

Chapter 2

Carbon Capture Technologies

Peter Markewitz and Richard Bongartz

Abstract This chapter focuses on carbon capture technologies that can be used in coal fired power plants and industrial processes. The three technology lines (post combustion, pre-combustion, oxyfuel) will be described in terms of state of the art, efficiency losses and advantages and disadvantages. An outlook will be given of further developments in the long term (second generation). Special attention will be paid to retrofitting options of existing coal fired power plants. An increasing share of highly volatile renewable power generation will change the flexibility requirements of coal fired power plants. Against this background flexibility options will be discussed for power plants with carbon capture technologies.

Keywords Carbon capture • Coal power plants • Industrial processes • Retrofitting • Flexibility

2.1 Introduction

According to the United Nations' Intergovernmental Panel on Climate Change (IPCC), CO₂ emissions must be reduced by at least 50 % if the expected temperature increase caused by the greenhouse effect is to be limited to around 2–3 °C. With an ambitious reduction scenario (Blue Map), the International Energy Agency (IEA) outlines what technical measures could be used to achieve a significant reduction (IEA 2010). In order to reduce emissions to 14 GtCO₂ by 2050, a wide range of reduction measures are essential (Fig. 2.1). The capture and storage of carbon dioxide (CCS) from large point sources is considered extremely important in this context. If CO₂ reduction targets are to be achieved, CCS technologies must reduce emissions by around 8.2 GtCO₂ according to the IEA. Accounting for a 19 % emissions reduction, CCS as a single measure makes the biggest contribution to reducing emissions in the IEA scenario.

This chapter is structured as follows. First, we will describe the carbon capture technologies that can be used in coal-fired power plants (Sect. 2.2). The state of the

P. Markewitz (✉) • R. Bongartz

Institute of Energy and Climate Research – Systems Analysis and Technology Evaluation (IEK-STE), Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

e-mail: p.markewitz@fz-juelich.de; r.bongartz@fz-juelich.de

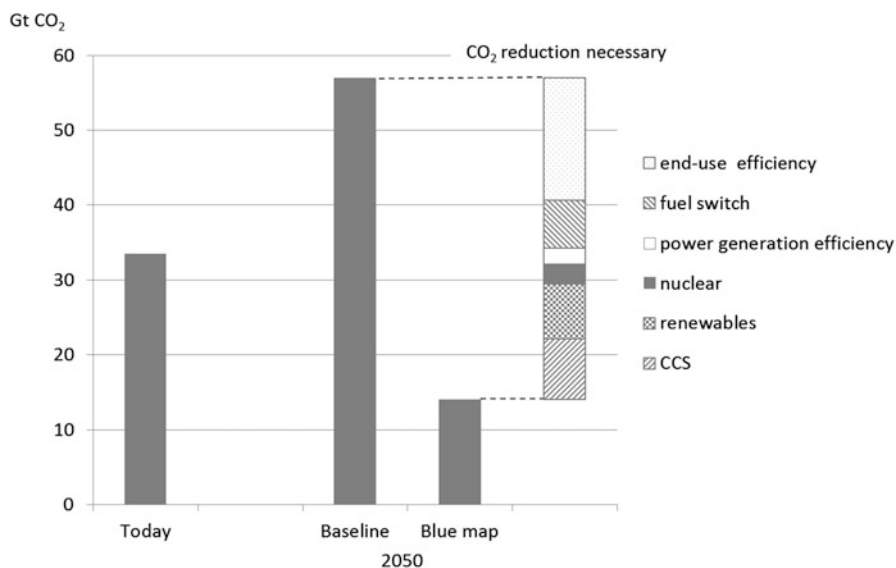


Fig. 2.1 Measures for reducing CO₂ in the International Energy Agency's Blue Map scenario (Source: IEA 2010)

art, efficiency losses and characteristics of the individual processes will be discussed. The current processes behind the three technology lines being discussed at present (post-combustion, oxyfuel, pre-combustion) will be outlined together with the 'second-generation' processes, which are still far from being ready for technical implementation but which have great development potential. Needless to say, an analysis of the operation of power plants with CCS cannot be performed in isolation, as the operating conditions must be taken into account within the context of the power generation system as a whole. For this reason, Sect. 2.3 takes a look at specific requirements (e.g. power plant operation mode, retrofitting the existing power plant fleet) that arise within a system context and are also decisive in implementing CCS technologies or CCS power plants.

As fossil-fired power plants cause a large share of global CO₂ emissions, present R&D work on carbon capture technologies concentrates on power plant application. However, the use of carbon capture processes is also conceivable in industrial processes, which likewise cause a significant share of global CO₂ emissions. Section 2.4 analyses which industrial applications and/or processes are of interest, as well as which of the capture processes currently being discussed could be used here. Carbon capture for biogas treatment, which has already been commercially implemented, will also be discussed.

2.2 Carbon Capture Technologies for Use in Coal-Fired Power Plants

Global CO₂ emissions in 2010 totalled approximately 31.3 GtCO₂ (Ziesing 2011a). With a share of 40 % of total emissions, fossil-fired power plants were the main emitters. The distribution is similar in Germany: fossil power plants produced around 42 % of total German emissions (2010: approx. 826 Mt) (Ziesing 2011b). Both the global and national CO₂ reduction targets can therefore only be achieved if CO₂ emissions from fossil-fired power plants can be significantly reduced. In addition to a transition to low-carbon fuels and an improved efficiency of power plants, carbon capture is another important reduction option. As carbon capture entails substantial efficiency losses, a high efficiency is generally desirable for the basic power plant process.

In the following, the focus will be on the carbon capture process and its integration into the power plant. For measures aiming to increase the efficiency of fossil power plants, the reader is directed to the existing literature (Markewitz et al. 2011a; Wietschel et al. 2010).

Numerous research and development projects are being conducted throughout the world on carbon capture. At present, three technical process routes are preferred (Fig. 2.2): these are post-combustion, the oxyfuel process, and pre-combustion. Other processes also exist which have a long way to go before they are ready for technical implementation but which also offer several advantages (e.g. great efficiency potential) compared to processes being developed today. They are often referred to as ‘second-generation’ processes. The three technology lines currently being discussed will be outlined here. The processes will be described in brief, as will the state of the art, the advantages and disadvantages, and an outlook will be given of further technical developments in the long term (second generation). The technical details of the processes will not be described here as these are provided in the extensive literature (see (Stolten and Scherer 2011; IPCC 2005; BMWi 2007; Kather et al. 2008)).

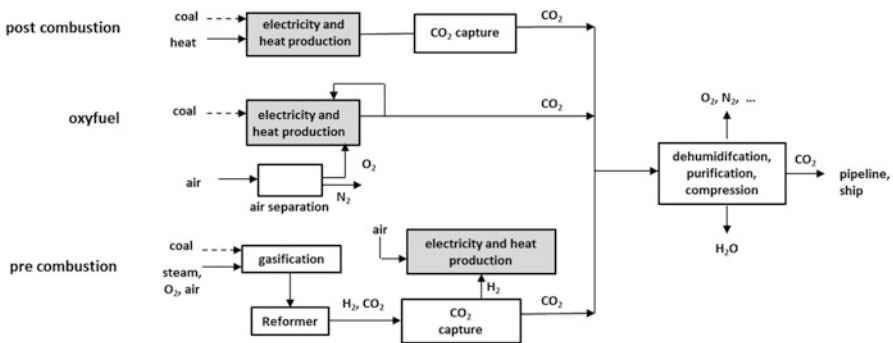


Fig. 2.2 Principles of CCS technologies

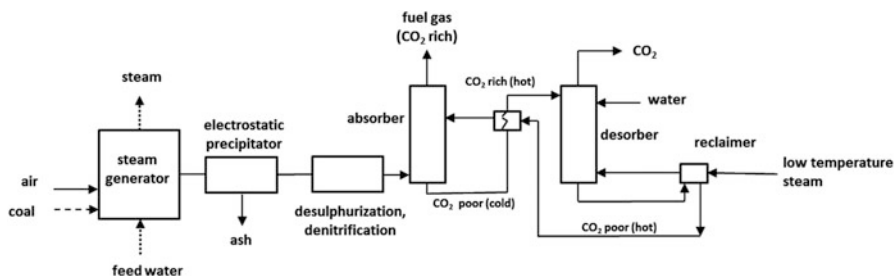


Fig. 2.3 Process flow diagram for post-combustion systems (amine scrubbing)

2.2.1 Post-combustion Processes

When carbon capture is performed after the combustion process (Fig. 2.2) including downstream flue gas purification (dedusting, desulfurization, denitrification), this is referred to as a post-combustion process. Using a suitable solvent, the carbon dioxide in the flue gas is chemically absorbed. In a second step, the CO_2 in the loaded solvent is excited by a temperature or pressure change and desorbed (Fig. 2.3). The CO_2 is then processed and compressed for transport to storage sites. The near CO_2 -free solvent is then recycled and used for a new separation cycle. Suitable solvents include organic substances (e.g. alkanolamines, known as amines) and inorganic substances (e.g. alkaline earth solutions, ammonia).

