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CO$_2$ Capture and Utilization in Cement and Iron and Steel Industries

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Abstract

This paper evaluates different technological options for the reduction of CO$_2$ emissions in heavy industries through their capture and use of best available technologies. This work presents the approach and the preliminary results obtained in the evaluation of iron and steel industry using oxygen blast furnace gas recycling after CO$_2$ capture in a pressure swing adsorber. It also evaluates the use of post-combustion capture using amines for retrofitting in the cement industry. Both are considered the most feasible technologies to be deployed in Europe in the medium term. Calibration, validation and future work regarding iron and steel industry model are described. The study finally assess the potential of cement and iron and steel industries to supply CO$_2$ to urea and methanol industries, which are two prospective users of CO$_2$ captured in Europe.

Keywords: Iron and steel industry; cement industry; carbon capture; carbon utilisation; oxygen blast furnace; post-combustion; conceptual design

1. Introduction

In the fight against climate change, energy-intensive industries have increasingly reduced their carbon emissions by enhancing their efficiency. However, industries like cement and iron and steel have inherent CO$_2$ emissions from mineral conversion. In Europe, the industries that release most of the CO$_2$ emissions are cement, responsible of 3-4% of the total CO$_2$ emissions [1], and iron and steel, contributing by 4-7% [2]. Therefore, the use of best available technologies (BAT), of highly productive reactors, and of more integrated plants, has reached a plateau in terms of carbon emissions reduction. While the energy sector can decrease its emissions by using alternative fuels, the only potentially significant alternative in the heavy industry sector is to capture the CO$_2$. Carbon capture and storage (CCS) and carbon capture and utilization (CCU), or carbon capture utilization and storage (CCUS), are then

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necessary to significantly reduce global carbon emissions from the energy intensive industrial sector, and in particular from iron and steel and cement industries.

Nomenclature

BAT	best available technology
BF	blast furnace
BFG	blast furnace gas
BOF	basic oxygen furnace
BOFG	basic oxygen furnace gas
CCS	carbon capture and storage
CCU	carbon capture and utilization
CCUS	carbon capture, storage and utilization
COG	coke oven gas
CRI	Carbon Recycling International
EU	European Union
FGD	flue gas desulphurization
GHG	greenhouse gases
HM	hot metal
HRC	hot rolled coil
IEA	International Energy Agency
JRC	Joint Research Centre
MEA	monoethanol amine
OBF	oxygen blast furnace
OBFG	oxygen blast furnace gas
PCI	pulverized coal injection
PG	process gas
PSA	pressure swing adsorption
SET	Strategic Energy Technology
TG	top gas
TGR	top gas recycling
TGRBF	top gas recycling blast furnace
ULCOS	Ultra-low Carbon dioxide Steelmaking

The European Union (EU) has set as objective the reduction of greenhouse gas emissions (GHG) of 40% below the 1990 levels by 2030, and of 80-95% by 2050. Recognizing that technology will play a pivotal role in achieving this challenge, the EU has identified the strategic R&D priorities in its Strategic Energy Technology Plan (SET-Plan), which can facilitate the transition to a decarbonized energy system. This plan has highlighted the relevance of CCS and CCU in energy and industry sectors.

The starting point of this work is a previous study of our research group that uses process flow modelling to identify the best capture method for the cement industry [1]. This work also relies on the economic analyses performed from the application of BATs in energy intensive industries [3,4]. In this paper, first we explain the modelling approach in iron and steel industry, which is then calibrated and validated against industrial plant data for the steel mill. Second, we summarize the main results from Ref. [1]. Finally, we compare cement and iron and steel plants in terms of captured and utilized carbon dioxide potentials. The present iron and steel plant analysis will be refined, and the detailed work will be presented in a future follow-up study that is under preparation.

