Abstract:

The industrial sector represents one-fifth of total global CO_2 emissions. Refining is the fourth largest contributor to CO_2 industrial emissions. The implementation of carbon capture and storage technologies (CCS) in the industrial sector is considered a key action by the International Energy Agency (IEA) to reduce CO_2 emissions. However, the majority of the development for CO_2 capture and storage is driven by the electric-utility sector. The aim of this paper is to assess the potential deployment of CO_2 capture within the refining sector as first step of the CCS chain.

The assessment starts with the description of the four main approaches for CO_2 capture, namely: post-combustion, pre-combustion, oxy-fuel combustion and chemical looping combustion.

Next, four principal CO_2 emissions routes are identified in the processes of a refinery: process heaters, hydrogen production, utilities and fluidised catalytic crackers (FCC). Suitable CO_2 capture technologies for each of these routes are analysed. Also, a brief overview of some CCS projects in the refining sector is presented.

The conclusions extracted point that there is an interest from the refining sector in CO_2 capture, as indicated by initiated demonstration projects. CO_2 capture is a technically feasible option for reducing CO_2 emissions from the refining sector through a range of post combustion, pre-combustion and oxy-fuel technologies. Yet, there are a number of challenges that are hindering widespread deployment of CO_2 capture that need to be overcome. Potential for early deployment of CO_2 capture in refineries exists for some high purity CO_2 streams, which are a by-product of certain hydrogen production processes. The latter, along with development of CCS in the power generation sector, could be the initial stage for a further progress of CO_2 capture in the refining sector.

Challenge of the integration of CO$_2$ capture in the refining sector

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1 Introduction

The industrial sector, mainly cement, iron and steel, chemicals and refining, represent one-fifth (7.1 $GtCO_2$) [1] of total global CO_2 emissions. Emissions from these sectors are expected to grow by around 35% up to 2050 [2] under current policies. Reducing CO_2 emissions from these sectors is crucial for the global action to prevent climate change.

In order to prevent climate change, the Intergovernmental Panel on Climate Change (IPCC) estimates that global CO_2 emissions need to decrease by between 50% and 85% of their 2000 levels by 2050 [3]. Carbon Capture and Storage (CCS) systems have emerged in recent years as feasible methods to mitigate CO_2 emissions. However, the majority of the development for CO_2 capture and storage is driven by the electric-utility sector, in which the emphasis is on large centralized units for electric power generation. Still, power generation sector accounted for 25% [4] of global greenhouse gas emissions in 2010, only 4% [4] more than industrial sector.

The International Energy Agency (IEA) considers the implementation of CCS in industrial sector as a key action to achieve the CO_2 reduction goal by 2050. Specifically, IEA estimates that CCS in industrial applications could represent around half of the emission reductions achieved through CCS by 2050 [1].

CCS encompasses a family of technologies and techniques that enable the capture of CO_2 from fuel combustion or industrial processes, the transport of CO_2 via ships or pipelines, and its storage underground, in depleted oil and gas fields and deep saline formations.

This paper intends to assess the deployment feasibility of CO_2 capture technologies in the refining sector, as one of the major industrial contributors to CO_2 emissions. Concretely, refining sector is responsible for 0.7 $GtCO_2$ out of the 7.1 $GtCO_2$ [1] from the industrial sector, being the fourth largest contributor, after iron and steel, cement and chemicals. This paper is structured as follows:

- Firstly, the main CO_2 capture technologies are presented. The aim of these technologies is to produce a stream of CO_2 highly concentrated from the flue gases originated in fuel combustion or industrial processes. Thus, this stream is next compressed and transported to its storage site. Both the functioning principles and the technical restrictions of each technology are described here.
- The third chapter assess the potential implementation of CO_2 capture technologies in a refinery. Four major emission routes in refineries potentially compatible with CO_2 capture are identified and analysed. Besides, a brief overview of current CCS projects in the refining sector is presented.
- Finally, chapter 4 contains the conclusions extracted from the analysis.

2 CO_2 capture options

In general, to economically sequester CO_2 derived from industrial plants, it is first necessary to produce a relatively pure, high pressure stream of CO_2. The process of producing this CO_2 stream is referred to as separation and capture, which encompasses all operations that take place at the plant site, including compression. For ease of transport, CO_2 is generally compressed to the order of 100 atm.

