or negative (-169%), respectively. Further research is required to complete the environmental perspective of using alternative fuels, but these results already illustrate a potential low-hanging fruit to improve the environmental performance of post combustion calcium looping in the cement industry.

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Peer-review under responsibility of the organizing committee of GHGT-13.

Keywords: cement plant; calcium looping; process modelling; LCA; biomass

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1. Introduction

Calcium looping is a promising CO₂ capture technology that can be applied to both power plants and industry processes. It is especially considered a favourable technology for the cement industry as they already have experience with solid handling, have in-place infrastructure for handling limestone and spent solids could potentially be utilized in the cement production process. Although integration of the calcium looping process with clinker production is shown to be more efficient [1], [2], post-combustion calcium looping can still be a valid retro-fitting option for existing cement plants [3].

Traditionally, coal is used for the production of cement due to its high heating value and the positive effect of coal combustion products such as fly ash on cement quality. Although an increasing amount of cement plants has started to co-fire less carbon intensive fuels, such as waste streams and biomass to reduce CO₂ emissions, coal is still the most dominant fuel used in cement production [4]. Consequently, coal is considered *the* fuel to support calcium looping processes at cement plants. However, the use of coal can have large repercussions for the environmental footprint of a cement plant as emissions associated with coal production and transport are reported to dominate the life cycle impact of calcium looping [5].

The performance of natural gas fired calcium looping has been studied in literature for natural gas fired power plants [6], [7] and natural gas fired industry processes [8], [9]. Furthermore, the feasibility of applying calcium looping to biomass fired power plants has been assessed in [10]. To date, no open-source literature addresses the performance of other fuels driving the calcium looping CO₂ capture process at a cement plant as it seems straightforward to use the same fuel that is already used for the production of cement. However, when post-combustion calcium looping to existing cement plants is considered, fuel handling capacity would need to be increased anyway, requiring additional investments. Therefore, coal does not necessarily need to be selected as the fuel driving the calcium looping in the decision making process.

This study aims to investigate (i) what the impact will be of using different fuels on the technical configuration and performance of the capture system and (ii) whether, and if so by how much, using low carbon intensive fuels might provide a low-hanging fruit to improve the environmental performance of calcium looping in cement plants.

2. Methodology

2.1. System boundaries

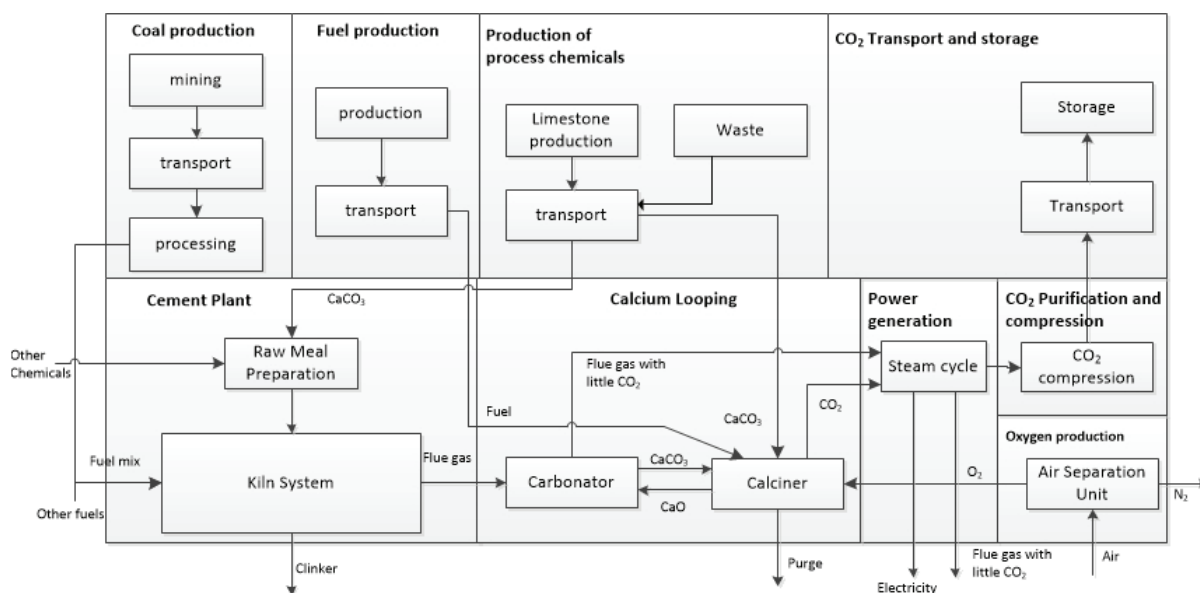


Figure 1. Simplified process flowsheet of clinker production with post-combustion calcium looping.