2. Modelling approach

The cement and iron and steel industries are modelled using a conceptual design approach, to identify and characterize the trade-offs between carbon capture and energy consumption. Only the most important operation units that influence the ratio between heat and power, and captured CO_2, are modelled. The models have been implemented in the commercial software Chemcad^{TM}. For calibration and validation, the modelling results are compared with the output streams of the reference plants in the cement and iron and steel carbon capture and storage (CCS) studies from the International Energy Agency (IEA) Greenhouse Gas (GHG) R&D Programme [5,6]. Steady-state conditions are assumed, e.g. the processes are continuous, and off-gases have constant flow rates. We have evaluated (i) post-combustion capture with methylethanol amine (MEA) for the cement industry, and (ii) oxygen blast furnace (OBF) with top gas recycling (TGR) and capture using pressure swing adsorption (PSA) for the iron and steel industry. These are the two most mature technologies that can be implemented in the medium term, as a retrofitting option. The following section describes the modelling strategy for iron and steel industry. The modelling strategy and results for cement industry are detailed in Ref. [1], and a summary is provided in sections 2.2 and 3.2.

2.1. Iron and steel industry

The iron and steel industry encompasses several and differentiated processes and routes. Different possibilities have been envisaged for carbon capture retrofit, since different CO_2 emission points exist. As reported in [7], 70% of the emissions come from flue gas produced in the blast furnace (BF) hot stoves and in the power plant, while coke oven gas (COG) and basic oxygen furnace gas (BOFG) are responsible for 9% and 7%, respectively [8]. This work models (i) a reference iron and steel plant based on Ref. [9], and (ii) an iron and steel plant with capture similar to that investigated in [6], but that follows the technological solution proposed by the Ultra-low Carbon dioxide Steelmaking (ULCOS) project. ULCOS aims at reducing CO_2 emissions by at least 50% from today's best known steelmaking routes. The technology selected as the most immediate (retrofitting) solution implementable at the industrial scale was the top gas recycling blast furnace (TGRBF), with CO_2 capture in a PSA. This configuration, instead of hot blast, uses low purity O_2 to produce the reducing gases from pulverized coal injection (PCI) coal. The BF is converted into an OBF. The gas obtained directly from the OBF is the top gas (TG) or the oxygen blast furnace gas (OBFG), and the reducing gas resulting after CO_2 capture is called process gas (PG) [9].

A previous Joint Research Centre (JRC) report [2] pointed out that carbon emissions in iron and steel industry have been already significantly reduced, thanks to energy saving policies and the use of electric arc furnaces. Different BATs options are identified and are readily available for steel mills: state-of-the-art power plants, coke dry quenching, waste gas heat recovery, optimized sinter/pellet ratio, among others. The current paper does not take into account possible BATs. The two plants described in the following sections consider as reference the work in [6] and different publications from the ULCOS project [9-12]. Ref. [6] uses mainly the Masmod model for the reference steelworks mass and energy balances. The modelling strategy in this paper is derived from that of [13].

2.1.1. Base case - The coke oven and blast furnace

The reference plant for the base case is a classical steelmaking plant in Western Europe, with a production of 4 Mtonnes of hot rolled coil (HRC) per year. A standard configuration route consists in coke oven or coking plant, sinter plant, BF, basic oxygen furnace (BOF) and hot rolling mill. In this work, only the coke oven and BF are considered, as being the two units on which modifications are required (i) for injecting of oxygen in the BF instead of air (OBF) and (ii) for PG recycling, since the blast furnace gas (BFG) is treated in the PSA, and coke production diminishes due to the reducing gases recirculated into the BF. It is supposed that the rest of the units remain the same.

The gases produced in the iron and steel plant have an important calorific value and can be used for different purposes. In the base case, COG is mainly used to meet the energy needs of the rest of the plant. In the coke oven, it is utilized to produce the pyrolysis gases, and in the BF to (i) dry the PCI coal and to (ii) heat up the cold blast. The BFG is used in the coke oven supporting the COG for the production of the pyrolysis gases, and in the BF, also with COG, to heat up the cold blast. The remaining is sent to the power plant. Fig. 1 shows a detailed distribution of the

by-product gases among the simulated units. The production of gases changes in the plant configuration with carbon capture. However, the needs satisfied by the base case gases, must be also fulfilled when capture is used.

2.1.2. CO₂ capture case – Oxygen blast furnace and pressure swing adsorption with cryogenic distillation

The main concepts that differ from the base case are:

- O_2 injection in the BF instead of air. Therefore, hot blast is not produced, and hot stoves are not required.
- Gas from BF recycled to the own BF, as a reducing agent for the iron ore. Since this gas must be at high temperature (i.e. adapted temperatures according to the injection zone), PG gas is pre-heated before entering the tuyeres and the shaft with hot gases from natural gas combustion.
- Use of CO_2 capture and CO_2 purification techniques.
- Extra combustible needs (i.e. to cover the demand of BFG that is not satisfied with the change of configuration), are covered by natural gas.

