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Recent developments on carbon capture and storage: An overview

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A B S T R A C T

The Intergovernmental Panel on Climate Change assumes the warming of the climate system, associating the increase of global average temperature to the observed increase of the anthropogenic greenhouse gas (GHG) concentrations in the atmosphere. Carbon dioxide (CO₂) is considered the most important GHG, due to the dependence of world economies on fossil fuels, since their combustion processes are the most important sources of this gas. CO₂ concentrations are increasing in the last decades mainly due to the increase of anthropogenic emissions. The processes involving CO₂ capture and storage is gaining attention on the scientific community as an alternative for decreasing CO₂ emission, reducing its concentration in ambient air. However, several technological, economical and environmental issues as well as safety problems remain to be solved, such as the following needs: increase of CO₂ capture efficiency, reduction of process costs, and verification of environmental sustainability of CO₂ storage. This paper aims to review the recent developments (from 2006 until now) on the carbon capture and storage (CCS) methodologies. Special attention was focused on the basic findings achieved in CCS operational projects.

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1. Introduction

The public concern about the climate change due to the greenhouse gas (GHG) emissions to atmosphere led to the United Nations Framework Convention on Climate Change (UNFCCC) in 1992. The main objective of this meeting was to discuss methods for stabilization of GHG concentrations in the atmosphere at a level that the anthropogenic emissions could not interfere with the climate system. Carbon dioxide (CO₂) is the most important GHG due to the dependence of world economies on fossil fuels as energy source, which led to increase the emissions of this gas to the atmosphere. For instance, CO₂ emissions from fossil fuel power generation (23 Gton-CO₂/year) represent approximately 26% of the total emissions (Holloway et al., 2007; IPCC, 2005, 2007). The stabilization of GHG concentrations in atmosphere can be achieved using non-carbon energy resources: the renewable energy such as biomass, solar and wind energy. The high costs of renewable energies associated with the abundance and availability of fossil fuels delay the introduction of this envi-

ronmental friendly energy forms. There are also some barriers for changing the technological systems, which are prepared to fossil fuel energy. Thus, in the coming years, the energy continues to be obtained mainly from fossil sources. Maintaining the main energy source, carbon capture and storage (CCS) is the most indicated technology to stabilize the CO₂ concentrations in the atmosphere. It involves the CO₂ capture at the point of generation, compressing it to a supercritical fluid, and then sequestering it. Fig. 1 shows a schematic view of CCS chain. CCS also covers biological processes, such as the use of trees or microalgae to capture CO₂ (Stewart and Hessami, 2005; Skjånes et al., 2007; Bilanovic et al., 2009). However, it is more usual to see these CCS methodologies associated with non-biological processes of capturing CO₂ from combustion.

The CCS methodologies comprise three steps: CO₂ capture, CO₂ transportation and CO₂ storage. CO₂ is captured at fixed point sources, such as power plants and cement manufacturing facilities, and different methods are studied with this aim. The most common are absorption, adsorption, separation by membranes and cryogenic separation (Steenveeldt

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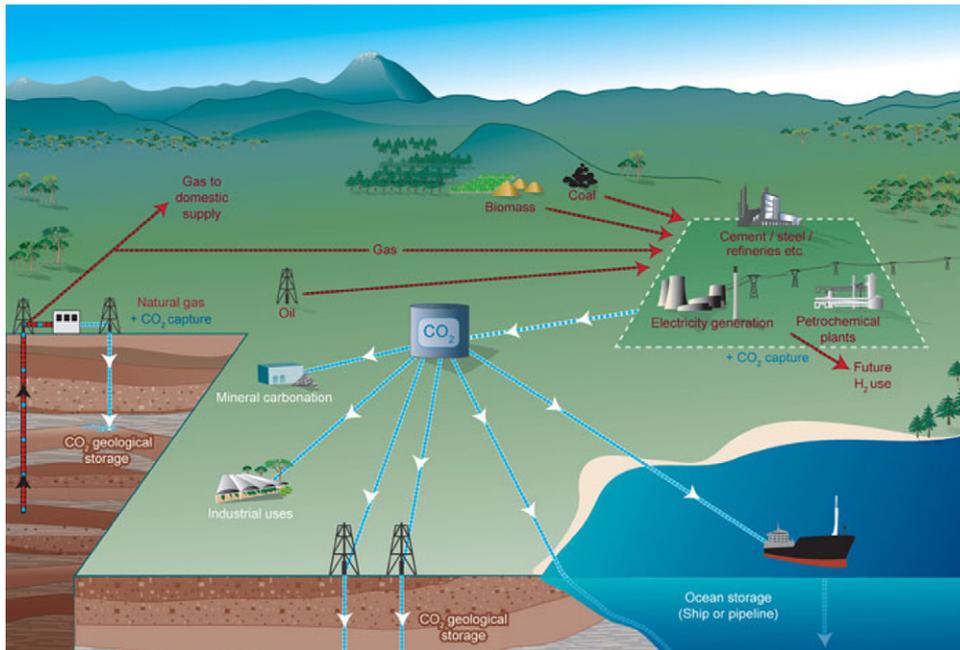