A variety of technologies for separating CO_2 from a mixture of gases are commercially accessible and widely used nowadays. Figure 1**¡Error! No se encuentra el origen de la referencia.** illustrates some of the technical approaches available. The selection of the technology is dependent on the requirements of product purity and on the conditions of the gas stream being treated (such as its temperature, pressure or CO_2 concentration).
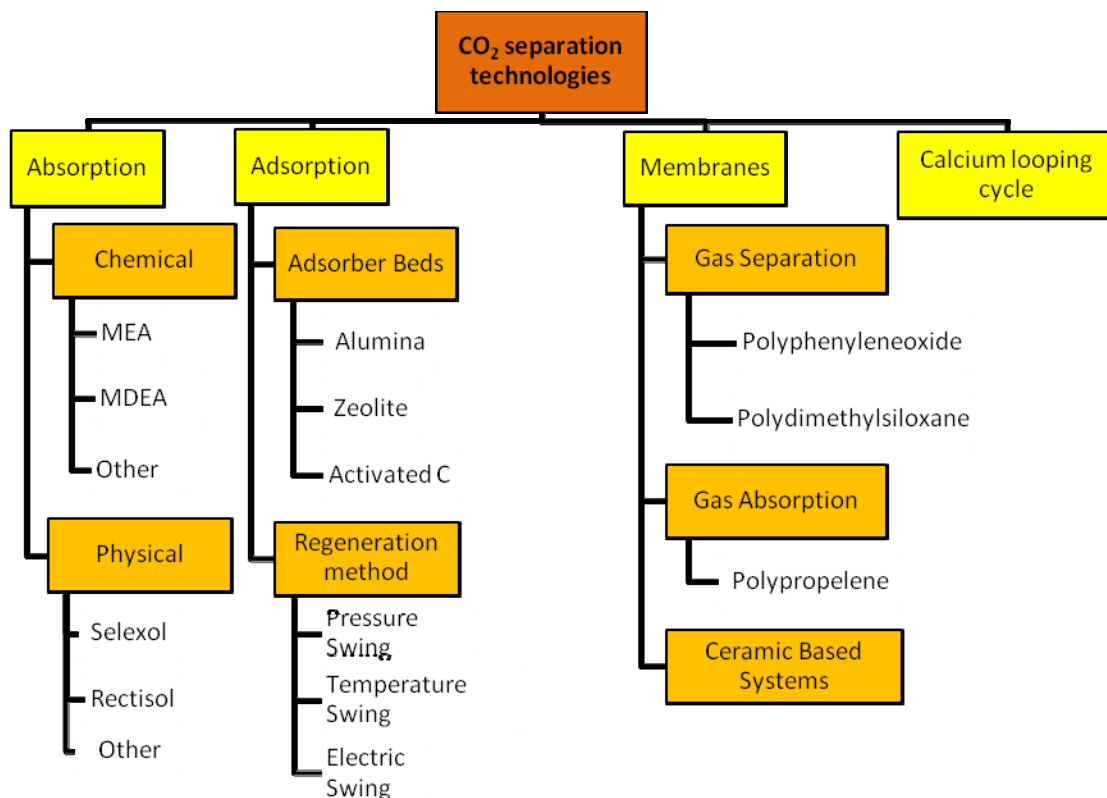
Figure 1. Technical options for CO_2 separation. Adapted from [5].

There are four main approaches to capture CO_2 from fossil-fuel-fired energy systems or industrial processes: pre-combustion, post-combustion, oxy-fuel combustion and chemical looping combustion. These technologies are suitable for large stationary carbon emission point sources, such as refineries. However, capture is only the first step of the CCS chain. Once captured, the CO_2 requires to be compressed, for its subsequent transportation to the storage site.

2.1 Post-combustion

Post-combustion capture is the removal of CO_2 from flue gases downstream of the emission sources. At first glance, these capture methods can be applied to flue gases from the combustion of any fossil fuel, but actually their content in impurities affect the installation in terms of design and cost. Hence, it becomes necessary to place upstream of the capture system, units for the removal of other pollutants, such as SO_x or particles [6, 7].

In broad terms, CO_2 can be captured in post-combustion using:

- Chemical absorption
- Physical absorption
- Adsorption
- Membranes
- Calcium looping cycle