Capture was performed in the ULCOS project using a PSA with tested pressures between 4.2 – 5.5 bar. The last CO_2 purification unit before compression and transportation, is a cryogenic distillation column that mainly separates CO_2 and CO [9,10]. The work in [14] outlines the PSA-cryogenic distillation flowsheet applied in the iron and steel industry. PSA Cryocap™ steel (a capture flowsheet developed specifically for this type of industry) is being developed by Air Liquide. In this plant, the TG is cleaned and compressed to PSA working pressure. After the PSA, the tail gas is fed to a cryogenic purification unit, to obtain pure CO_2. The non-condensable gases are recycled back to the compressed gases before the PSA and the PSA product gas, the reducing gas, is re-injected into the OBF. In the CO_2 capture case, following the ULCOS configuration, the hot blast is replaced by oxygen. The so-called Version 4 achieves a 25% of carbon reduction. The recycled PG enters the furnace through tuyeres and shaft, at 1250 °C and 900 °C, respectively. COG production, which is lower due to a minor need of coke, is used again to supply all the needs of the rest of the plant that does not change with the addition of the capture configuration, to dry the PCI coal and to produce the pyrolysis gases. PG is used to supply reducing agents to the BF, and to produce the pyrolysis gas. Fig. 1 presents an outline of the distribution of by-product gases for the configuration with carbon capture.

2.1.3. Model outline and validation

The model is implemented in Chemcad™. A production year of 7,884 hours (90%) is assumed. Ambient conditions are 15 °C and 1.01325 bar. Air composition is, in molar fractions, 78.17% N_2, 20.72% O_2 and 1.11% H_2O. The modelling approach uses as K-value model the Soave-Redlich-Kwong state equation (SRK) with the Enthalpy model SRK for all the units, except for pressurized CO_2 system, that uses as Enthalpy model the modified Soave-Redlich-Kwong (MSRK) state equation. The iron and steel model, comprising the coke oven and the BF, has *six* well differentiated zones (see in Fig. 1 a scheme of the modeled zones and their layout).

For the coke oven:
- Production of pyrolysis gases.
- COG cleaning and share.

For the BF:
- Preparation zone.
- Hot stoves.
- Tuyeres.
- Elaboration zone.

The *pyrolysis* simulation is adapted from the approach described in [15], then calibrated to obtain the representative COG composition. The empirical equations that model solid transformation into gases and char [16,17], are introduced into an Excel™ spreadsheet and connected to Chemcad™ simulation using Data Maps. On the one hand, the pyrolysis gases are produced in the combustion of COG and BFG with air in a Gibbs reactor. On the other hand, the resultant COG is cleaned from impurities (using component separators) and divided into 5 streams: COG to (i) users (outside the coke oven and the BF), (ii) for pyrolysis gases production, (iii) for PCI coal