Fig. 1 – Schematic view of CCS chain (IPCC, 2005).

et al., 2006; Kanniche and Bouallou, 2007; Figueroa et al., 2008; Thiruvenkatachari et al., 2009). Then, the captured gas mixture is compressed to a liquid and supercritical fluid to be transported by pipeline or ship (Svensson et al., 2004; Zhang et al., 2006; McCoy and Rubin, 2008) to the place where it will be stored. The CO₂ storage options comprise geological storage, ocean storage and mineralization. In essence, CCS keeps CO₂ out of atmosphere by capturing it from exhaust gas and injecting it in deep reservoirs that contain fluids for thousands of years. CCS is an important technological option because it allows the societies to maintain their existing carbon-based infrastructure, while minimizes the effects of CO₂ on earth climate system. However, this technology is still under development.

The CO₂ capture represents the major fraction of the total costs, with values ranging from 24 to 52 €/ton-CO₂ (Bode and Jung, 2006). The transportation cost varies with the pipeline dimensions (length and diameter), pressure of CO₂ and landscape characteristics, ranging from 1 to 6 €/ton-CO₂ per 100 km of pipeline. The CCS total costs can vary from –3 to 106 €/ton-CO₂. The negative values are expected for the injection of CO₂ in enhanced oil recovery (EOR) fields (Solomon et al., 2008).

There is a relationship between the CO₂ emissions (CD) with the population (P), economic development (represented by gross domestic product, GDP), energy production (E), carbon based fuels used for energy production (C) and CO₂ sinks (S_{CO₂}), expressed by the modified Kaya's identity (Bachu, 2008):

$$CD = P \frac{GDP}{P} \frac{E}{GDP} \frac{C}{E} - S_{CO_2} \quad (1)$$

Eq. (1) shows that the CO₂ emissions increase with population, per capita GDP, the energy intensity of the economy (E/GDP) and carbon intensity of the energy system (C/E). There are five ways to reduce the CO₂ emissions, which the first two (reduction of population and decline of the economy output) are unacceptable as policy decisions. The increase of energy efficiency (E/GDP) and the change of the fossil fuels to non-carbon forms of energy (renewable and nuclear energy) are

measures to reduce the CO₂ emissions to the atmosphere. The fifth term, representing the CO₂ sinks, corresponds to the contribution of CCS technology. van der Zwaan and Gerlagh (2009) presented a study comparing three main options to reduce the CO₂ concentrations in the atmosphere: energy savings, a carbon to non-carbon energy switching and the use of CCS. The results showed that CCS is a valuable option, even with CO₂ leakage of a few percentages (lower than 2% v/v).

Steenneveltdt et al. (2006) presented an overview on the most important research directions for CO₂ capture and some projects that had been initiated around the world. The current paper aims to review the recent CCS methodologies (from 2006 until now). Special attention is given for the main results obtained in the cited CCS projects.

2. CO₂ capture technologies

The CO₂ capture can be performed following three different technological concepts: post-combustion capture systems, pre-combustion capture systems and oxy-fuel capture systems (Damen et al., 2006b; Kaggerud et al., 2006; Gibbins and Chalmers, 2008; Thiruvenkatachari et al., 2009). Fig. 2 shows the three technological concepts.