1. Chemical absorption

The most mature post-combustion capture techniques comprise liquid absorption using chemical solvents. Liquid absorption involves a mass transfer process with a chemical reaction process. Figure 2 shows a scheme of a post-combustion plant based on chemical absorption.
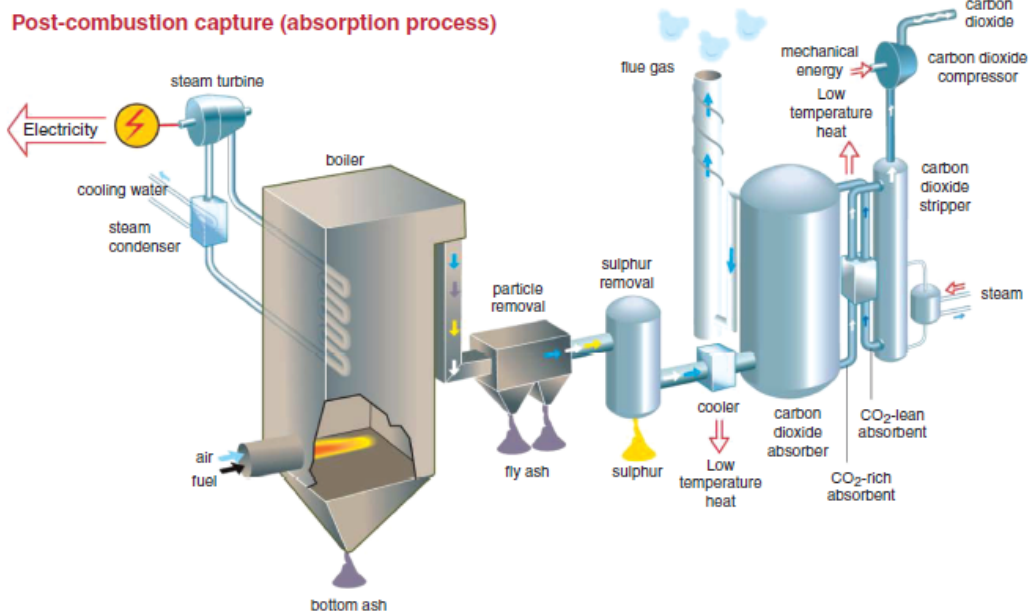
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Figure 2. Post-combustion capture schematic [6]

Low CO_2 partial pressures in flue gases are the main reason for the selection of CO_2 capture based on chemical absorption. During the capture process, the exhaust gas is cooled and channelled to an absorber, where it flows counter current of a chemical solvent. Here, most of the CO_2 is absorbed within the solvent, which is then sent to a stripper where CO_2 is recovered by the application heat, regenerating the solvent. The recovered CO_2 is next dehydrated and compressed for transport.

The most commercially mature post-combustion technology is Fluor's Econamine FG Plus. It uses a 30 wt% monoethanol amine (MEA) solution that can achieve CO_2 recovery efficiencies ranging from 85% to 95%. Although MEA is the best-known solvent, its high energy requirements and degradation rates are major drawbacks. Another process, developed by BASF, uses activated methyldiethanolamine (MDEA) for H_2S and CO_2 removal [6, 8].

2. Physical absorption

Physical absorption methods are explained below for pre-combustion technologies. These methods can be applied for post-combustion separation of CO_2; however, the exhaust gases conditions (low pressure) are not the most adequate.

3. Adsorption

The CO_2 adsorption process comprises the capture, fixation and accumulation of the CO_2 gaseous molecule on the surface of another one, typically a solid. This phenomenon depends on the adsorbent mass, pressure, temperature and the nature of both the gas and the solid surface. The key for the consolidation of this process remains on developing an adsorbent with high adsorption capacity for CO_2 and high selectivity for different concentrations of CO_2 in the gas stream. The adsorption process also involves a sorbent regeneration process. The most common regeneration methods are those based on temperature and pressure changes. The main three regeneration methods are:

- PSA (Pressure Swing Adsorption): under high pressure, gases tend to be attracted to solid surfaces, or "adsorbed". The higher the pressure, the more gas is adsorbed; when the pressure is reduced, the gas is released, or desorbed.
- TSA (Temperature Swing Adsorption): with this method, sorbent is regenerated when temperature is increased.
- ESA (Electric Swing Adsorption): regeneration occurs when a low voltage currents is applied to the sorbent.

4. Membranes

Membranes are permeable materials that can be used to selectively separate CO_2 from other components of a gas stream. They effectively act as a filter, allowing only CO_2 to pass through the material. The driving force for this separation process is a pressure differential across the membrane, which can be created either by compressing the gas on the feed side of the material or by creating a vacuum on the downstream side.

Despite it is a relatively simple technology, the volume of the gas stream to be treated in post-combustion systems is very large, and CO_2 is usually very dilute, so the separation efficiency falls. Another inconvenience is derived from the high degradation rate of membranes, caused by the flue gas conditions (e.g. temperature, impurities) [5, 8, 9].

5. Calcium looping cycle

Some new post-combustion capture processes employ solid sorbents rather than liquid solvents to capture CO_2 from the flue gas.