drying, (iv) to the hot stoves and (v) the remaining is flared. The coke temperature is decreased with a stream of water.
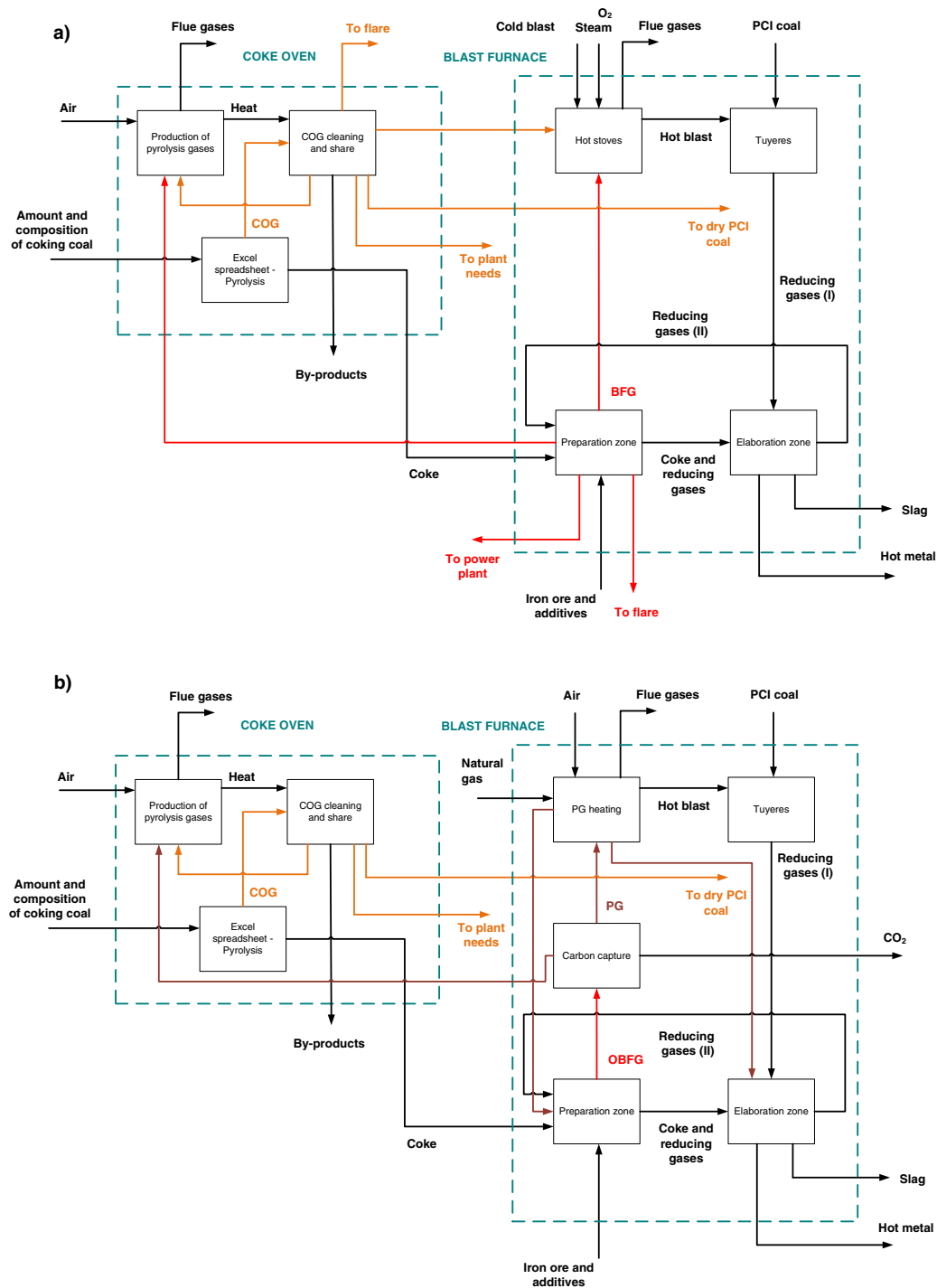

Fig. 1. Block diagrams that outline the modelling blocks and the COG, BFG, OBFG and PG connections: (a) Base case; (b) CO_2 capture case.

Coke and iron mineral with quartzite and limestone, enter the BF in the so-called *preparation zone*. In this zone, the mineral is mainly transformed into wüstite (FeO). This section has been modeled with five consecutive stoichiometric reactors, with adapted conversion rates, as a result of the calibration.

The production and elaboration zones are "separated" by the so-called *reservation zone*. The temperature in this zone is assumed of 950 °C, and all the streams connecting preparation and elaboration zones are at this temperature.

The main purpose of the *elaboration zone* is the reduction of wüstite into iron to obtain the hot metal (HM). First, the remaining magnetite is converted into wüstite. This modelling approach assumes that the HM is 100% *Fe*. The reactions that take place are simulated in stoichiometric reactors with specified conversions. At the end, a Gibbs reactor contributes as a final step to obtain HM and slag, and the reducing gases to be recycled to the preparation zone.

The reducing gas from the *tuyeres* is the result of the gasification of the PCI coal (previously dried and mixed with N_2) with the hot blast. The gasification reactions are represented by stoichiometric reactors with adjusted conversion rates.