Figure 1 presents a simplified flow sheet of system boundaries and process flows included in this study. This study considers a cement plant located in north-Western Europe producing around 1 million tonnes of clinker per year. The kiln system in the cement plant under study is fired by a fuel mix comprising coal (> 50%) and various types of (biogenic) waste [11]. The flue gas of the kiln system contains c.a. 18%_{vol.} CO₂ [11]. 85% of the CO₂ is captured from the flue gas via calcium looping using limestone. A small share of the solid sorbents is purged after the calciner to keep the content of calcium sulphate and unburned limestone below critical levels. The heat required for the carbonator and calciner is provided by the fuels, and the available heat from flue gas and CO₂ stream is used to produce electricity via a steam cycle. The produced electricity is assumed to replace general electricity production in the LCA. Oxygen demanded by the calcining process is provided via available air separation technology and the captured CO₂ is compressed to 110 bar, transported and stored in an offshore aquifer.

Three different fuel types are considered to drive the calcium looping process, namely: coal, natural gas and woody biomass. Characteristics of the selected fuels are presented in Table 1. A reference case in which no CO₂ capture is applied is also included in the analysis.

Table 1. Coal and biomass composition

<i>Component</i>	Mass fraction (%) in coal ¹⁾	Mass fraction (%) in biomass ²⁾	<i>Component</i>	Mole fraction (%) in natural gas ³⁾
<i>Water</i>	1.2	3.5	<i>CH4</i>	83.2
<i>Ash</i>	14.3	1.6	<i>C2H6</i>	3.7
<i>C</i>	71.7	47.0	<i>C3H8</i>	0.6
<i>O</i>	5.9	41.9	<i>C4H10</i>	0.2
<i>H</i>	3.9	5.6	<i>C5H12</i>	0.4
<i>Cl</i>	0.1	0.01	<i>CO₂</i>	1.0
<i>N</i>	1.7	0.4	<i>N</i>	10.9
<i>S</i>	1.2	0.04	<i>S</i>	0.0
<i>HHV (MJ/kg)</i>	27.2	19.9	<i>HHV (MJ/kg)</i>	51.5

1) Based on the coal type used at the cement plant under study [11].

2) Wood pellets produced from residues [12].

3) Natural gas composition of European benchmarking task force [13].

2.2. Approach

The general approach of this study is depicted in Figure 2. The technical assessment is carried out by developing an Aspen+ process model which includes the calcium looping process, electricity production and CO₂ compression. Results of the process model are used to carry out a life cycle assessment (LCA) to assess the environmental performance. The inventory of the LCA comprises the results of the technical modelling and data on up- and downstream processes taken from the Ecoinvent database [14]. The functional unit of the LCA is 1 kg of clinker produced by the cement plant.

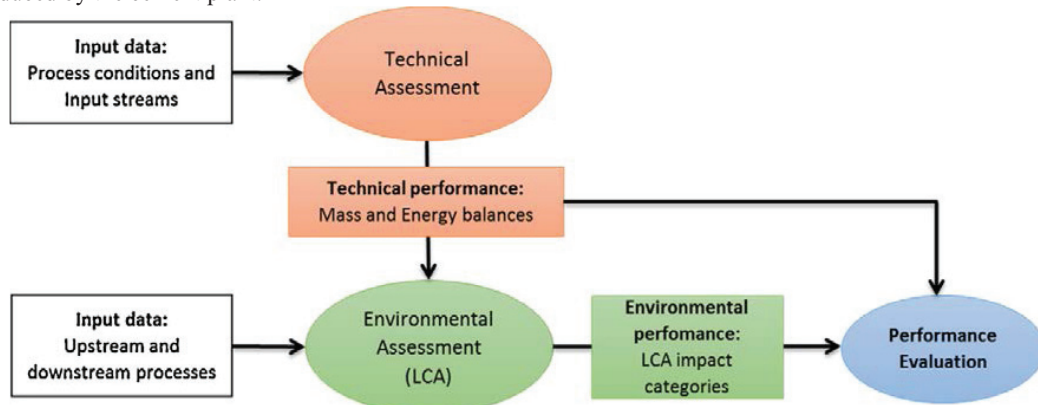


Figure 2. Schematic overview of general approach [18].