An advanced concept called calcium looping cycle or carbonate looping cycle is currently believed to be a breakthrough in the cost of avoided CO_2. Studies indicate that calcium looping has the potential to be more efficient and lower in cost than amine-based capture methods. In this process the desulfurized flue gas passes through a carbonator bed where calcium oxide (CaO) reacts with CO_2 in the flue gas to form calcium carbonate ($CaCO_3$). The carbonate is then heated in a separate reactor called the calciner where the reverse reaction takes place, releasing the CO_2. Energy for this reaction is supplied by combusting a fuel such as coal with high-purity oxygen. The CaO formed in the calciner is then sent back to the carbonator to complete the loop. A schematic of this process is shown in Figure 3 [5, 8].
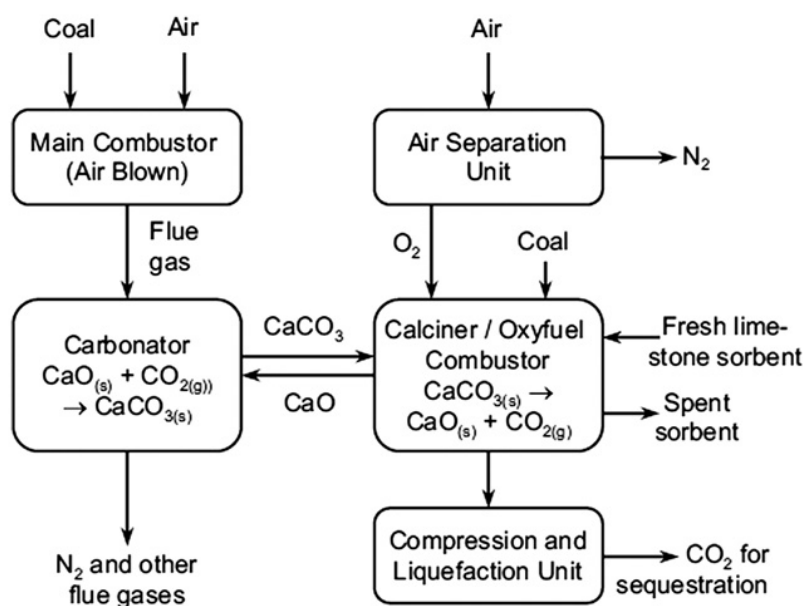

Figure 3. Calcium looping cycle schematic [5]

2.2 Pre-combustion

In pre-combustion capture, CO_2 is separated from a gaseous fuel mixture under reducing conditions before its combustion. The fuel entering the combustion chamber has been previously decarbonized; thereby the flue gas produced is emitted to the environment free of CO_2.

CO_2 separation techniques for pre-combustion are those described above for post-combustion, but applied to the gas stream derived from the decarbonization process. The most commercially available technologies rely on liquid solvents for CO_2 absorption, usually by scrubbing the gas stream with physical solvents. A simplified diagram is depicted in Figure 4.
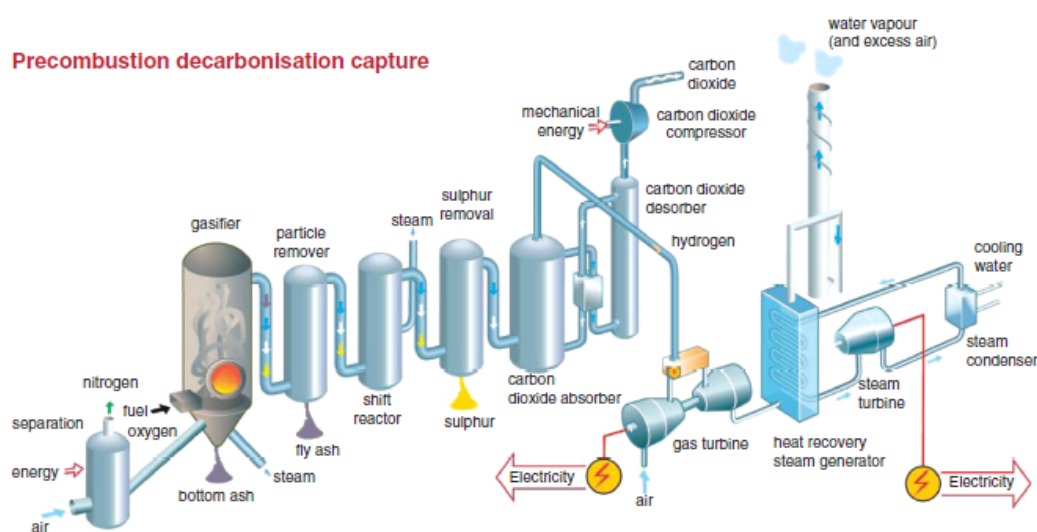

Figure 4. Pre-combustion capture schematic [6]

Pre-combustion capture starts with the conversion of the carbonaceous fuel (e.g. natural gas, biomass, or oil) to a gaseous mixture consisting primarily of H_2 and CO, named syngas. This fuel-conversion step, called gasification for solid fuels and reforming for gaseous fuels, is endothermic and requires supplementary heating, typically supplied by the partial oxidation of the fuel. In CO_2 capture application, high-purity O_2 obtained from an air separation unit (ASU) is often used as oxidant, yielding the high temperatures required to produce syngas. The CO in the syngas is converted to H_2 and CO_2 through the water/gas shift (WGS) reaction. The WGS reaction is facilitated by commercially available catalysts. CO_2 can now be separated from the gaseous mixture.