The cold blast is heated up to 1118 °C with the hot gases from the *hot stoves*. The cold blast is a mixture of air, high purity O_2 and steam. Hot gases are obtained in the combustion of COG and BFG with air. The combustion is represented by an adiabatic Gibbs reactor.

To accommodate the *CO₂ capture* configuration, the base case is modified as follows:

- Instead of hot blast, low purity O_2 is directly injected into the tuyeres. The injection of PCI coal remains the same. Hot stoves are "replaced" by the heating system of the PG. Hot gases come from the combustion of natural gas.
- Since apart from coal and coke, PG is acting as reducing gas, the injection, and hence, the production of coke, is adjusted to achieve the final TG composition.
- PG gas is obtained by separating CO_2 from the rest of the gases using a PSA. Then, the gas composed mainly by CO_2 (and CO) is purified in a cryogenic distillator. PG gas is recycled back to the BF. PG not recycled is used in the coke oven.
- Purified CO_2 stream (100%) is further compressed up to 110 bar, and then prepared to be transported by pipelines.
- Natural gas is used to top the heating value not provided in this case by the TG to the power plant.

The *PSA* and the *cryogenic distillation*, at this stage, are modelled using component separators. The PSA is assumed to take place at 5.5. bar, and the resulting stream has a molar composition of 10.1% CO, 66.7% CO_2 and 23.1% H_2O (13% CO and 87% CO_2 dried). The desorption step of the PSA cycle is supposed to take place at 1 bar. The PG gas is then separated to be distributed to the OBF and the coke oven. The CO_2 rich stream at 1 bar is further compressed, in a multi-stage process that separates water, until the working pressure of the cryogenic unit. Once this pressure is obtained, the gas is cooled-down to the working temperature of the cryogenic unit. The cryogenic temperature is obtained by heat exchange with a refrigeration fluid. The gas is previously cooled down while warming up the off-gases of the distillation column that are recycled back before the PSA. Off-gases are expanded in a turbo-expander before being recycled.

Validation was performed after the model was calibrated. The current models were validated against the published input and output streams from [6], which also mentions some of the results of ULCOS programme. It is important to verify the composition of the COG, BFG for the base case, and of the OBFG and PG for the capture case, since it is crucial to ensure that the capture option fulfills the gas requirements in the base case. As example of the followed validation strategy, see in Fig. 2 the comparison between the simulated compositions with that of the reference data for the case study with carbon capture.

The relative errors in major streams flow rates and compositions predictions are in range of +/- 13%. Overall, the results show that in a conceptual design stage the developed general model for an iron and steel plant comprised by a coke oven and a BF, provides satisfactory accuracy for the purposes of our study.
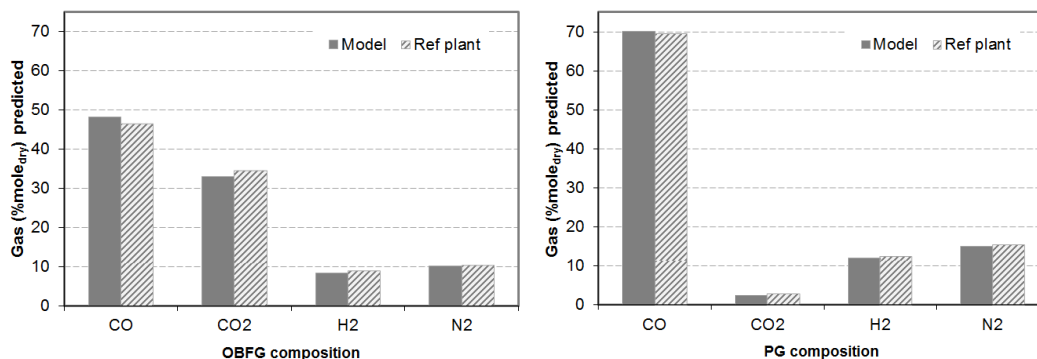
Fig. 2. Comparison of the composition obtained for the main gases of the simulated CO_2 capture case: (a) OBFG; (b) PG.

2.2. Cement industry

The base cement plant and the plant adapted for CO_2 capture technologies, post-combustion absorption with MEA, oxy-combustion, and calcium-looping are simulated and validated in [1]. The three options are compared in terms of specific energy consumption, CO_2 footprint, energy penalty, raw material consumption and energy recovery potential. The selected reference plant selected produces 1 Mtonne of clinker per year [5]. The model is again focused on only those units that are influenced by the carbon capture implementation, but it considers the energy consumption of the whole plant. The simulated functional units are:

- Pre-heater.
- Pre-calciner.
- Rotary kiln.
- Clinker cooler.