For the first one (post-combustion technology), the exhaust gas contains CO₂ with low concentration (4–14% v/v) represents an important limitation for CO₂ capture (Zanganeh et al., 2009; Olajire, 2010). The most used CO₂ separation process capture is the absorption using an amine as the absorbent (Damen et al., 2006b; Kaggerud et al., 2006; Gibbins and Chalmers, 2008). The low CO₂ concentrations in flue gas require powerful chemical solvents and, when applied, high energy amounts have to be expended to regenerate the solvents. It is applied to produce high purity CO₂, which can be applied in enhanced oil recovery, urea production and in the food/beverage industry. Other separation processes can also be applied. Adsorption, gas-separation membranes and cryogenic distillation are also discussed in this review.

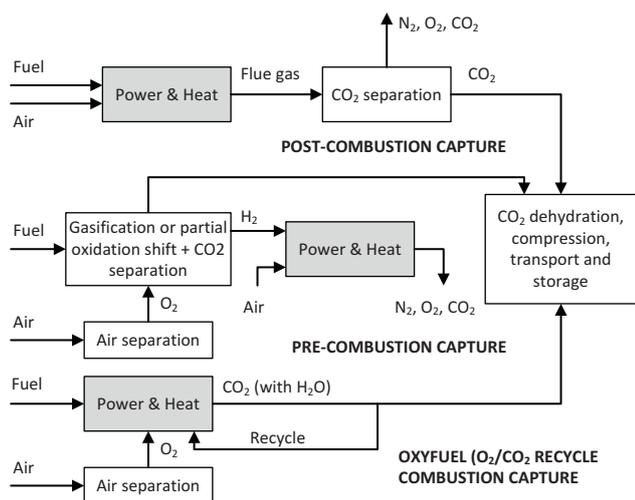


Fig. 2 – Technology concepts for CO₂ capture (adapted from Gibbins and Chalmers, 2008).

In pre-combustion capture systems, fuel is reformed by oxygen and/or steam to form a mixture of H₂ and CO₂. The CO₂ can then be separated from H₂, and the pure hydrogen is combusted with air in the power plant. The separation of CO₂ and H₂ can be done by absorption, adsorption or membranes (Blomen et al., 2009; Mendes et al., 2010). Biomass and natural gas can be used for pre-combustion capture technology. The aim of these systems is to convert the carbon fuel to carbonless fuel (Olajire, 2010): the chemical energy of carbon is transformed to chemical energy of hydrogen. An important advantage relative to post-combustion systems is the higher CO₂ concentration and pressure achieved in the output stream. Thus, the applied equipment to separate CO₂ from the referred stream can be smaller and different solvents can be used with lower energy penalties for refrigeration. The main disadvantage of pre-combustion capture is the high investments costs (Olajire, 2010).

For oxy-fuel combustion, the process is performed with pure oxygen (keeping nitrogen out of the system) produced using a cryogenic air separation or membranes (Pfaff and Kather, 2009). The combustion products are essentially CO₂ and H₂O, which are separated by condensing water (Zanganeh et al., 2009). The combustion with pure oxygen is associated to high temperatures when compared with the combustion with air (Kather and Scheffknecht, 2009). The nitrogen present in air is the major heat sink. In oxy-fuel process, cooled flue gases are used as heat sink instead of atmospheric nitrogen. The recycle of CO₂-rich flue gas reduces the temperature to usual values. Consequently, the concentration of CO₂ in the output stream is high, with values above 80% v/v (Olajire, 2010). Moreover, NO_x is not formed, which is another advantage to post-combustion systems (Kather and Scheffknecht, 2009). The main costs of oxy-fuel technology are related with the separation of O₂ and N₂ with the air separation unit. The cryogenic distillation is a very expensive process and requires high energy consumption (Bolland and Undrum, 2003; Blomen et al., 2009; Burdyny and Struchtrup, 2010; Zhu et al., 2010). To reduce the energy needed in this separation process, Burdyny and Struchtrup (2010) proposed a hybrid system composed by an O₂/N₂ permeable membrane and the cryogenic distillation. The membrane process obtains a stream of oxygen enriched air, which is turned into high purity oxygen using cryogenic distillation.