Syngas production from gasification and partial oxidation is generally carried out at high pressure, resulting in high CO_2 partial pressures. For this reason, physical solvents are the preferred means to capture CO_2 in pre-combustion applications. The CO_2-rich solvent is regenerated by pressure reduction, releasing the CO_2, and the regenerated solvent is reused for CO_2 absorption. The recovered CO_2 is dried and compressed for its transport and storage, while the hydrogen-rich stream can be used as fuel. The most extended processes employ Rectisol and Selexol as solvents.

Physical absorption is subject to Henry's law, which results in a linear dependency between the CO_2 partial pressure and the solvent capture effectiveness. As a result, although the fuel-

conversion process is laborious and costly, CO_2 separation is straightforward, avoiding the need of a chemical reaction for CO_2 capture [5, 6, 8].

2.3 Oxy-fuel combustion

Oxy-fuel combustion systems are being developed as an alternative to post-combustion technologies. In oxy-fuel combustion, pure oxygen is used as oxidant instead of air. This eliminates the huge amounts of N_2 in the flue gas stream.

After the particulate matter (fly ash) is removed, the flue gas consists of CO_2, smaller amounts of pollutants such as SO_2 and NO_x, and water vapour. The latter, can be removed by condensation, and the remaining impurities using the adequate conventional technology, thus obtaining a nearly-pure CO_2 stream that can be sent directly to storage, as shows Figure 5.
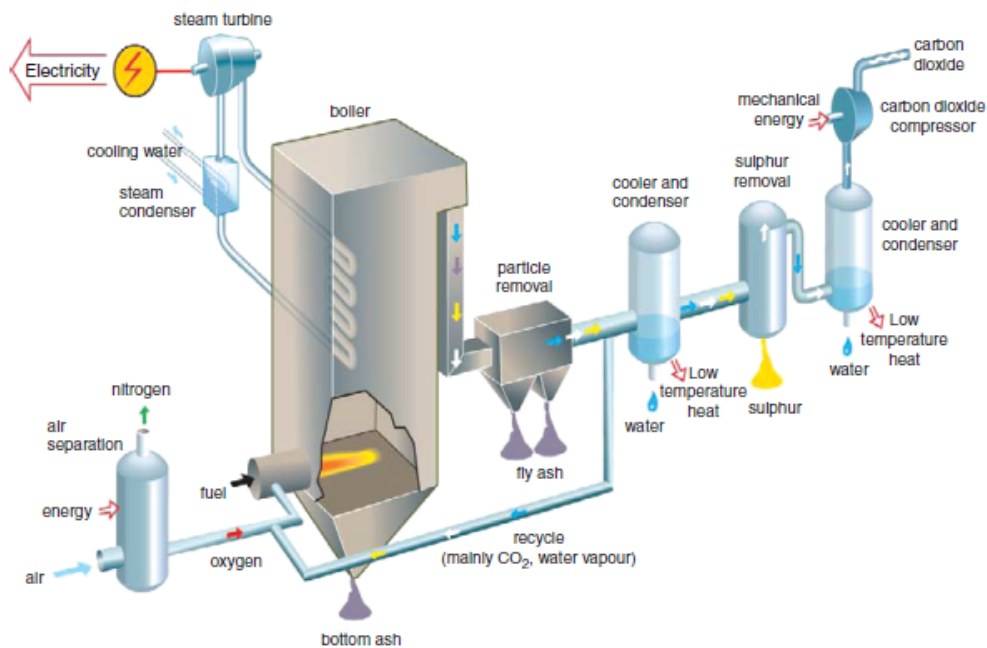
Figure 5. Oxy-fuel combustion capture schematic [6]

Using oxygen instead of air reduces the oxidant mass, reaching higher temperatures than in conventional combustion. Hence, a certain portion of flue gas is recycled to the combustion process to moderate the flame temperatures. Temperature can be also reduced by water injection.