2.2.1.1 State of the Art

The main aim of ongoing research and development activities is to identify suitable solvents. Considerable experience has already been obtained with alkanolamines (e.g. monoethanolamines), which are used commercially today in chemical industrial processes (e.g. ammonia production, natural gas treatment). Unfortunately, this cannot be transferred to the power plant process as the flue gas composition and the flue gas volume flow in a power plant are very different to the requirements of a chemical industrial process. A simplified process flow diagram of an amine-based post-combustion facility is shown in Fig. 2.3. CO_2 is absorbed in an absorber at a temperature between 40 and 60 °C. The amine liquid loaded with CO_2 is then fed into a desorber. The steam for regenerating the liquid (releasing CO_2) is extracted from the power plant process at a temperature of 100–140 °C and is thus no longer available for power generation. In addition to the effort required to compress the captured CO_2 , this is one of the major reasons for the considerable efficiency losses of the process as a whole. An important development goal is to reduce the energy consumption for regeneration. Against this background, the use of sterically hindered or tertiary amines (aMDEA, MDEA, KS-1) is considered promising. They require considerably less desorption energy but have disadvantages with regard to the absorption process.

Basic problems include decomposition of the solvent in the presence of oxygen, as well as solvent degradation via reactions with sulfur dioxide or nitrogen oxide. Both effects mean that the solvent must be continuously replaced. Solvent decomposition caused by oxygen can be prevented by adding ‘inhibitors’. Reaction with SO_2 and NO_x produces degradation products in the form of salts, which are removed at high temperatures in a reclaimer (see Fig. 2.3). Degradation can be minimized by decreasing the amount of SO_2 in the flue gas before carbon capture. It is assumed that the degradation effect could be prevented at SO_2 concentrations of $10\text{--}25\text{ mg/m}^3$ (current value in coal-fired power plants: $150\text{--}200\text{ mg/m}^3$) (Rao and Rubin 2002). This in turn demands the construction of much more efficient flue gas desulfurization units (see Sect. 2.3.2).

Inorganic solvents are an alternative to amine scrubbing. The use of basic alkaline and alkaline earth solutions is an interesting alternative as these are characterized by high thermal stability, resistance to oxygen, and relatively low absorption and desorption heat. The relatively low reaction rate is disadvantageous but it can be increased by including additives. The basic feasibility of this has yet to be demonstrated (BMW 2007). A further option involves the use of ammonia. With the aid of ammonia and the addition of water, the CO_2 contained in the flue gas can be bound. The process based on the use of ammonia is characterized by a low energy demand for absorption and desorption. Furthermore, ammonia is an absorbent that can be produced cost-effectively. A variant of the ammonia-based process is the chilled ammonia process, which absorbs the CO_2 at a much lower temperature. The advantages compared to the conventional ammonia process are small volume and mass flows, and thus a lower energy demand and a lower NH_3 slip (Kozak et al. 2009).

Today, around 20 smaller post-combustion plants can be found throughout the world designed as pilot or demonstration plants. Of these, two are in operation in Germany (lignite-fired power plant Niederaussem: amine scrubbing; hard-coal-fired power plant Staudinger: amino acid salts). Another 20 post-combustion projects are currently planned worldwide (GCCS 2011). In today’s test facilities, the amount of CO_2 captured ranges from 0.125 to 500 t CO_2 per day, which corresponds to an electric power of much less than 1 MW (Fahlenkamp and Dittmar 2011). In addition to optimally integrating CCS technology in the power plant process, up-scaling to larger plants will thus be a crucial development step in the future. Within the framework of a European Union research programme for the demonstration of CCS, three post-combustion projects for coal-fired power plants (Porto Tolle, Belchatow, Maasvlakte) are being funded with plant sizes equivalent to a generation capacity of around 250 MW_{el} (ZEP 2008).

2.2.1.2 Efficiency Losses

Different authors have assessed the efficiency losses associated with carbon capture as ranging from around 9–14 percentage points (including compression, liquefaction and conditioning) (IPCC 2005; Finkenrath 2010; Kather et al. 2008). The

proportion of losses caused by the compression and liquefaction of carbon dioxide is between 2 and 3 percentage points. To improve efficiency, there are a number of options. These include the development of new solvents as well as an optimal thermal integration of the capture process (incl. CO₂ compression) into the power plant process. If this potential is fully exploited, according to (Kather et al. 2008), the efficiency losses for the post-combustion process should decrease in the optimal case to 9.1 percentage points (incl. compression) at a capture efficiency of 90 %.

2.2.1.3 Advantages and Disadvantages of Post-combustion Processes

An advantage of the post-combustion process is that extensive experience already exists for amine scrubbing from applications in chemical industrial processes. Furthermore, there is considerable potential for improving efficiency. Of all CCS technologies, the post-combustion process achieves the highest degree of purity for captured carbon dioxide (>99.99 %) based on what we know today (see Chap. 3). Although integrating a post-combustion process into a power plant process entails modifying the low-temperature steam part, it does not require fundamental changes in the power plant process. All necessary components are commercially available and no fundamental new developments are necessary. In addition, the post-combustion process (end-of-pipe technology) can be retrofitted in existing power plants (see Sect. 2.3.2). The investment costs, which are still quite high, are a major disadvantage (see Chap. 7). Another drawback is that it is not yet known how flexibly a power plant equipped with post-combustion technology can be operated (see Sect. 2.3.1).

2.2.1.4 Second-Generation Post-combustion Processes

Second-generation post-combustion processes include the carbonate looping technique and membrane-based processes. Carbonate looping is based on the principle of dry sorption. The main element is a dual fluidized bed reactor in which calcium oxide (CaO) is circulated continuously as the CO₂ carrier in a loop between the carbonator (absorption) and the calcinator (desorption). The process is based on a high-temperature reaction (600–700 °C) involving the reversible exothermic absorption of calcium oxide combined with the endothermic calcination of the calcium carbonate (CaCO₃, approx. 900 °C). In contrast to conventional scrubbing techniques, the absorption heat can be used in the power plant process, thus leading to increased efficiency. Efficiency losses are estimated by (Abanades et al. 2004) as around 7.2 percentage points (with CO₂ compression and conditioning). Ströhle et al. (2008) quote an efficiency loss of around 3 percentage points (without CO₂ compression and conditioning) at a capture rate of 70–95 %. The advantages of the process, which is currently being tested on a laboratory scale, are the high purity that can be achieved as well as its theoretical suitability for retrofitting.

Another option involves the use of membranes. Polymer and organic/inorganic hybrid membranes are the preferred membrane types for the capture of CO_2/N_2 (Reijerkerk et al. 2011). Membrane processes for the post-combustion route demand a considerable energy expenditure for flue gas compression in order to produce the required pressure difference for gas separation (cf. Göttlicher 1999). At present, membranes do not yet have the necessary characteristics (e.g. selectivities) to achieve high capture rates and purities in a single-stage membrane configuration with a reasonable energy demand. One solution that has been proposed involves a multiple-stage configuration of membrane modules and compressors/turbines. Furthermore, recirculating the retentate is one way of increasing the CO_2 concentration in the feed gas. With such concepts, efficiency losses can be limited to 7.8–9.2 percentage points depending on the respective membrane properties, and a capture efficiency of 90 % and purity of approx. 95 % can be achieved (Zhao et al. 2009, 2011).

2.2.2 Oxyfuel Processes

Oxyfuel is a term used to describe the combustion of carbon-containing fuels with pure oxygen, which achieves a high concentration of carbon dioxide. Compared to the flue gas of current power plants, which contains concentrations of CO_2 of around 12–15 vol. %, this figure is around 89 vol. % in oxyfuel plants (BMW 2007). After flue gas purification and scrubbing, the flue gas is mainly comprised of a carbon dioxide/steam mixture. By condensing the steam out of the flue gas, a CO_2 -rich flue gas is produced, which is then compressed ready for transport to the storage site. Oxygen for the combustion process is produced using a cryogenic air separation unit, which separates the oxygen out of the air via condensation at low temperatures ($< -182\text{ }^\circ\text{C}$). Combustion with pure oxygen leads to much higher combustion temperatures than in existing power plants, and it necessitates modifications in the burner and furnace chamber due to heat- and flow-specific conditions. In order to limit the combustion temperatures, some of the CO_2 -rich combustion gas is fed back into the firing chamber (Fig. 2.2). At the same time, oxygen that was not converted is fed back into the combustion process and the residual oxygen concentration in the flue gas is reduced.