3. CO₂ separation techniques

The technologies referred in the previous section include physical and chemical CO₂ separation processes. This study focuses the most common processes: (i) absorption; (ii) adsorption; (iii) gas separation membranes; and (iv) cryogenic distillation. The hybrid system based on membranes associated with cryogenic distillation is also mentioned in this review. Other hybrid processes (membranes associated with solvent absorption) and chemical looping combustion are not included in this study.

3.1. Absorption

Absorption is a physical or chemical process in which atoms, molecules or ions are dissolved in a bulk phase. This is a different process from adsorption, since molecules undergoing absorption are taken up by the volume, not by the surface (as in the case for adsorption). Absorption is a common process in the chemical industry and it is used among others in the treatment of the industrial gas streams containing acid gases like H₂S, NO_x and CO₂ (Majchrowicz et al., 2009). In these gas treating processes, aqueous solutions of particularly alkanolamines are commonly used (Damen et al., 2006b; Kaggerud et al., 2006; Gibbins and Chalmers, 2008). These systems are also important to capture CO₂ from flue gas in a regenerative absorption-desorption process. However, absorption has some drawbacks (Knudsen et al., 2009): (i) the applied solvents have limited cyclic CO₂ loading capacity; (ii) they promote the corrosion of the equipment; (iii) solvent regeneration requires high energy consumption; (iv) a significant amount of solvent is lost by evaporation; and (v) the solvent degrades in an oxygen rich atmosphere. The environmental impact assessment of an absorption-based CO₂ capture was performed by Thitakamol et al. (2007) giving some recommendations to reduce the impacts of this process.

Between the several technologies for CO₂ capture from flue gas, the absorption process with monoethanolamine (MEA) could be the first to be available for immediate industrial applications in the next few years (Kittel et al., 2009). The EU project CASTOR (2004–2008) aimed the development of new technologies for separation CO₂ from flue gas and its geological storage. A 1 ton-CO₂/h absorption pilot plant was tested to study the post-combustion capture technology for a coal-fired power station in Denmark (Knudsen et al., 2009). Moreover, this pilot plant was used to test new energy efficient solvents having 30% MEA and other amine solvents. The total operation time was about 4000 h. The results showed that it is possible to maintain this process for extended periods achieving 90% v/v CO₂ capture. The required energy for solvent regeneration was found to be 3.7 GJ/ton-CO₂ and MEA consumption was 1.4 kg/ton-CO₂. In this process, the corrosion is the main problem, being MEA more corrosive than secondary or tertiary amines. The flue gas contains oxygen that reacts with amines and produces corrosive degradation products. The corrosion was evaluated in pilot plants using MEA as solvent, identifying the equipment that is more susceptible for this operational problem (Kittel et al., 2009). Amine blends and other type of amines were tested to overperform the MEA absorption. Dey and Aronwilas (2009) studied the mass transfer in CO₂ absorption using blends of MEA and 2-amino-2-methyl-1-propanol, developing an empirical correlation with important process parameters: liquid flow rate, temperature and total

Table 1 – CO₂ capture based on absorption processes.

Process	Relevant issues	Reference
Amine-based process	Study of CO ₂ recovery as function of several operational parameters in a pilot plant of 2 ton-CO ₂ /day for CO ₂ recovery.	Lee et al. (2008)
	Evaluation of future cost reductions of amine-based CO ₂ capture process.	Rao et al. (2006)
	Development of tertiary amine based absorbents with high absorption rate and low heats of reaction.	Chowdhury et al. (2009)
	Experimental study of CO ₂ capture in a pilot plant using three solvents.	Mangalapally et al. (2009)
Chilled ammonia process	Initial operation of the chilled ammonia process (CAP) to capture 35 ton-CO ₂ /day from flue gases.	Kozak et al. (2009)
	Extended UNIQUAQ electrolyte model to describe the properties of the NH ₃ -CO ₂ -H ₂ O system.	Darde et al. (2010)
	Application of an approximate model to evaluate mass, energy and entropy flows.	Valenti et al. (2009)
	Thermodynamic analysis and process simulation of the chilled-ammonia process.	Mathias et al. (2010)
Carbonation/calcination cycles	Experimental results from a small test facility (30 kW) operated in continuous mode using two interconnected CFB reactors as carbonator and calciner.	Alonso et al. (2010)
	Integration and evaluation of a power plant with a CaO-based CO ₂ capture system.	Yang et al. (2010b)
	Study of CO ₂ capture from flue gas in a small bed reactor with limestone.	Fang et al. (2009a)
	Process analysis of CO ₂ capture (heat and mass balances) using three sorbents.	Li et al. (2008b)
Amino acid salt solutions	Test of amino acid salts for CO ₂ capture from flue gas under varying operational conditions.	Majchrowicz et al. (2009)
	Evaluation of the CO ₂ capture of amino acid salts with the addition of activation agents (phosphates).	Lu et al. (2009)
	Study of the kinetics of CO ₂ absorption in several amino acid salts at temperature of 298 K.	van Hoist et al. (2009)
	Assessment of CO ₂ capture potentiality of amine amino acid salts.	Aronu et al. (2010)