The flue gas from the pre-heater is treated in a flue gas desulfurizer prior to the MEA absorption to reach the appropriated amount of NO_x and SO_2. Based on the preliminary results, calcium-looping appears promising. Its shows lower specific energy consumption and capture energy penalty, and demonstrates higher potential to recover the produced waste heat, than the other analysed options. However, its integration in the cement plant is significantly more complicated. Post-combustion capture through MEA is a more feasible technology, which could be deployed in European cement plants in the medium term, and is used for further comparison in this study.

The thermodynamic models selected in Chemcad[TM] are, the Latent Heat Enthalpy model and the non-random two-liquid model (NRTL) K-value model, and the AMINE Enthalpy and K-value models for the specific MEA section. A Gibbs type reactor is used to model the pre-calciner and the rotary kiln. The cement kiln is assumed to operate in steady state conditions at atmospheric pressure and a temperature of 1450 °C. All solid/gas separations are modelled with flash vessels.

The flue gas is treated to reach acceptable levels of NO_x and SO_2, in a flue gas desulphurisation (FGD) unit, prior the amine absorption unit. The pre-calciner flue gas is cooled down to 40 °C before entering the absorption column. The absorption and the stripping units from the MEA process are modelled using SCDS rigorous multi-stage vapour-liquid equilibrium module. The thermophysical properties are modelled using the available Amine Kent-Eisenberg model, which is a simplified approach to model the reactions in a gas sweetening system. The reboiler temperature, at 120 °C, is an input in the model.

In order to verify the validity of the modelling approach, Ref. [1] compares energy consumption, CO_2 capture energy penalty, CO_2 footprint and CO_2 capture rate with different public bibliography. The main differences are justified.

3. Preliminary results

3.1. Iron and steel plant

As previously demonstrated, one of the main characteristics of an iron and steel plant is the integration and use of the produced gases. In this work, it is assumed that the needs of the BOFG are fully supplied. The main changes come from the demand fulfilled by the COG and the BFG, and specifically, from the power plant requirements, which is satisfied in the base case, but not when PG recycle is used in the CO_2 capture case. See in Fig. 3 and Fig. 4 the main-gases distributions for the two simulated cases. Natural gas consumption in the CO_2 capture case is 61,170 kg/h. Coking coal consumption decreases by around 25% in mass terms (73,700 kg/h). The emissions of CO_2 are 0.621 tCO_2/t_{HRC} for the base case, and 0.333 tCO_2/t_{HRC} for the CO_2 capture case. The flow rate of the captured CO_2 stream is of 445.2 t/h.
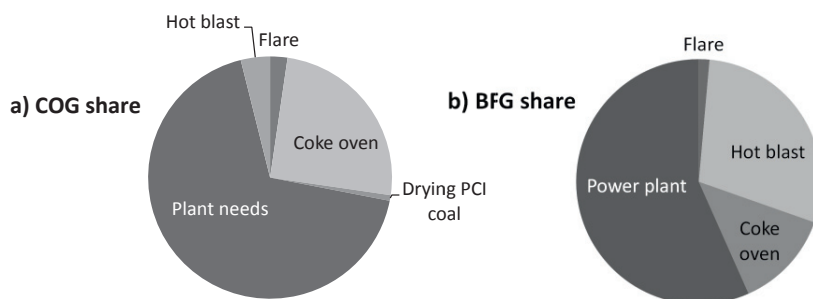

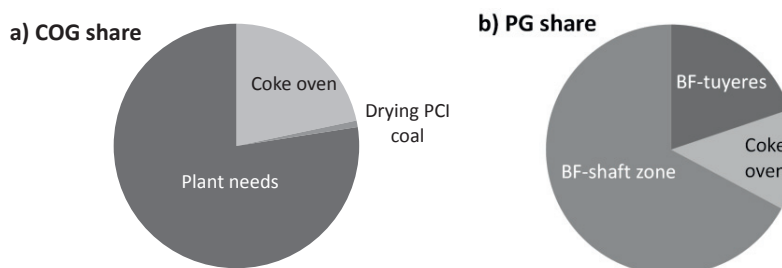Fig. 3. Base case gases share: (a) COG; (b) BFG.