The principal benefit of oxy-fuel combustion is that it avoids the need for a costly post-combustion CO_2 capture system. However, it requires an air separation unit (ASU) to generate the relatively pure oxygen (95%-99%) needed for combustion. In addition, this technology requires three times more O_2 than pre-combustion methods, elevating costs related to the ASU [5, 6].

2.4 Chemical looping combustion

This technique involves splitting the combustion of a hydrocarbon in two independent oxidation and reduction reactions. For this purpose, a suitable metal oxide acts as carrier of O_2 between two reactors. The carrier transfers O_2 from the combustion air to the fuel, thus, direct contact between the fuel and the combustion air is avoided. Figure 6 shows a simplified diagram of the chemical looping combustion.

In this process, the air passes through a reactor containing a powdered metal at $800 – 1,200^\circ C$. The metal is oxidized; next enters into another reactor where it is reduced by the fuel, releasing

the O_2 required for the combustion. The cyclone in Figure 6 is used to separate the metal oxide from the excess air.
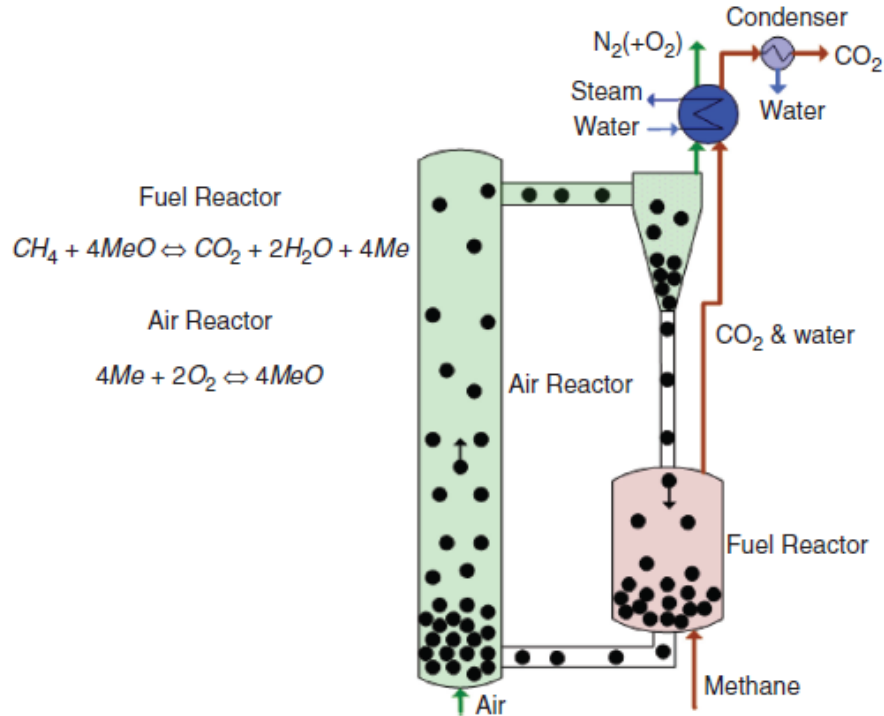

Fuel Reactor

$$CH_4 + 4MeO \Leftrightarrow CO_2 + 2H_2O + 4Me$$

Air Reactor

$$4Me + 2O_2 \Leftrightarrow 4MeO$$

Figure 6. Chemical looping combustion capture schematic [6]

An ASU is not needed for this kind of CO_2 capture. The advantage of avoiding direct contact between air and fuel is that the products of combustion, CO_2 and water, are separated from N_2 and any excess O_2. After water is condensed, a nearly-pure CO_2 stream is obtained which does not require any additional treatment except compression. If impurities such as NO_x or SO_x were present, conventional technology could be applied.

Chemical looping combustion has a great potential to significantly reduce CO_2 capture costs. However, it is still in the initial phase of its development, with challenges to overcome regarding the materials used and the selection of O_2 carriers [5-7, 10].

3 CO_2 capture in Refining Sector

The application of CCS to refineries is challenging due to the fact that CO_2 is emitted from many sources, which may be dispersed and could be relatively small compared to other industrial sectors.

Most applications of CCS in industry require a capture step to concentrate the relatively dilute streams of CO_2 to a level that will enable economic transportation and storage. Combustion-derived emissions produce the majority of CO_2 at refineries, generally at low pressure and concentrations (4% to 15%, similar to power plants). There are also some refining processes that produce flue gas streams with higher CO_2 concentration, allowing lower capture costs.

3.1 Deploying CO$_2$ capture in Refining Sector

There are four major emission routes at refineries which are potentially compatible with CO$_2$ capture technologies. These routes are associated to the following production processes in refineries: process heaters, hydrogen production, utilities and fluidised catalytic crackers (FCC).