amine concentration. Lee et al. (2008) tested several amine solvents to capture CO₂ from flue gas of a power installation unit using a 2 ton-CO₂/day pilot plant. However, even varying some experimental conditions, the best CO₂ recovery was achieved with primary amines. Freeman et al. (2010) study the CO₂ capture using concentrated aqueous piperazine (novel amine solvent) and achieved fast kinetics and low degradation rates when compared with MEA process. Another important research was the study of the solvent degradation, which main mechanisms were proposed by Lepaumier et al. (2010).

An alternative to amine-based process is the chilled ammonia process (CAP) that is a solvent-based regenerable process that absorbs the CO₂ from the flue gas at low temperatures (0–20 °C) using ammonia as solvent. In this temperature range, the precipitation occurs in the absorber. The desorption takes place at high temperatures (100–200 °C; Darde et al., 2010). Comparing CAP to the previous process, the heat of absorption of CO₂ is significantly lower; thus, this process shows good perspectives for low energy requirement. Powerspan Corp. has designed a post-combustion process for CO₂ capture using ammonia-based solution. The constructed pilot plant was designed to capture 20 ton-CO₂/day from the 50-MW Burger Plant ECO unit (McLarnon and Duncan, 2009). CAP was also implemented in We Energies power plant using a pilot plant constructed by Alstom (USA) and aiming to capture 35 ton-CO₂/day from flue gases. Kozak et al. (2009) presented the results from the initial operation of this pilot plant. The extended UNIQUAQ electrolyte model was applied to better understand the phenomena that occur in this process (Darde et al., 2010). This model was developed by Thomsen and Rasmussen (1999) to describe the properties of the NH₃-CO₂-H₂O system (temperature range from 0 to 110 °C and pressure up to 100 bar). The results showed that: (i) ammo-

nium carbonate and bicarbonate precipitates in the absorber; (ii) the gas phase in the absorber contains a high fraction of ammonia (a procedure should be defined to avoid the emission of ammonia); and (iii) CAP allows for a significant reduction of the energy consumption in the desorption when compared to the energy consumption of the process using amines.

An interesting alternative for CO₂ capture is the process based on carbonation/calcinations cycles. It consists in two fluidized-bed reactors (absorber and regenerator) operating at atmospheric pressure. In absorber, CaO is carbonated to CaCO₃ at a temperature of about 600–700 °C. The absorbent is regenerated by calcinations of CaCO₃ that produces a concentrated stream of CO₂ at higher temperature (greater than 900 °C). This process presents high efficiency (greater than 80%) and it is considered an economically competitive technology (Li et al., 2008b; Lu et al., 2008; Fang et al., 2009a; Manovic et al., 2009b). The major concern is the decay of absorbent activity with the carbonation/calcinations cycles. Blamey et al. (2010) presented some processes to reduce the rate of decay in sorbent reactivity, such as thermal pre-treatment and chemical doping of natural sorbents. However, even fully degraded, the uptake of CO₂ is higher than many other sorbents. According to the same reference, several pilot plants using this process have been constructed around the world.