3. Results

Table 1 presents technical results for the calcium looping process for the three fuels considered. In all cases, 85% of the CO₂ (0.74 kg/kg clinker) is captured (nominal) from the flue gas of the kiln system. However, as part of the CO₂ formed by the combustion of fuel needs to be captured as well, the real capture ratio (the total amount of CO₂ that is captured from the system) exceeds 85%. As coal is the most carbon intensive fuel, relatively more CO₂ is formed during combustion and a higher capture ratio is required to avoid 85% of the CO₂ emissions. However, CO₂ purity slightly decreases when using natural gas or biomass due to the different composition of these fuels, due to a higher O₂ (biomass) or N₂ concentration (natural gas).

Table 2. Technical performance results

Parameter	Coal	Natural Gas	Biomass
CO ₂ avoided (kg/kg clinker)	0.74	0.74	0.74
Nominal CO ₂ capture ratio (by default)	85.0%	85.0%	85.0%
CO ₂ captured (kg/kg clinker)	1.42	1.16	1.30
CO ₂ purity (mass)	98%	96%	97%
Real CO ₂ capture ratio	91.6%	90.0%	90.9%
F0/FCO ₂ ¹⁾	0.11	0.08	0.09
Fr/FCO ₂ ²⁾	4.25	4.25	4.25
Thermal input calciner (MW)	0.21	0.18	0.19
Fuel consumption (kg/kg clinker)	0.21	0.14	0.26
Limestone consumption (kg/kg clinker)	0.22	0.15	0.18
Oxygen consumption (kg/kg clinker)	0.48	0.46	0.37
Ash production (kg/kg clinker)	0.030	0	0.004
Purge (CaO and CaSO ₄) (kg/kg clinker)	0.12	0.09	0.10
Gross Electricity production (MW)	52.1	51.9	51.6
Electricity consumption (MW)	32.2	28.5	28.0
Net Electricity production (MW)	19.9	23.4	23.6

1) Fresh limestone molar ratio (kmol CaCO₃ in fresh limestone/kmol CO₂ in flue gas)

2) Circulating lime molar ratio (kmol CaO in circulating solids/kmol CO₂ in flue gas)

The limestone to CO₂ ratio in the process (Fr/FCO₂) is kept constant in all cases, but the limestone make-up to CO₂ ratio (F0/FCO₂) slightly differs due to different limestone consumption values in each case. Limestone consumption is mostly affected by the amount of CO₂ in the system and the purge ratio. A lower purge ratio is required for the natural gas and biomass calcium looping processes due to the lower Sulphur content in these fuels. As the total amount of CO₂ is also lower in the system, the limestone consumption is considerably reduced in the biomass and natural gas driven calcium looping processes. As a result, the required fuel (thermal input to the calciner) also slightly decreases when firing natural gas or biomass instead of coal.

Firing biomass noticeably reduces the amount of oxygen required due to the relatively high oxygen content of biomass. Electricity production only differs slightly per case, however, electricity consumption is reduced in the natural gas and biomass cases as a result of electricity savings in CO₂ compression and O₂ production, respectively.

To assess the environmental performance, climate change potential is used as indicator (expressed in kg CO₂ equivalent per kg clinker). Figure 3 shows the results for the life cycle assessment of the calcium looping processes for each fuel as well as the reference case in which there is no capture of CO₂. Results are divided into contributions from coal production and transport (for clinker production), production and transport of additional fuel for the calcium looping process, clinker production (direct emissions), oxygen production and CO₂ compression, transport and storage. Besides, negative contributions in the form of electricity production (assuming regular electricity is replaced) and CO₂ captured from biomass (assuming that biomass is CO₂-neutral) decrease the CCP impact.

Due to the CO₂ capture and the avoided emissions from regular electricity production, CCP is reduced by 92%, 100% and 168% for the coal, natural gas and biomass cases, respectively. The larger reduction for natural gas compared to coal is mostly the result of lower emissions during natural gas production and transport and a reduction in CO₂ compression electricity consumption. The substantial reduction in the biomass case is predominately caused by the capture of CO₂ from the biomass (negative emissions), although reduced electricity consumption for the production of O₂ also contributes.