2.2.2.1 State of the Art

The oxyfuel process has been tested in several test facilities worldwide, as well as in two smaller pilot plants (approx. 30 MW_{th}) in Germany (lignite-fired power plant Jämschwalde) and France (gas-fired project power plant Lacq). However, feasibility for the usual size of power plants today has not yet been demonstrated.

Cryogenic air separation units are state of the art today and are used on a large scale for industrial applications (e.g. steel industry, syngas production). Air

separation demands a high energy expenditure, which causes considerable efficiency losses in an oxyfuel power plant. The challenge therefore involves optimizing the energy demand of the air separation unit and integrating it into the power plant process. By improving cryogenic air separation from a process engineering point of view, e.g. three-column process, the specific energy demand could be decreased by 20 % compared to today's plants (BMW 2007). An important framework condition here is the required oxygen purity, which should be around 99.5 vol. % (residual components: Ar, N₂). Measures for refining conventional air separation units, however, often lead to a lower oxygen purity and a higher residual gas content. This gives rise to a need for additional gas purification, which in turn is connected with a higher energy expenditure as well as with a higher compression effort (cf. (Kather et al. 2008; Castillo 2009; Kather and Klostermann 2011)).

Combustion with pure oxygen leads to considerably lower flue gas volumes and a modified radiant heat transfer. This in turn necessitates a new design for the heat exchange surfaces, the firing chamber geometries, and the flue gas channels. In general, it is assumed that the oxygen excess is lower than for conventional firing, which could cause burn-up problems and corrosion of the combustion chamber walls.

Another problem is the infiltration of undesired air, which amounts to several percent of the total flue gas volume flow and may increase with the lifetime of the power plant. This means that the required CO₂ purity can no longer be achieved, and that additional energy is required for compression.

Several of the problems outlined here are currently being investigated in ongoing R&D work. Of particular note is the biggest oxyfuel demonstration plant in the world at the moment: a lignite-fired power plant at the Jämschwalde site with a rated thermal capacity of 30 MW. The demonstration plant went into operation in 2009, and can be operated with pure oxygen as well as with air. Tests have shown that a CO₂ purity of 99.7 % can be achieved at a capture efficiency of 90 %. Other large pilot plants (e.g. Callide Power Plant, Australia, 30 MW_{el}) are currently being planned (cf. GCCS 2011). An oxyfuel power plant is also planned for the Compostilla site in Spain as part of the EU programme for the demonstration of CCS. The envisaged design is an oxyfuel power plant with a fluidized bed reactor (phase 1: 30 MW_{th}; phase 2: 300 MW_{el}).

2.2.2.2 Efficiency Losses

The efficiency losses of an oxyfuel power plant are currently estimated to range between 8 and 11 percentage points. The operation of the air separation unit alone causes an efficiency loss of 7 percentage points. The remaining losses are due to CO₂ compression and conditioning. According to calculations performed by (Kather and Klostermann 2011), the efficiency losses are around 10 percentage points when an improved air separation unit and improved conditioning technologies are used. By optimally integrating CO₂ compression into the heat balance of the power plant, the efficiency losses can be reduced further by one percentage

point. A transition to the three-column air separation process would allow total efficiency losses to be limited to around 8 percentage points. In general, it should be noted that the efficiency should always be considered in correlation with the carbon capture efficiency, the required CO₂ purity and the oxygen purity (cf. Kather and Klostermann 2011; Castillo 2011).

2.2.2.3 Advantages and Disadvantages of Cryogenic Oxyfuel Processes

As was the case for the post-combustion process, the oxyfuel process also has considerable efficiency potential. In addition, all necessary technical components are commercially available, which means that no new developments are required. As air separation units are already being built and operated as large plant units today, the oxygen volumes required for power plant operation do not pose any problems. Compared to scrubbing processes, the oxyfuel process is less complex, which is advantageous. Furthermore, there is no need to dispose of by-products. As things currently stand, the relatively high investment costs are considered a disadvantage. Whether oxyfuel processes can also be retrofitted into existing power plants is still unclear (see Sect. 2.3.2). As was the case for post-combustion processes, no conclusive statements can be made on the flexibility of the operation mode of an oxyfuel power plant (Sect. 2.3.1).

2.2.2.4 Second-Generation Oxyfuel Processes

An alternative to cryogenic air separation is chemical looping, which uses metal oxides as an oxygen carrier and where the metal oxide is circulated continuously between two reactors in a loop. In the combustion reactor, oxygen is separated from the metal oxide in an endothermic reaction, and then used for fuel combustion. The reduced metal oxide is regenerated in a second reactor using atmospheric oxygen as an oxidizing agent at temperatures of 1,200 °C. The heat flow from the oxidation reactor and the flue gas is used to generate heat (BMW 2007). The main challenge and the focus of several ongoing research projects is the identification of suitable metal oxides with an appropriate stability and a sufficient reactivity with relevant regeneration properties (Ausfelder and Bazzanella 2008; Lyngfelt and Mattisson 2011). Eppler and Ströhle (2011) assume that the efficiency losses of chemical looping are around 8 percentage points (incl. compression).

Another alternative to cryogenic air separation is to separate oxygen using oxygen-conducting membranes. Ceramics (perovskites, fluorites) are used for this purpose as they are characterized by a specific conductivity for oxygen ions and are permeable to oxygen ions at a temperature above 700 °C. A particular advantage of this process is that relatively high oxygen purities can be achieved. In addition to membrane development, thermally integrating the membrane process into a power plant process also remains challenging. Different concepts are currently being developed and tested for this (three-end process, four-end process)

(Kneer et al. 2010; Engels et al. 2010). Depending on the concept chosen and other parameters, efficiency losses range from 6 to 10 percentage points according to the literature (Kneer et al. 2010; Engels et al. 2010; Stadler et al. 2011; Castillo 2011; Beggel et al. 2011).

2.2.3 *Pre-combustion Processes*

In integrated gasification combined cycle (IGCC) power plants, coal or other materials (e.g. refinery residues) are partially oxidized at high temperatures and under high pressure (approx. 30 bar), and converted into a raw gas (CO , H_2 and CO_2) (Fig. 2.2). A downstream catalytic conversion uses steam as an oxidizing agent to convert the carbon monoxide into carbon dioxide and hydrogen (CO shift). The resulting syngas has a high pressure level, which allows carbon capture to be performed using physical absorption on the basis of methanol (Rectisol wash) or dimethyl ether/polyethylene glycol (Selexol scrubbing). The remaining hydrogen is subsequently used in a gas and steam process to generate power (BMW 2007; Wietschel et al. 2010).

2.2.3.1 *State of the Art*

The gasification of coal and other energy carriers is state of the art, and is used throughout the world to produce syngas, which in turn is the starting material for several other industrial applications (e.g. methanol synthesis). However, gasification technology has not yet become established for power generation, and only a few IGCC power plants are commercially operated worldwide today (BMW 2007; Wietschel et al. 2010). The efficiencies that can be currently achieved are comparable to those of conventional coal-fired power plants. Due to the high efficiency potential of coal-fired combined cycle processes and the feasibility of using established and tested processes for CO_2 separation, IGCC power plants are attracting increasing attention within the context of carbon capture.

For carbon capture, the use of Rectisol wash is preferred because it can simultaneously remove CO_2 along with the undesired accompanying substances H_2S and COS (carbonyl sulfide) from the syngas. The Rectisol process comprises several process steps, such as the absorption of sulfur-containing components, the absorption of carbon dioxide, and the desorption of the loaded solvent. In addition, the sulfur-containing accompanying substances must be removed from the solvent and treated accordingly (e.g. Claus unit). The solvent here is an absorbent with a methanol basis. Absorption occurs under pressure (30–60 bar) and at very low temperatures ($-40\text{ }^\circ\text{C}$). CO_2 is separated from the loaded solvent using nitrogen and temperature changes. The CO_2 is then conditioned and compressed. The Rectisol process can generate relatively high purities of over 99 % (Kunze and Spliethoff 2010).

Syngas preparation is already common in several industrial sectors, which means that prior experience exists in this area, and the technical challenge therefore lies in implementing the basic IGCC process. Gasification technology must be improved, raw gas cooling must be refined (e.g. optimal use of waste heat, partial quench), and it must be rendered feasible to use H_2 in gas turbines (BMW 2007; Wietschel et al. 2010). As is the case for other CCS technology lines, the optimal thermodynamic design is a crucial prerequisite for exploiting the existing efficiency potential.