Other absorption process is CO₂ capture using amino acid salt solutions. These solutions are characterized by: (i) low volatility (due to the ionic nature) and higher surface tension; (ii) higher stability in the presence of oxidative compounds (smaller production of toxic degradation products); and (iii) high chemical reactivity with CO₂ (formation of solid precipitates when absorbing CO₂; Majchrowicz et al., 2009). The capture of CO₂ from flue gas using amino acid salts was studied under different operating conditions (Majchrowicz et al.,

2009). From this study, it was concluded that: (i) higher temperatures and lower CO₂ partial pressures lead to higher solubility limits; (ii) at higher amino acid salt concentration precipitation starts at lower CO₂ partial pressures; and (iii) the resulting precipitates are composed by the amino acids and more complex species (CO₂-containing). Some relevant issues concerning recent studies about CO₂ capture based on absorption processes is presented in Table 1. The energy requirement is an important drawback of absorption process and other technologies are being studied.

3.2. Adsorption

Adsorption is also considered a feasible process for CO₂ capture at an industrial scale (IPCC, 2005; Drage et al., 2009). It is a selective process in which molecules contained in liquid or gaseous mixtures adhere on a solid surface, the adsorbent. These molecules, even in small concentrations in the streams, can be captured by these selective materials. The properties of the adsorbed particles (molecular size, molecular weight and polarity) and the adsorbent surface (polarity, pore size and spacing) determine the adsorption quality. As the adsorption is an exothermic process, the regeneration of the adsorbents through desorption can be performed by rising the temperature. However, adsorption presents lower energy requirements and avoids the shortcomings when compared to absorption (Drage et al., 2009). In post-combustion process, adsorption can be applied to capture CO₂ from flue gases. On the other hand, in pre-combustion process, the most important reactions are the steam reforming of hydrocarbons and the water-gas shift reaction to produce H₂ (Damen et al., 2006b; Mendes et al., 2010). To increase the yields of H₂, the CO₂ is captured to shift the reaction equilibrium to the right according to Le Chatelier's principle. This improvement has several benefits: (i) the reaction may be performed in a single reactor, which reduces capital costs, (ii) the reduction of steam needed in the reaction, and (iii) the process captures CO₂, which is environmentally friendly and may eliminate the need for a dedicated CO₂ capture unit (Damen et al., 2006b).

In the industrial context, a continuous CO₂ capture process should be applied and the technologies often studied are the cyclic processes. These adsorption processes can be grouped in two types, according to the way in which the adsorbent is changed between the adsorption and desorption steps: by changing the pressure or the temperature. In pressure swing adsorption (PSA), the adsorption is performed at pressures higher than atmospheric pressure. The desorption occurs at atmospheric pressure. However, a vacuum desorption pressure can be applied and, for commercial adsorbent 13X, the costs of CO₂ capture are comparable with the cost of MEA absorption (Ho et al., 2008b). The operation with an adsorbent with higher adsorption capacity and higher selectivity to CO₂ would increase the profitability of PSA. The vacuum swing adsorption (VSA) operates at near-ambient temperature and pressures and the desorption is performed at lower pressure. It is a promising technology due to the relatively low power consumption and ease of operation (Li et al., 2008a; Zhang et al., 2008). The main drawback of adsorption processes (both PSA and VSA) in post-combustion CO₂ capture is the need of cooling and drying the flue gas (and also, in some cases, the removal of SO_x which represents poison for several adsorbents). For VSA, the presence of water can drop significantly the process productivity and the formation of a "water zone" increases the vacuum level, reducing the CO₂ capacity (Li et al.,

2008a). However, CO₂ capture can be performed using VSA without pre-treatment of the flue gas (to remove the impurities). Zhang et al. (2009) performed experiments where CO₂, NO_x, SO₂ and H₂O were captured in the same VSA column within different function layers. The application of a lower vacuum level resulted in high purity CO₂ stream and high CO₂ recovery. Moreover, NO desorption is quick and influenced by the O₂ concentration in the gas mixture and SO₂ desorbs slowly due to the high affinity to the adsorbent surface. However, the interactions of the different species in the process should be studied and more cycles should be developed to evaluate the decay of the process performance.