Suitable CO$_2$ capture technologies for each of these processes are analyzed below. Many of these technologies are already available but are not yet mature for CCS, or are only at demonstration stage.

3.1.1 CO$_2$ capture from process heaters

Refineries employ numerous fired heaters and boilers of different sizes and capacity throughout the facility. These could have capacities ranging between 2 MW and 250MW, and a typical refinery could have between 20 and 30 different interconnected processes around the site. This heating equipment usually uses different types of fuel that are available on-site, thus producing flue gas with a wide-ranging CO$_2$ composition. Together, these dispersed emission sources can in some cases be the largest producers of CO$_2$ in the refinery, being able to reach up to 60% of the total emissions.

Post-combustion capture and oxy-fuel combustion currently offer possibilities for reducing emissions from process heaters in refineries. Technologies that could potentially be implemented in the future in new facilities include chemical looping combustion, and pre-combustion capture in the production of hydrogen fuel for use in boilers and heaters.

The retrofit of heaters with post-combustion capture technologies is limited due to the wide distribution of heating units within the refinery complex. Some experts have proposed to resolve this by ducting the gases from dispersed heaters to a central location where CO$_2$ could be separated and compressed. Others, have questioned the feasibility of such an approach and proposed instead to capture only the CO$_2$ from the largest on-site stacks.

Oxy-fuel offers another potential mechanism for capturing the CO$_2$ from heaters and boilers. For this scenario it is proposed that all heaters and boilers on site are modified for firing with pure oxygen, produced at a central location, and that flue gases from the combustion plants will be initially treated at each CO$_2$ generation point (where water will be removed and CO$_2$ will be compressed to 30bar). Final compression of CO$_2$ will take place at one central location. One of the challenges will be the large distances for piping oxygen and CO$_2$ across the site between oxygen facilities and CO$_2$ compression stations [11-13].

3.1.2 CO$_2$ capture from hydrogen production

Between 5% and 20% of refinery CO$_2$ emissions are linked to the production of hydrogen (H$_2$). Hydrogen is a by-product of the catalytic reformer and fluid catalytic cracker (FCC) processes, but as demand of H$_2$ has increased (due to changes in fuel specification to reduce sulphur content), demand now exceeds supply from these processes in most refineries. To meet the increased demand, hydrogen is produced either through the steam methane reforming (SMR) of natural gas or through the gasification of heavy residues and fuel oil. The hydrogen produced in both of these processes needs to be separated from other constituents in the flue gases.

Hydrogen is most commonly produced through SMR. Traditionally, hydrogen produced in SMR plants was purified using chemical absorbents such as amines MDEA, resulting in high purity CO$_2$. However, in the past three decades has emerged a trend towards separation using PSA. In the current refining market, PSA offers two advantages over amine chemical absorption: 1) PSA produces very high purity hydrogen, and 2) the overall energy efficiency of the hydrogen production process is increased compared with chemical absorption. The change to PSA has been driven by the market need of high purity hydrogen. But PSA results in much lower concentration CO$_2$ in streams which contain 20-30% impurities. The impurities include H$_2$, CO

and methane (CH_4) making the gas suitable for reuse as fuel in the SMR furnace, but reducing the feasibility of CO_2 capture and increasing the cost.

Gasification plants for hydrogen production are generally larger than SMR and operate at high pressures of 50-70 bar. These conditions are suitable for the use of physical absorption solvents over chemical absorption solvents because they have higher loadings, require less energy input and produce dry CO_2 under these conditions. With gasification, all the CO_2 emissions associated with conversion end up in the flue gas stream and therefore, the rate of capture is higher than with SMR [13, 14]

3.1.3 CO_2 capture from utilities

Refineries require a large amount of steam and electricity to meet the energy demand of the different processes, and can account for 20% to 50% of the total refinery emissions. Steam is provided on-site, and in order to increase efficiency, it may be undertaken in conjunction with electricity production via combined heat and power (CHP). As these processes closely mirror those used in the power sector, this implies that opportunities for applying CO_2 capture in the utility installations of the refineries will follow the development of CO_2 capture technology in the power sector. Post-combustion and oxy-fuel combustion are the most promising technology for CHP. In the case of Integrated Gasification Combined Cycle (IGCC) pre-combustion would be the most suitable technology for CO_2 capture [11, 12].

3.1.4 CO_2 capture from fluidized catalytic cracking

In those refineries that operate fluidised catalytic cracking (FCC) units, such units can account for 20% to 50% of the total CO_2 emissions from the refinery. Unlike most of the other emissions from a refinery, the emissions from FCCs are process-related rather than combustion-related. During processing, carbon is deposited on the surface of a catalyst powder. The catalyst is regenerated by the oxidation of coke with air.