Planning has begun on an IGCC power plant, which is being funded within the framework of the European Union's CCS demonstration programme. The coal-fired combined power plant (Stainforth, UK) with a capacity of 650 MW_{el} is part of the Don Valley Power Project. It is currently the only planned coal-fired IGCC plant in Europe. Further IGCC projects are planned in the USA and in China (GCCS 2011).

2.2.3.2 Efficiency Losses

The efficiency losses for an IGCC power plant with physical CO_2 scrubbing compared to an IGCC power plant with no carbon capture range from 9 to 12 percentage points (Scherer and Franz 2011; Kather et al. 2008; Göttlicher 1999; Kunze and Spliethoff 2010; Grabner et al. 2010; Rubin et al. 2007). The IGCC demonstration power plant (450 MW_{br}, full quench, 40 bar), which was originally planned for the German site Hürth, was supposed to achieve an efficiency of 34 % at a gross efficiency of 48.5 %. The transition to a partial quench would have improved the efficiency by around 1–1.5 percentage points. By exploiting the considerable efficiency potential, a gross target efficiency of 44 % was reported for a capture rate of >90 % (Renzenbrink et al. 2008).

2.2.3.3 Advantages and Disadvantages of Pre-combustion Processes

A significant advantage of the pre-combustion process is that physical scrubbing (Rectisol, Selexol) is already used commercially on a large industrial scale to purify syngas and there is thus a direct analogy to IGCC power plants. The high purities that can be achieved are also a benefit of this process. In addition, the considerable efficiency potential of the basic IGCC process must also be considered, which includes gasification, thermodynamic optimization of the entire system, and the improvement of gas turbine technology (BMW 2007). Another key advantage of IGCC plants is the high fuel flexibility and the option of generating other products in addition to power (polygeneration). For example, it is possible to use syngas to produce other gaseous or liquid products (SNG, methanol, synthetic fuels, etc.). The high complexity of these plants compared to conventional power plants, however, is a major obstacle. A major barrier is also posed by very high investment costs for the basic IGCC power plant process, which also explains the low number of IGCC plants globally. High investment costs were also stated as the reason why

some planned IGCC power plants with carbon capture were not implemented in the recent past (cf. Markewitz et al. 2012).

2.2.3.4 Second-Generation Pre-combustion Processes

Due to favourable pressure conditions, membranes can be used in the pre-combustion process as a replacement for physical scrubbing in the long term. Either H_2 -selective membranes (e.g. microporous zeolite membranes, microporous sol-gel membranes, MPEC membranes) or CO_2 -selective membranes (polymer membranes) can be used (Scherer and Franz 2011). The type of membrane chosen depends mainly on the IGCC concept, as this determines the prevailing framework parameters (pressure, temperature, syngas composition, etc.). Scherer and Franz (2011) reported efficiency losses ranging between 8.7 and 10.5 percentage points for the use of CO_2 -selective membranes. For H_2 -selective membranes, the authors reported a range of 9.1–11.1 percentage points. Compared to the efficiency losses associated with scrubbing (9–12 percentage points), membranes demonstrated only slight efficiency advantages. However, the use of catalytic high-temperature H_2 membranes, which combine H_2 separation with the CO shift reaction in one engineering process, have great potential. The advantage here is that the shift reaction can occur in a stoichiometric ratio (steam to CO). Compared to present concepts, which require a super-stoichiometric ratio, this would allow the steam demand to be reduced considerably. In contrast to a Rectisol wash, no temperature decrease is required here for the conversion reaction. This allows a gain in efficiency potential of between 0.8 and 2.9 percentage points compared to the previously mentioned membrane concepts (Scherer and Franz 2011). However, hot gas dedusting must be performed upstream of the membrane reactor.

In general, it should be noted that the membrane types currently being discussed do not yet demonstrate the necessary characteristics (e.g. selectivity, permeability, stability) for implementation in various separation concepts, and that there is a need for fundamental R&D work in the field of membrane development and fabrication.

2.3 Future Framework Conditions and Requirements for the Implementation of Power Plants with Carbon Capture

While the previous section outlined the individual carbon capture technologies and evaluated them in terms of their advantages and disadvantages, Sect. 2.3.1 will analyse CCS power plants in terms of their flexibility and the main technical requirements arising from power plant operation.

In general, it is assumed that power plants with carbon capture technologies will only be available commercially from 2020 at the earliest. This means that the

installed power plant fleet in 2020 will not be equipped with CCS technology. Against this background, Sect. 2.3.2 will look at how suitable the discussed technology routes are from a technical perspective for retrofitting the installed power plant fleet and the potential that exists.

2.3.1 Flexibility of Power Plants

The liberalization of the European electricity market, which began in 1998, has meant that power plant operators have had to change track because their expected revenues now depend on market conditions on the power exchanges. This in turn has a major impact on how power plants are operated, and the demand for flexibility has increased substantially. The accelerated deployment of renewables for power generation must also be taken into account, as this will continue in future according to the plans of many countries. This large share of highly volatile power generation (wind, photovoltaic) is frequently fed into the grid preferentially in accordance with legal requirements. Covering the remaining residual load (consumer load minus volatile feed-in) necessitates controllable conventional power plant capacity with high power gradients as well as large load ranges (maximal/minimal capacity), while simultaneously accounting for a much higher number of load changes (RWE 2009).

This change in the basic conditions means that new coal-fired power plants currently being constructed will be much more flexible to operate than older existing power plants. This applies both to the power gradients and the load ranges, as shown in Table 2.1 for selected existing coal-fired power plants and new plants currently being built.

In the future, it can be assumed that not only will the number of load changes increase considerably but so too will the number of start-up and shut-down cycles (Trautmann et al. 2007; RWE 2009). According to estimates, the number of warm start-ups (>8 h downtime) and hot start-ups (<8 h downtime) will increase over the entire lifetime of a coal-fired power plant by a factor of around four and eight, respectively. Other important factors include the time required to start up and shut down a power plant. It is assumed, for example, that the usual start-up times today (cf. Table 2.2) will have to be considerably reduced (by 20–30 %) in order to meet market requirements (RWE 2009).

Table 2.1 Power gradients and load ranges for selected old and new coal-fired power plants

	Hard coal		Lignite	
	Old	New	Old	New
Maximal gradient	+/-8 MW/min	+/-27 MW/min	+/-11 MW/min	+/-30 MW/min
Maximal capacity	600 MW	800 MW	300 MW	1,000 MW
Minimal capacity	420 MW	200 MW	200 MW	500 MW

Source: RWE (2009)

Table 2.2 Start-up times of coal-fired power plants for different types of start-ups

	Start-up time
Cold start-up (after <72 h downtime)	Approx. 6 h
Warm start-up (after <48 h downtime)	Approx. 4 h
Hot start-up (after <8 h downtime)	120 min
Hot start-up (after <2 h downtime)	90 min

Source: Trautmann et al. (2007)

The requirements outlined above apply to all conventional power plant types in general and thus also to power plants with CCS technology. However, ongoing R&D activities in the area of carbon capture are currently focused on the development of CO₂ separation processes and their integration into the power plant process. Most technical and economic analyses on CCS power plants are therefore founded on a base-load mode of operation. Different publications (Chalmers 2010; Davison 2011) have therefore concluded that the flexibility of CCS power plants and the resulting technical requirements have not received enough attention and that there is a considerable need for research. Reliable estimates of temporal behaviour (start-up and shut-down cycles, load ramps) demand dynamic modelling of plants and power plant components. As almost all existing CCS power plant analyses with very few exceptions (Ziaii et al. 2009; Kvamsdal et al. 2009) have been performed with static models, a well-founded evaluation of the temporal behaviour of CCS technology routes is not yet possible. The same applies to the possibilities afforded by partial load operation, and the associated efficiency losses. Studies by Linnenberg and Kather (2009) suggest that the efficiency loss of a CCS power plant in partial load operation is greater than that of a power plant with no carbon capture in partial load operation.

The following will take a look at some options that are currently being discussed to increase the partial load flexibility of the preferred CCS technology routes in order to operate at as large a load range as possible. The discussion is based predominantly on a comprehensive literature review performed by (Chalmers 2010) on this particular topic.

2.3.1.1 Post-combustion Processes

One way of operating post-combustion power plants with a more flexible load is to bypass the carbon capture system. The low-pressure steam required for desorption could then be used for electricity generation and/or to increase the load. It should be noted, however, that the option of bypass operation must be devised when a new plant is being planned because it requires modifications in the design of the low-pressure turbines, the generator, and other components. The expected response times for load activation depend on the steam extraction concept. Economic considerations must reflect the fact that a higher load must be fed into the grid in times of high load demand and high electricity prices in order to generate higher

revenues. These revenues must be balanced against the additional investments and CO₂ allowance costs for the additional CO₂ emissions.