Fig. 4. CO_2 capture case gases share (a) COG; (b) PG.

3.2. Cement plant

At the current status, the main challenge for implementing post-combustion capture with amines, is the scale-up of the process, since the results from the demonstration projects in power plants are not directly transferable to the cement industry. Specific energy consumption (in kJ/kg clinker) increases by 45% when integrating post-combustion capture with MEA absorption in the cement plant. The largest contributor to this value is the heat demand of the reboiler in the desorption column. The main performance indicators of the cement plant integrated with MEA absorption are summarized in Table 1, which also summarizes the values of the reference plant.

3.3. Utilization of CO_2

This section aims at a preliminarily compare the captured streams in the reference plants for cement and iron and steel production, with the two most developed utilization options: urea synthesis and methanol production. *Urea synthesis* already uses CO_2 as raw material. The so-called "urea yield boosting" aims at using captured CO_2 to

consume all the NH_3 produced for a given urea plant scale. *Methanol synthesis* from CO_2 is being built at a commercial scale. Carbon Recycling International (CRI) started the operation of the first commercial demonstration plant, in Iceland, in 2011. In Table 2, the values of captured CO_2 from simulations for heavy industries are compared with the values estimated for urea[1] and methanol[2] plants. The values reported for cement and for iron and steel correspond to 0.85 $tCO_2/t_{clinker}$ and 0.87 tCO_2/t_{HRC}, respectively.

Table 1. Main CO_2 capture performance indicators for the cement plant.

Plant	Base case	Amine scrubbing
Capture efficiency	-	85%
CO_2 capture energy penalty (kJ/kg CO_2)	-	4952.2
CO_2 in flue gas	34.1%	2.5 t/h
CO_2 in CO_2 stream after cooling	-	95%

Table 2. Comparison between the CO_2 captured and the CO_2 utilized at typical scales, cement and iron and steel on the one side, and urea and methanol on the other.

Plant	Scale (per year)	Stream of CO_2
Cement	1 Mtonne clinker	86.8 t/h
Iron and Steel	4 Mtonne TRC	445.2 t/h
Urea	666,000 tonne urea	2.5 t/h
Methanol	4,000 tonne methanol	0.6 t/h

The results in Table 2 underline the difference between the captured streams and the CO_2 utilization needs, if compared plant-to-plant. Less than 1% of the captured emissions can be used in a utilization plant. Provision of small clusters or networks would probably facilitate the implementation of capture and utilization techniques. The creation of new materials (and markets) with CO_2 will increase its utilization. For instance, if one of the products, such as methanol, can be used as fuel in transportation applications, the prospect of the market is higher. A detailed study on CCU will be published by the JRC by the beginning of 2015.

4. Conclusions and perspectives

This work presents a validated conceptual model of a reference iron and steel plant to observe its performance in terms of CO_2, energy and mass balances. Two cases have been investigated: (i) the reference plant with a classical plant configuration, and (ii) the plant with CO_2 capture, i.e. using oxygen blast furnace gas recycling and capture through a PSA, plus further CO_2 purification and compression. The model has been presented, explained and validated. Cement production was also analyzed through the study of a reference plant and a plant retrofitted with post-combustion capture. This paper underlines the difference of scale between carbon capture and utilization and sets the basis for further work in the area of heavy industry and utilization. However, it has been pointed out that it is feasible to retrofit iron and steel and cement industries with carbon capture technologies, and that CCU is part of the storage/use of captured CO_2 solution. As future work, both iron and steel and cement models will be refined. The JRC is preparing two reports in the areas of carbon capture in heavy industry and carbon utilization. The objective of the whole approach is to develop a useful tool to evaluate economics and environmental parameters in the field of CCUS, to recommend viable pathways, from capture to disposal.

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[1] Typical size and flow rates obtained from Stamicarbon.

[2] Current industrial scale and CO_2 consumption from the CRI presentation in http://setis.ec.europa.eu/setis-deliverables/setis-workshops-hearings/workshop-co2-re-use-technologies .

Acknowledgements

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