For all adsorption processes, the success of the adsorbent is defined by their CO₂ selectivity and adsorption capacity at flue gas temperature. Two parameters that are also important are the spent time for regeneration and the adsorbent lifetime. The regeneration for PSA and VSA is performed by changing the pressure. As the captured CO₂ should be pressurized to be transported and stored, the application of these adsorption technologies results in a large energy penalty (Drage et al., 2008). Thus, alternative processes for regeneration can be considered by changing temperature: temperature swing adsorption (TSA) and electric swing adsorption (ESA). In ESA, the increase of temperature is performed by passing electricity through the conductor (Joule effect). In both cases, the regeneration of the adsorbent is performed by reducing the equilibrium capacity of the material. Several materials were tested for TSA to reduce the required energy for regeneration (Grande and Rodrigues, 2008; Drage et al., 2009; Grande et al., 2009a, 2009b; Sjostrom and Krutka, 2010), evaluating their potentiality for CO₂ capture. Besides the good results achieved in these studies, the reduction of regeneration time (an important drawback of TSA) should be studied. For TSA, the regeneration may take hours, while for PSA, it can be performed in few seconds. For that reason, some reports considered that PSA is technically and economically more viable than TSA (IPCC, 2005; Othman et al., 2009; Lackner et al., 2010).

Besides the cyclic processes, several authors have been concerned about the performance evaluation of CO₂ capture using different materials. Carbon nanotubes were tested with and without surface modification to increase the affinity to CO₂ (Su et al., 2009). This adsorbent has high adsorption capacity of pollutants due to their pore structure and existence of several surface functional groups which can be achieved by chemical modification or thermal treatment. However, the presence of water and the high temperature reduce the adsorption capacity. The adsorption is mainly physical; thus, less energy is required for regeneration of the adsorbent. Several hydrotalcites (Mg/Al/Zr containing materials for use as carbon dioxide sorbents) were tested to remove CO₂, enhancing the methane steam reforming (Oliveira et al., 2008) and the water-gas shift reaction (van Selow et al., 2009). In the methane steam reforming reaction, a sorption mechanism was proposed, assuming the combination of physical adsorption and chemical reaction. This adsorbent presented a small loss after 75 sorption/desorption cycles. In water-gas shift reaction, the carbon monoxide conversion increases from 55% to 100% in the presence of the adsorbent. In this study, the adsorbent continued to be stable after 500 cycles, presenting high reaction conversions without changing the operating conditions. Additionally, several adsorbents have been proposed for CO₂ capture, including carbon fibre monolithic adsorbents (Thiruvengatchari et al., 2009), activated carbon fibre-phenolic resin composites (An et al., 2009),

Table 2 – CO₂ capture based on continuous adsorption processes.

Process	Relevant issues	Reference
Pressure/vacuum swing adsorption	Evaluation of the economical feasibility of PSA for CO ₂ capture from post-combustion power plant flue gas.	Ho et al. (2008b)
	Simulations to evaluate the effect of mass transfer on CO ₂ capture from flue gas by PSA.	Reynolds et al. (2006)
	Evaluation of the effect of feed gas temperature, evacuation pressure and feed concentration on the performance of CO ₂ capture by VSA.	Zhang et al. (2008)
	Evaluation of the effect of operating and design parameters of VSA on CO ₂ capture by a numerical model.	Xiao et al. (2008)
	Development of a model targeted for CO ₂ capture using PSA and validation with experimental results achieved with a pilot scale PSA.	Zhang and Webley (2008)
	Investigation and report of the effects of impurities on process performance such as CO ₂ purity, recovery and process power.	Zhang et al. (2009)
Thermal/electric swing adsorption	Performance evaluation of 24 different sorbent materials for CO ₂ capture in a cyclic temperature swing adsorption.	Sjostrom and Krutka (2010)
	Experimental investigation of post-combustion CO ₂ capture by TSA with internal heat-exchanger using 13X and 5X zeolites.	Merel et al. (2008)
	Comparison of different methods of desorption in experiences of CO ₂ capture using TSA with 5A zeolite: by heating, purge or vacuum.	Tlili et al. (2009)
	Determination of suitable operating conditions of ESA for CO ₂ capture from flue gases to reduce the size and energetic consumption of the capture plant.	Grande et al. (2009b)

melamine–formaldehyde highly porous adsorbents (Pevida et al., 2008, 2009), amine immobilized adsorbents (Chatti et al., 2009), red mud (Yadav et al., 2010), between others (Drage et al., 2007; Liu et al., 2007; Singh et al., 2009). In this context, Sjostrom and Krutka (2010) compared several adsorbents, showing that carbon-based materials present excellent stability, while zeolites work well under dry conditions. However, no type of material can be applied for CO₂ capture without concerns. Table 2 presents some relevant issues of recent studies about adsorption processes for CO₂ capture.