Depending on the process, the concentration of CO_2 in the flue gas typically ranges from 10% to 20%. Two technology options exist for the capture of CO_2 from the FCC: post-combustion capture, the most mature, and oxy-fuel combustion of the regeneration process, still in development. The potential of both has been compared, and despite the relatively high capital cost of oxy-fuel, the potential of lower operating costs makes it attractive option too [13-15].

3.2 Status of CCS in Refining sector

Some CCS projects in the refining sector are already underway, and others are planned. Some of them are presented below:

- In Norway, Statoil, Gassnova (which represents the Norwegian Government in matters relating to CCS), Norske Shell and Sasol have established an agreement to develop, test and verify solutions for carbon capture, in Statoil's refinery in Mongstad. They have built a centre for carbon capture technologies at Mongstad, known as the "CO_2 Technology Centre Mongstad" (TCM). TCM aims to test CO_2 capture from flue gas streams in the refinery cracker and in the cogeneration power plant of the refinery. They are also planning to develop a full-scale capture plant at Mongstad [16].
- In Brazil, Petrobras is operating a demonstration project for CO_2 capture by oxy-firing FCC in a refinery.
- In Canada, the Alberta government financially supports the North West Upgrading bitumen refinery project, which will capture CO_2 from a gasification process used to produce hydrogen [14].

- In Rotterdam, CO_2 from Shell's Pernis refinery is captured, transported and used in nearby greenhouses. Plans to transport more CO_2 from the refinery and store it in the depleted Barendrecht gas field were cancelled because of public resistance to storage [17].
- In France, Total has been testing since 2010 oxy-fuel combustion capture at the countries' largest production site of liquid hydrocarbons, in Lacq [18].

4 Conclusions

This paper has analysed the potential integration of CO_2 capture technologies in the refining sector. There are four main approaches for CO_2 capture: post-combustion, pre-combustion, oxy-fuel combustion and chemical looping. To explore their potential within the refining industry the main emission sources of a refinery have been identified: process heaters, hydrogen production, utilities and FCC. The contribution to the total refinery emissions of each source is shown in Table 1.

CO_2 emitter	% of total refinery emissions
Process heaters	30%-60%
Hydrogen production	5%-20%
Utilities	20%-50%
FCC	20%-50%

Table 1. CO_2 emission sources at a typical refinery complex

Process heaters represent the largest CO_2 sources. The technical feasibility for CO_2 capture from process heaters is highly dependent on plant configuration, and the availability and accessibility of combined stacks. Retrofitting process heaters in a refinery with post-combustion capture technologies is limited by the wide distribution of heating units. The possibility of ducting multiple flue gas streams to a single CO_2 capture unit has been proposed, but the feasibility of these solutions is contested. Oxy-fuel combustion is emerging as an alternative to post-combustion. On the one hand, the energy penalty for CO_2 capture is reduced by avoiding the solvent regeneration need. On the other hand, inconveniences related to piping need to be overcome.

Hydrogen production accounts from 5% to 20% of CO_2 emissions from a refinery. It produces concentrated streams of CO_2 often at a high pressure. Thus, it offers a low-cost option for CO_2 capture deployment. However, not all hydrogen production routes are equally suitable for CO_2 capture. Two pre-combustion separation processes dominate: chemical absorption and PSA. Chemical absorption could lead to a very pure stream of CO_2, which could be directly compressed. By contrast, PSA produces lower CO_2 concentrations and is more focused on re-firing the gas stream. Although PSA produces hydrogen of higher quality and at a lower cost, this may not be the most adequate option for a carbon constrained world.

Regarding utilities, capture technologies for the power generation sector will probably reach development before other sectors, so refinery utilities may be a good opportunity for the deployment of their learnings. CHP also makes CO_2 capture technologies more applicable because emissions are centralized.

Finally, for FCC the potential to reduce the energy penalty of CO_2 capture derived from post-combustion may support oxy-fuel combustion in the long term.

There is an interest from the refining sector in CO_2 capture, as indicated by initiated demonstration projects. CO_2 capture is a technically feasible option for reducing CO_2 emissions from the refining sector through a range of post combustion, pre-combustion and oxy-fuel technologies. Yet, there are a number of challenges that are hindering widespread deployment of CO_2 capture that need to be overcome. Potential for early deployment of CO_2 capture in refineries exists for some high purity CO_2 streams, which are a by-product of certain hydrogen production processes. The latter, along with development of CCS in the power generation sector, could be the initial stage for a further progress of CO_2 capture in the refining sector.

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