Another possible method of increasing load is to store the CO₂-loaded solvent. This means that desorption of the solvent loaded with CO₂ is not performed for a certain period of time, and that the solvent is stored in special tanks for this interim period. The low-pressure steam originally intended for desorption can then be used for electricity generation and/or to increase load. In times when load demands and revenues are low, the power plant output can be reduced, and desorption of the stored loaded solvent can be performed using low-pressure steam.

The two concepts outlined above are still only of a purely theoretical nature, and detailed technical and economic analyses have yet to be performed. Furthermore, in addition to carbon capture technology, compression and thermal connection to the actual steam cycle as well as possible impacts on pipeline injection and operation must also be taken into account.

2.3.1.2 Oxyfuel Processes

Current concepts are designed so that the first step involves starting up an oxyfuel power plant with air and switching over to oxygen operation as soon as sufficient flame stability has been achieved. High oxygen concentrations, which could arise in the transition phase and pose a danger of explosion, are considered a potential problem. To avoid this problem, optimized burner technology is essential. The duration of the start-up process is estimated to be around 15–30 min and it depends mainly on the burner configuration (Kluger et al. 2009; Grathwohl et al. 2009).

Both start-up and shut-down cycles as well as partial load operation assume a variation in oxygen supply. The speed of this variation also depends on the flexibility of the air separation unit, for which load ramps of 1–3 % per minute have been estimated (White et al. 2009). The load change rate could be enhanced by storing oxygen in liquid or gaseous form and then using it during periods of high load change rates.

2.3.1.3 Pre-combustion Processes

Only six IGCC plants are currently in operation worldwide, which means that only limited experience exists with coal-fired combined power plants. It is therefore almost impossible to predict the load behaviour of IGCC plants with carbon capture from our point of view today. One method of varying load would be to store part of the hydrogen temporarily, and to use it as required for high load demand. This variant would have the advantage that the gasifier would not have to meet the load demands in full. As the hydrogen is used in a gas turbine, both the load flexibility and load change rate are very high. An advantage of IGCC plants is the basic feasibility of producing other products such as methanol in addition to electricity and hydrogen (polygeneration), as this could help to boost the plant flexibility. In

the case of a low demand for electrical load, for example, operation could be switched to another product, reducing electricity generation and operating the coal gasifier in base load mode.

2.3.2 *Retrofitting the Existing Power Plant Fleet*

Some 41 % of global anthropogenic CO₂ emissions originate from power plants (IEA 2011). Around 73 % of these emissions come from coal-fired power plants, clearly indicating how important they are in the context of CO₂ mitigation strategies. In general, it is assumed that carbon capture technologies will be commercially available from 2020 at the earliest. The International Energy Agency (IEA) estimates that by 2020 the globally installed power plant capacity will increase from around 1,580 GW today by between approximately 350 GW (450 ppm scenario) and 700 GW (current policies scenario) (IEA 2011). Which of the existing coal-fired power plants could be retrofitted with carbon capture technologies, supplementing the predicted new builds, depends on the technical and economic prerequisites. It can be assumed that these will be relatively modern plants (cf. Finkenrath et al. 2012), namely those built between 1999 and today. Between 1999 and 2009, plants with a combined global power plant capacity of 525 GW were built (Fig. 2.4).

Since 2005, a global power plant capacity of around 272 GW has been installed. If these installed plant capacities are added to the additional new-build capacities predicted by the IEA by 2020, the potential to retrofit coal-fired power plants with carbon capture technology ranges from 875 to 1,225 GW. This emphasizes the

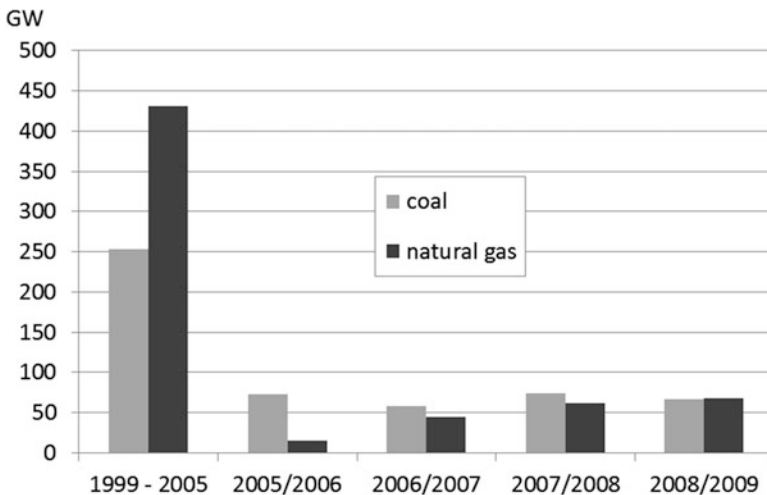


Fig. 2.4 Global expansion of coal- and gas-fired power plants (1999–2009) (Source: IEA 2000, 2006, 2008c, 2009, 2011)

growing importance of retrofitting existing power plants with carbon capture technologies.

2.3.2.1 Excursus: Germany

The installed capacity of coal-fired power plants in Germany is currently around 47 GW (lignite: 19.9 GW; hard coal: 26.8 GW). Coal-fired power plants with a capacity of 11.5 GW are currently being built and are due to go into operation by 2013 at the latest (Markewitz et al. 2011b). As a countermove, around 12 GW coal-fired power plant capacity will be taken off the grid by 2020 due to obsolescence. In the current scenario framework for the Network Development Plan approved by the Federal Network Agency for Electricity, Gas, Telecommunications, Post and Railway (Bundesnetzagentur) (Bundesnetzagentur 2011), it is assumed that coal-fired power plants will be further expanded by 2022. Depending on the scenario, additional capacity ranges between 11 and 20 GW. These capacities in particular are extremely relevant for retrofitting with carbon capture technology.

Due to the high efficiency losses associated with the use of carbon capture technologies, electricity generation drops and the net power is reduced compared to existing power plants that have not been retrofitted with CCS. These losses must be compensated by the respective operator, for example by buying additional quantities of electricity or by building new power plants. However, the limiting resource situation must be considered when building new lignite-fired power plants (Markewitz et al. 2009).

2.3.2.2 Suitability of Carbon Capture Technologies for Retrofitting

There is an ongoing discussion as to whether power plants currently being built and those planned for the near future (up to 2020) should be 'capture ready'. TÜV Nord, for example, offers certification for this (Climate Change Standard TN-CC006). This standard comprises requirements for the technological and site-specific feasibility of retrofitting a capture unit, the availability of the necessary space, the feasibility of disposing of the captured CO₂, and/or of storing it, as well as possible impacts on plant safety and the environment. Compliance with these criteria aims to ensure the basic feasibility of a retrofit. Additional investments must also be earmarked for technical measures in advance.

For such existing power plants, the question arises as to whether component design should be planned in advance in order to simplify retrofits at a later point in time from an engineering perspective and also to improve efficiency. In other words, additional investments for this at the time of power plant construction would then be offset from the point in time of the retrofit, e.g. by a higher efficiency and shorter retrofit times (see Irons et al. 2007a). From an operator's point of view, there are potential risks and uncertainties associated with advance investment (Irons et al. 2007a). These include future provisions of regulatory law, the future

development of energy prices and CO₂ allowances, and the future organization of the electricity market. From a technical point of view, there is an inherent risk that planning for components in advance would commit operators to a technology concept that may be preferred today but by the time the plant is retrofitted could be considered antiquated.

The current discussion on retrofitting concentrates on the post-combustion process and the oxyfuel process. A special case is that of IGCC power plants: although retrofitting existing IGCC power plants with CCS is currently a topic of discussion internationally, the small number of existing and planned plants worldwide means that it is still insignificant and is therefore not discussed in the following.

2.3.2.3 Oxyfuel Processes

How suitable the oxyfuel process is for retrofitting existing conventional coal-fired power plants has yet to be clarified. The degree to which higher temperatures, altered thermal radiation conditions and flue gas compositions (e.g. via fuel gas recycling) could possibly cause high-temperature corrosion or carburization of the boiler materials and thus impair the functionality and even the lifetime of the existing steam boiler is unclear. Another challenge lies in ensuring the necessary tightness of the boiler in order to prevent air ingress (cf. Irons et al. 2007a, b). Model-based simulations of the firing and the steam boiler (cf. Tigges et al. 2009) indicate that power plant operation with air or pure oxygen is theoretically possible. In addition to providing sufficient space for the installation of new components (e.g. CO₂ compressor and conditioning unit, air separation unit), (Irons et al. 2007b) also state that the burner must be replaced, and the air and flue gas channels and peripheral units must be modified (e.g. induced draught fans). No alteration of the heat exchange surfaces is necessary according to Irons et al. (2007b). It can be assumed that the flue gas desulfurization unit must be modified in order to achieve the levels of purity required for pipeline transport.