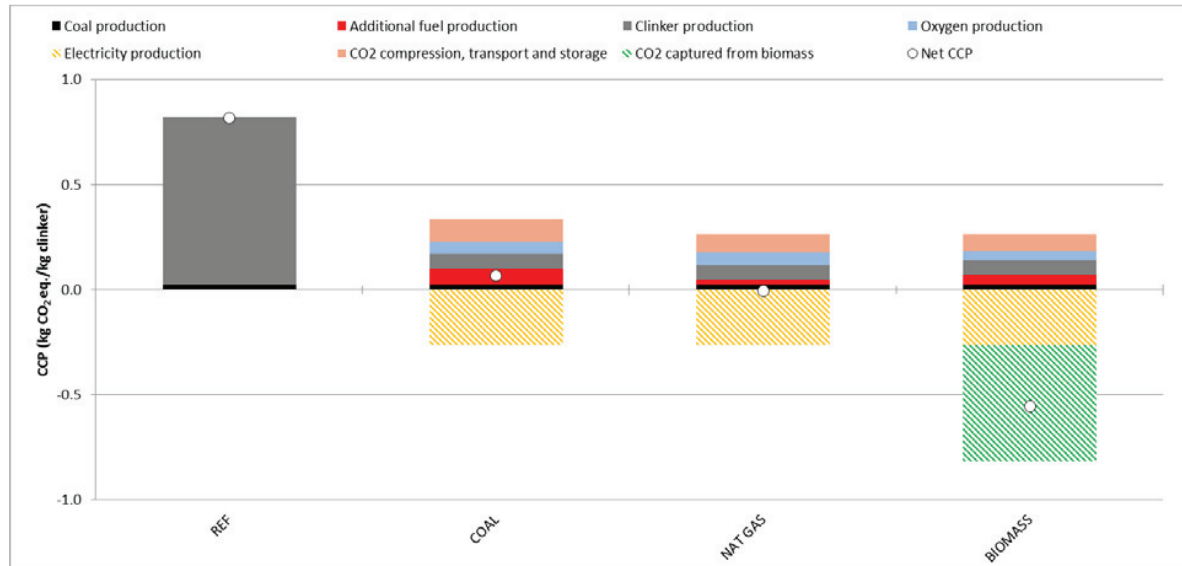


Figure 1. Climate Change Potential (CCP) for the reference case without Ca-looping (REF), Ca-looping fuelled by coal (COAL), calcium looping driven by natural gas (NAT GAS) and Ca-looping fuelled by biomass (BIOMASS).

4. Discussion and Conclusion

This study aimed to assess the technical and environmental performance of a calcium looping process at a cement plant for different fuels. Using a different fuel significantly alter the mass and current results already indicate climate change reduction potential of using natural gas or biomass as a fuel for post combustion calcium looping instead of coal. Assumptions and simplifications applied in this study are addressed here to better understand the results and to highlight potential areas for further research.

To start with, process optimization by heat integration with the cement plant was not carried out. Complete integration of the clinker production process and the calcium looping process is shown to drastically improve the efficiency of calcium looping [1], [3]. In the case of add-on calcium looping, integration of heat streams might also substantially improve the efficiency of the process, especially taking into account the large amount of heat available in clinker production and calcium looping.

An important assumption was that the purge taken from the calcium looping processes (CaO with small amounts of CaSO₄) is free of impact. The purge seems to be too valuable to be considered a waste stream as it has been shown that spent sorbent can be used as a limestone replacement in cement production [15]. However, it was difficult to assess what material the purge directly could replace in the cement plant under study and a potential environmental benefit from using the purge in the cement production process was therefore not considered. Further research is necessary to assess how to adequately allocate the environmental footprint to the purge stream.

Biomass was considered to be carbon neutral in this study. The CO₂ captured after the combustion of biomass was assumed to be taken from the atmosphere. However, this assumption is strongly disputed in literature [16], [17], and the actual reduction of climate change potential depends on how well forests or biomass plantations are

managed and on the so called carbon debt, the time between adsorbing the CO₂ and the combustion of the biomass. Most LCA studies still assume biomass to be carbon neutral as it remains difficult to assess the carbon footprint of biomass in LCA research. Nevertheless, by doing so, the climate change reduction potential of using biomass as fuel for calcium looping might be overestimated.

In this paper only CCP was used as indicator of the environmental performance of the process. Further work including a complete life cycle assessment addressing all environmental indicators is necessary to provide a comprehensive picture of the environmental performance. This work, currently under development, will also include a more detailed analysis of the impact of fuel selection on the technical performance and a sensitivity analysis to explore the potential impact of some of the assumptions mentioned.

Acknowledgements

This research has been carried out as part of the EDDiCCUT project (www.eddiccut.com). EDDiCCUT is a 4 year research project (2012-2016) aiming to assess the environmental performance of carbon capture and utilization technologies. The project is led by the Industrial Ecology Programme at Norwegian University of Science and Technology in collaboration with the Copernicus institute at Utrecht University and Tel-Tek in Porsgrunn. Project is supported by Gassnova and industrial partners.

We thank Lars-André Tokheim from the University College of Southeast-Norway for his contribution to this research by participating in scoping, brainstorm and discussion sessions.

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