Within the framework of the Australian research project CALLIDE, an existing coal-fired power plant (site: Callide; 120 MW_{el}) is being retrofitted with oxyfuel technology. Test operation is planned under real conditions over a period of 5 years.

2.3.2.4 Post-combustion Processes

Of all post-combustion processes, amine scrubbing is the most intensively studied with regard to retrofitting existing power plants (cf. Irons et al. 2007b; Ploumen 2006). Compared to the oxyfuel process, the main advantage of the post-combustion process is that the firing process does not have to be modified. It is also assumed that no modifications are necessary for the steam generator. As previously mentioned, too much residual SO₂ in the flue gas can cause degradation of the amine-based solvent used today. In order to prevent degradation, the flue gas

should contain no more than 10 ppm ($<25 \text{ mg/m}^3$) SO_2 (Rao and Rubin 2002). Initial tests with flue gas desulfurization at the German power plant site in Niederaussem indicate that such SO_2 volumes will most likely be achievable using a lime scrubbing (REAplus) process that is very different to present flue gas desulfurization systems (Reissner 2009). Whether existing lime scrubbing systems can be retrofitted or whether they will have to be completely replaced is still unclear.

Large amounts of low-pressure steam are necessary for desorption of the amine liquid loaded with CO_2 . Thermodynamic calculations show that around 50–65 % of the total low-pressure steam in a power plant is required for this, leading to a situation where several stages of the low-pressure turbine could no longer be operated (cf. Ploumen 2006; Irons et al. 2007b). Retrofitting therefore necessitates considerable modifications of the low-pressure turbine section, the pre-heating section (heat exchanger), the condenser, and the cooling water pumps. Moreover, partial load operation poses another problem. It involves a reduced steam flow combined with a lower turbine inlet pressure in the low-pressure section. If the pressure level sinks below the pressure of the low-pressure steam required for desorption, steam must be extracted from the medium-pressure section, which then leads to additional efficiency losses (Ploumen 2006).

Compared to new power plants with carbon capture, a retrofitted existing power plant has a lower efficiency because, for example, the thermal optimization between absorption/desorption, CO_2 compression and the pre-heating section is only suboptimal.

Capture-ready measures, in contrast to retrofitting with no advance measures, can lead to substantial efficiency gains depending on the type of technical measure. According to Irons et al. (2007b), capture-ready measures for the low-pressure turbine and the flue gas desulfurization system facilitate efficiency gains of 1.4–2.3 percentage points.

Retrofitting amine scrubbing leads to an increased demand for cooling water, which is around 30 % higher than that of a conventional power plant without carbon capture (Ploumen 2006). Most of the additional water demand can be attributed to amine scrubbing components, flue gas cooling, and CO_2 compression. Flue gas cooling upstream of the absorber, in particular, leads to a clear increase in the total heat load to be dissipated. Which capture-ready measures are to be implemented for this depends on the respective cooling system. For a closed cooling loop, for example, additional space must be reserved for cooling towers. For fresh water cooling, an additional withdrawal of fresh water in accordance with the legal requirements must be possible.

2.4 Carbon Capture Processes for Industrial Applications

The development of carbon capture technologies today is generally aimed at applications in fossil-fired power plants. However, the CO_2 emissions caused by industrial processes are not without relevance. A total of around 25 % (2008: 7.4 Gt)

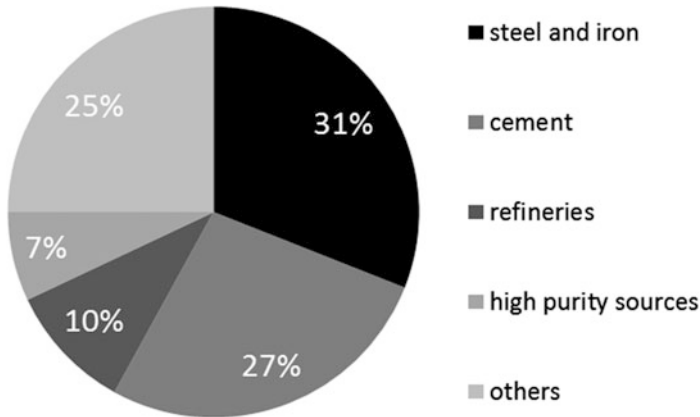


Fig. 2.5 Global share of industrial CO₂ emitters in 2008 (Source: Trudeau 2011)

of global CO₂ emissions are caused by industrial processes (Trudeau 2011). It is estimated that 75 % of these emissions (Fig. 2.5) are caused by larger point sources in the sectors iron and steel, cement, refineries and other large industrial processes (gas processing, H₂ production, ammonia production, Ctl,¹ ethylene/propylene production). According to predictions by the International Energy Agency, global emissions from these sectors will increase from around 5.6 Gt today to some 10.3 Gt by 2050 (Trudeau 2011; IEA 2010).

In industry, it is important to distinguish between energy-related and process-related emissions. Process-related emissions are those CO₂ emissions where CO₂ is created as a product of a chemical reaction other than combustion. Process-related emissions are created as a result of the material conditions of the production process (e.g. pig iron production, cement and lime production) and generally cannot be avoided. Energy-related emissions are those CO₂ emissions resulting from a combustion process (e.g. process heat and power generation).

In 2010, industry was responsible for around 18.9 % of energy-related and process-related CO₂ emissions in Germany (Ziesing 2011b). This is equal to around 156 MtCO₂. About 98 Mt (approx. 63 %) of these emissions were generated by industrial plants involved in emissions trading. Table 2.3 provides a breakdown of plants involved in emissions trading shown according to industrial sectors.

According to this breakdown, the iron and steel sector, refineries and the cement and lime branches are the largest emitters of all industrial plants which trade emissions accounting for almost 80 %. The plant-specific emissions indicate how large the point sources are. Smelting plants are the largest industrial point sources with approx. 3.5 Mt/a, followed by coking plants (approx. 0.89 Mt/a) and refineries

¹ Coal to liquid.

Table 2.3 Number and emissions of industrial plants involved in emissions trading in Germany in 2010

	Number of plants	Emissions ktCO ₂ /a		Ranking	Ø Emissions per plant ktCO ₂ /a	Ranking
				Absolute emissions %		Ø Emissions per plant
Refineries	26	22,272	22.8	1	856	3
Coking plants	4	3,568	3.7	9	892	2
Pig iron and steel production	26	5,877	6.0	5	226	6
Integrated smelting plants ^a	6	21,392	21.9	2	3,565	1
Steel processing	8	967	1.0	11	121	8
Cement clinker	39	18,577	19.0	3	476	5
Lime	69	7,757	8.0	4	112	9
Glass	85	3,701	3.8	8	43.5	12
Mineral fibres	8	356	0.4	13	44.5	11
Ceramics	134	1,327	1.4	10	9.9	14
Cellulose	5	142	0.1	14	28.4	13
Paper	122	5,715	5.9	6	46.8	10
Propylene/ ethylene	8	5,169	5.3	7	646	4
Carbon black	5	725	0.7	12	145.0	7
Total	545	97,545	100		179.0	

Source: Hohlfeld et al. (2011), own calculations

^aBlast furnace, converter, sintering and pelleting unit, hot blast stove, etc

(0.86 Mt/a). Propylene or ethylene production plants emit an average of 0.646 Mt/a and cement clinker plants around 0.48 Mt/a.² Although the lime branch is a considerable emitter, the large number of plants means that plant-specific emissions are much smaller. In general, it should be noted that a significant number of large industrial point sources are located in Germany.

Whether and which carbon capture process could help to reduce these emissions will be analysed in the following using examples of important industrial processes. A direct analogy to or transferability of the CO₂ process routes discussed previously for use in power plants does not exist. We will therefore discuss which capture processes could be useful for industrial applications. A special case is the oxyfuel process, which does not directly capture CO₂. The oxyfuel process aims at a high CO₂ concentration in the combustion off-gases by using pure oxygen for the combustion process. Whether this technique can be transferred to industrial processes will also be discussed in the following.

²For comparison, a hard-coal-fired power plant (500 MW, 4500 full load hours) emits around 1.7 MtCO₂ per year.

Treating biogas for injection into the natural gas grid is becoming increasingly more common. Biogas treatment comprises several technical process steps and includes the separation of CO₂ that arises during the fermentation process. Different carbon capture processes are already used commercially for this today. Although biogas treatment is not considered an industrial process, it is of significant relevance. It will therefore be dealt with here separately.

2.4.1 *Steel and Iron Production*

Around 1.5 Gt crude steel were produced globally in 2011. Of this, around 70 % was produced with the emissions-relevant oxygen steel process and around 30 % by the electric steel process (Wirtschaftsvereinigung Stahl 2011). In 2008, global emissions from the pig iron and steel sector amounted to around 2.3 GtCO₂ (cf. Fig. 2.5). In 2011, crude steel production in Germany was approx. 44.2 Mt, and 68 % of this was produced using the oxygen steel process. The emissions by the German pig iron and steel sector (excluding coking plants) amounted to some 30 MtCO₂ in 2010 (Table 2.3).

An integrated smelting plant for producing oxygen steel comprises several plant components, which are usually emission sources. The largest point source is the blast furnace, which accounts for around two-thirds of the total emissions of an integrated smelting plant (Fleer 2011). The CO₂ formed during the blast furnace process (energy- and process-related) is part of what is known as the top gas, which is also used for other purposes (e.g. hot blast stove, power plant). The proportion of carbon dioxide contained in the top gas depends primarily on the fuels and reducing agent used. The ranges for a typical composition are given in Table 2.4.

There are numerous options for reducing CO₂ emissions. The spectrum ranges from improved heat recovery, new coal-based processes (direct reduction of melt and iron) to the substitution of fuels (e.g. natural-gas-based direct reduction) (Fleer 2011). Carbon capture is another option.

Considerations focus on the use of CCS technologies along the blast furnace converter section. A very promising option is top gas recycling, where the CO₂ is captured using chemical or physical scrubbing, and then fed back into the blast furnace process as a reducing agent. By blowing oxygen into the blast furnace, the nitrogen concentration in the top gas is reduced and the properties of the top gas are improved (Fleer 2011; UNIDO 2010).

Table 2.4 Composition of top gas

Components	Concentration vol. %
N ₂	50–55
CO	20–28
CO ₂	17–25
H ₂	1–5

Source: Hohlfeld et al. (2011)

Another option involves converting the carbon monoxide in the top gas into CO_2 via a shift reaction and then using physical scrubbing to separate the CO_2 contained in the syngas. The remaining H_2/N_2 mixture could be utilized with the aid of a gas turbine process (Gielen 2003).

As previously mentioned, direct reduction processes are another option for steel production. Direct reduction occurs with a reduction gas based on natural gas. The reduced iron is subsequently smelted with an electric arc furnace. One advantage of the process is that coking plants would no longer be needed, which would considerably cut CO_2 emissions. The CO_2 emissions resulting from the reduction process could then be separated using conventional CCS techniques. A key obstacle to the use of direct reduction processes is the poor economic efficiency and the limited spectrum of starting materials (only high-quality iron ore) compared to conventional oxygen steel processes (Fleer 2011; UNIDO 2010; IEA 2010).

It is also possible to integrate carbon capture processes into a melt reduction process (e.g. Hlsarna smelting technology) as coal gasification with pure oxygen leads to a relatively high CO_2 concentration in the off-gas. However, this process is still in the R&D phase, which means that the classic oxygen steel process will not be replaced any time in the near future (IEA 2010).

To summarize, there are a number of options for deploying carbon capture processes for crude steel production. According to UNIDO (2010), the range of chemical and physical scrubbing processes also includes pressure swing adsorption and cryogenic processes. The combination of different processes is also believed to be possible.

2.4.2 Cement and Clinker Production

Cement is a hydraulic binder used to produce building materials (e.g. mortar, concrete). It contains clinker (approx. 70 %), which is produced in a clinker burning process, and calcium sulfate (gypsum). During cement clinker production, direct CO_2 emissions are released (approx. one-third) through rotary kiln operation. Around two-thirds of the emissions are process-related and are released in the calciner. Possibilities for reducing CO_2 emissions include optimizing the processes, fuel substitution for kiln operation, and reducing the clinker volume in cement. Another conceivable variant is the use of carbon capture processes, whereby the CO_2 in the off-gas (approx. 14–33 vol. % CO_2) is separated downstream of the clinker kiln with the aid of chemical scrubbing (IEA 2008a; Barker 2010). In contrast to a power plant, the regeneration of the loaded solvent occurs in a separate steam process. Another possibility involves operating the calciner with pure oxygen in analogy to the oxyfuel process. This concentrates the CO_2 in the off-gas, which is then cleaned and compressed. One problem associated with this process is the higher CO_2 partial pressure, which could impact negatively on the calcination reaction (Fleer 2011; IEA 2008a). Compared to today's plants, the calciner would have to be redesigned (De Coninck and Mikunda 2010).

2.4.3 Refineries

A refinery transforms crude oil into marketable products (e.g. petrol, fuel oil, kerosene, lubricants, etc.). In contrast to other industrial plants (e.g. blast furnace, cement), several refinery processes produce a large number of products. Thermal energy is required for the main process groups, namely distillation, conversion (cracking, coking, reforming), and the post-treatment and refining of products. Accordingly, the CO₂ emissions produced in a refinery are distributed among different sources (Table 2.5). In some refineries, the necessary process steam is produced in power plants where CCS technologies can be used, as discussed in the previous sections. In addition to an improved efficiency of the furnaces, improved thermal integration, better process control and CHP utilization, the use of carbon capture processes (integrated in the refinery process) has also been discussed as a further mitigation measure for the refinery process (De Coninck and Mikunda 2010).

In principle, all of the CCS technologies currently being discussed could be implemented here. For furnaces and steam generators, both chemical absorption scrubbing and physical absorption techniques are possible. The latter is often discussed in combination with a gasification of petroleum coke. A potential problem is the relatively low CO₂ concentration in the off-gas flow (Table 2.5), which could be solved by increasing the concentration using the oxyfuel process (Fleer 2011).

Approximately 5–20 % of CO₂ emissions result from the production of hydrogen, which is required for diverse cracking processes, and is currently produced using the steam reforming of natural gas or the gasification of heavy refinery residues (e.g. flexicoking) (De Coninck and Mikunda 2010). The deployment of CCS technologies (e.g. physical scrubbing) would also be feasible here.

In summary, different carbon capture systems could be implemented in different processes in a refinery. This applies in particular to off-gas flows with a high CO₂ content. In addition to the lack of economic efficiency, technically integrating these

Table 2.5 CO₂ emission sources in a typical refinery

Emission source	Description	Share of CO ₂ emissions (%)	CO ₂ concentration (vol.) in the off-gas flow (%)
Process furnaces	Heat generation via combustion of fossil energy carriers for distillation columns and reactors	30–60	8–10
Steam generators	Process steam generation via combustion of fossil energy carriers	20–50	4–15
Catalytic crackers	Burn-up of petroleum coke	20–50	10–20
Hydrogen production	Reforming of hydrocarbons to H ₂ and CO ₂	5–20	20–99

Source: Fleer (2011), Brown (2010), Van Straelen et al. (2009), own data

processes into the refinery process also poses a problem. Furthermore, the need for additional equipment could lead to space problems. We are not aware of any demonstration project at the moment testing the application of carbon capture processes in refineries.

2.4.4 Ammonia Synthesis

For the production of ammonia, hydrogen is required, which is produced in today's plants via the steam reforming of natural gas (share: 85 %) or via the gasification of solid energy carriers (share: 15 %) (IPCC 2005). The resulting syngas (CO and H₂) is then converted in another step to CO₂ and H₂. Following this, chemical absorption is used in most plants to separate the CO₂ (IEA 2008b). Around two-thirds of the total CO₂ is used in many ammonia synthesis plants for the further production of uric acid (end product: fertilizers) and thus does not enter the atmosphere directly (see contribution on CO₂ utilization). Some of the excess CO₂ is also further utilized (e.g. enhanced oil recovery). As long as the captured CO₂ is not intended for further use, it has to be prepared for storage and compressed. Ammonia synthesis including chemical scrubbing is an established process and is used throughout the world on a large industrial scale. It is estimated that worldwide around 180 Mt of unused CO₂ could be stored. For Germany, this figure is estimated to be around 3 Mt (McKinsey 2007).

2.4.5 Ethylene Oxide Production

Ethylene oxide is an important raw material that is required for the fabrication of a variety of products (e.g. ethylene glycol, organic insulating materials, textile fibres). The starting point for the production of ethylene oxide is ethylene (C₂H₄), which is usually fabricated today in refineries using naphtha steam cracking. Ethylene oxide is produced by adding oxygen and using suitable catalysts (e.g. silver) at temperatures of around 200–300 °C and a pressure of 10 bar. In addition to the main reaction, a number of side reactions occur in which carbon dioxide is formed (via over-oxidation) and must be separated in a subsequent process. In 2008, around 19 Mt of ethylene oxide were produced worldwide. In Germany, production is estimated to be almost 1 Mt. However, it is unclear how much CO₂ is produced. De Coninck and Mikunda (2010) estimates that for every unit of ethylene oxide around 0.3 units of carbon dioxide are produced. The amounts of CO₂ assumed by the IPCC (2005), on the other hand, are lower by a factor of four. Ausfelder and Bazzanella (2008) estimate that the annual global CO₂ emissions caused by ethylene oxide production amount to some 5.1 Mt.

2.4.6 *Excursus: Carbon Capture During Biogas Treatment*

In 2011, there were some 83 biogas treatment and feed-in plants in Germany, and their biomethane feed-in capacity totalled around 460 million m³ (DBFZ 2012). Around two-thirds of the plants have a plant-specific treatment capacity of 350–700 Nm³/h (DBFZ 2012). According to the German federal government's plans, biogas injection is to be increased by 2020 to 6 billion m³ by 2020.

Before biogas can be fed into the low-pressure gas grid, diverse requirements must be met. These are set down in the relevant regulations (DVGW worksheets G280, G685). This includes compliance with the combustion parameters, a set CO₂ content in the conditioned biogas (max. 6 %), a set oxygen content (max. 3 %), and a maximal permissible water content. In order to achieve as high a methane content as possible and adhere to the necessary limits, technical processing that also captures the CO₂ contained in the raw biogas is essential. The CO₂ contents in the untreated biogas range between 25 and 55 % depending on origin (Table 2.6), and is thus much higher than the CO₂ contained in flue gases from coal-fired power plants (see Section 2.2).

According to DBFZ (2012), different processes are used to capture CO₂ in Germany. Around 25 % of all plants are equipped with pressure swing adsorption, 29 % with pressurized water scrubbing (chemical absorption), 31 % with amine scrubbing, and 10 % with physical scrubbing (e.g. Genosorb®)³. While amine scrubbing and physical scrubbing for biogas treatment are still only at the prototype stage, pressure swing adsorption and pressurized water scrubbing are established commercial and technically mature processes.

Although amine scrubbing and physical scrubbing achieve the highest methane purities with extremely low methane losses, the regeneration of the loaded scrubbing liquid necessitates process heat, which is not available at every plant site. Amine scrubbing also requires upstream desulfurization of the biogas. Another drawback is that the degraded scrubbing liquid must be continuously replaced, which has a negative effect on economic viability. The use of pressurized water

Table 2.6 Biogas composition

	Share (ranges)
CH ₄	45–70 %
CO ₂	25–55 %
Air	0.05–5 %
H ₂ S	10–30,000 mg/m ³
Organic sulfur	<0.1–30 mg/m ³
Ammonia	0.01–2.5 mg/m ³
Steam (at 25 °C and 1 bar)	Approx. 3 %

Source: Urban et al. (2012)

³ Absorbent is tetraethylene glycol dimethyl ether.

scrubbing has a number of key advantages. Even though the CO₂ loading capacity of water is lower than that of amine-based liquids and larger quantities of scrubbing liquid are required, leading to higher pumping efforts, the amount of water needed can be reduced by increased pressure (higher loading capacity). Water also has the potential to dissolve other acidic components (e.g. H₂S), which does away with the need for an additional purification step. The regeneration of the loaded scrubbing water is simple and does not require process steam. It is done by relieving the pressure and stripping the scrubbing water with air (Urban et al. 2012). A key advantage over amine-based scrubbing is the almost unlimited availability and inexpensiveness of water. In addition, the waste heat can be used, for example for the fermentation process. The specific capacity of plants today ranges from 300 to 1,250 m³/h. All scrubbing processes (chemical and physical scrubbing) are connected to a subsequent gas drying unit, which is not required for pressure swing adsorption. Pressure swing adsorption compresses the raw biogas (4–7 bar) before cooling it down to below 40 °C, which considerably improves the adsorption properties. The gas is then fed through an adsorber where a carbon molecular sieve retains the carbon dioxide. The adsorbents are regenerated using a vacuum pump. The advantages of the process include high stability and the inexpensive availability of the carbon molecular sieve. Another advantage is that pressure swing adsorption needs no additional process heat. As with pressurized water scrubbing, the waste heat can be used for the fermentation process. Disadvantages include pre-treatment of the raw gas, which is necessary for high H₂S contents, and the relatively low purities (>96 %), which however still meet the relevant regulations (DBFZ 2012).

All of these deliberations show that a variety of industrial processes are theoretically suitable for separating carbon dioxide. The emissions are – depending on the industrial process – either energy-related and/or process-related. Whether or not a capture technology is suitable for use depends on the individual constraints associated with the respective industrial process. The large heterogeneity of industrial processes means that different carbon capture technologies are conceivable, as shown in Table 2.7.

Table 2.7 Carbon capture processes for selected industrial applications

	Chemical absorption	CO ₂ -upgraded oxyfuel	Pressure swing adsorption	Cryogenic	Physical absorption
Cement	×	×			×
Ammonia synthesis	×				
Natural gas treatment	×			×	×
Biogas treatment	×		×		×
Iron and steel	×	×	×		×
Refineries	×	×			×

The discussion on biogas treatment confirms that the use of carbon capture processes is not necessarily confined to large point sources, but that these processes can also be technically implemented in smaller plants – and to some extent in a cost-effective manner today. In addition, the range of techniques currently used for biogas treatment also confirms that site-specific criteria (e.g. unavailability of process heat) play a decisive role in determining which carbon capture process is ultimately implemented.

2.5 Summary and Conclusions

Over 40 % of global carbon dioxide emissions are caused by fossil-fired power plants. According to estimates by the International Energy Agency (IEA), power generation from fossil-fired power plants will continue to increase considerably in the future. In order to limit the temperature rise expected because of the greenhouse effect, carbon dioxide emissions must be drastically reduced. A key role here is played by reducing CO₂ emissions from fossil-fired power plants. Capturing carbon dioxide is an important mitigation measure in this context and is the subject of numerous research and development projects throughout the world. At present, three technology lines are favoured: post-combustion, oxyfuel, and pre-combustion. Both post-combustion and oxyfuel processes are currently being tested in smaller demonstration plants. It is not yet clear whether the transition to commercial power plant block sizes (i.e. up-scaling) will be accomplished in one stage. The plant capacity of the demonstration power plants funded by the European Union suggests that up-scaling to commercial plant sizes will probably occur in two stages.

All three technology lines have considerable potential for increasing efficiency, which can be improved depending on the process using different measures. The thermodynamic integration of the carbon capture process is particularly challenging for all three technology lines. In the long term, there are a number of interesting options that could replace physical and chemical scrubbing, which is preferred today. Alternatives include the use of membranes and carbonate looping. For the oxyfuel process, the cryogenic air separation process could be improved (three-column process) and the transition to other oxygen production processes (use of membranes, chemical looping) is also possible.

The mode of operation of a power plant will also have to meet much higher demands in the future (e.g. higher load ramps, larger load ranges, higher number of start-up and shut-down cycles). Whether power plants with carbon capture will be able to fulfil these requirements is a question that will have to be answered in the future. Due to the considerable efficiency losses caused by carbon capture, a basic power plant process with the highest possible efficiencies is generally assumed. A significant efficiency increase can only be achieved in coal-fired power plants by

increasing live steam parameters, which has adverse effects on the flexibility. Increasing the flexibility of coal-fired power plants with and without carbon capture is one of the main challenges for the future from a technical point of view.

It is generally assumed that CCS technology will be commercially available from 2020 at the earliest. Against the background of planned fossil-fired power plants worldwide, retrofitting with carbon capture technologies will play a particularly important role. Based on information available today, post-combustion processes appear to be the most promising technology line for retrofitting. A big advantage compared to other technology lines is that the modification of the power plant process would not involve too much effort.

According to estimates by the International Energy Agency, carbon dioxide emissions from industrial processes (iron/steel, cement production, etc.) will increase substantially in future. This often involves large point sources. For iron and steel production, refineries or cement production, there is a range of different options for implementing various carbon capture processes. In the long term, considerable potential is also seen here in Germany to reduce emissions by some 38 MtCO₂ by 2030 (McKinsey 2007). Envisaged applications include blast furnaces, ammonia synthesis, and clinker production. At the same time, it should be noted that these technologies are still not economically viable.

The motivation for carbon capture in biogas treatment is rather different compared to the previously discussed power plants and industrial processes. However, biogas treatment is a good example showing how carbon capture is also possible and indeed viable for smaller CO₂ sources depending on the local conditions. The processes currently in use (pressurized water scrubbing, pressure swing adsorption) are technically mature and are particularly suitable for capturing small amounts of CO₂.

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