3.3. Gas separation membranes

The separation process using membranes is a low cost technology, when high purity gas streams are not fundamental. Several studies have been published comparing the performance of amine absorption-based and membrane in the CO₂ capture from flue gases (Bounaceur et al., 2006; Favre, 2007; Okabe et al., 2008). The gas permeation processes have several advantages: (i) higher separation energy efficiency relative to equilibrium-based processes; (ii) current application to industrial processes (air separation, hydrogen recovery and carbon dioxide capture from natural gas); and (iii) high packing density which requires small installations (Bounaceur et al., 2006). The separation of CO₂ from N₂ was already studied and is easily performed by membranes with high CO₂ permeance and CO₂/N₂ selectivity. However, the treatment of an enormous flow rate of flue gas emitted by power plants requires a very large membrane area and consequently increases the cost of this capture technology. Another drawback is the need of very large, expensive and energy-consuming compression equipment. This limits the maximum pressure ratio attainable by feed compression and/or permeate vacuum to about 10. Merkel et al. (2010) developed membranes in collaboration with the United States Department of Energy with CO₂ permeances ten times higher than commercial CO₂ membranes and CO₂/N₂ selectivity of 50 at 30 °C. Those authors considered the increase of membrane permeance an important step for the reduction of the membrane technology cost to capture CO₂ from flue gas.

Polymeric membranes are of particular interest in gas separation applications. They separate gases based on the

solution-diffusion mechanism and are typically limited by the well-known relationship between permeability and selectivity. The polymeric membranes present several advantages: (i) low cost; (ii) high performance separation; (iii) easy of synthesis; and (iv) mechanical stability (Scholes et al., 2009; Xomeritakis et al., 2009). Powell and Qiao (2006) presented a review about the polymeric membranes for CO₂ capture from flue gases (separation of CO₂/N₂). The authors focused on design strategies, synthesis, fabrication and role of novel materials. Concerning the separation of CO₂ from flue gases, polymeric membranes cannot be applied without cooling of the gas mixture because high temperatures rapidly destroy the membrane. These membranes can also be applied to pre-combustion processes. In steam reforming reaction, membranes separate CO₂ from hydrogen, contributing to increase of reaction yield. In oxy-fuel combustion systems, the membranes can be applied to produce oxygen-enriched streams from air. These two applications are attempts to increase the CO₂ concentration in the output stream and, consequently, to increase the CO₂ capture system performance (Powell and Qiao, 2006).

Bounaceur et al. (2006) assessed that the energy demand for CO₂ capture using polymeric membranes can be reduced substantially when compared to amine absorption-based process. However, this achievement was only valid for recovery ratio and permeate composition less than 0.8 and feed CO₂ composition greater than 0.2. This feed composition can be found in the flue gases of some industries: cement industry, steel production, etc. The cost associated to membranes can also be reduced if vacuum pumping is applied in place of upstream compression. When the CO₂ purity is an important requirement, the amine absorption is the best option. To improve the performance of the membranes, Favre et al. (2009a, 2009b) proposed the CO₂ separation from the fuel combustion with oxygen-enriched air. High CO₂ purities and recoveries were achieved (greater than 90%), as the flue gas presents a higher CO₂ concentration due to the absence of N₂. For separation of CO₂ from flue gas, minor components influence the gas permeation performance. Water presented in the flue gas reduces the CO₂ recovery, through competitive sorption and plasticization of the polymer (Scholes et al., 2009, 2010). Besides the stream humidity, the presence of SO₂ also decreases the mem-

Table 3 – CO₂ capture based on separation with membranes.

Process	Relevant issues	Reference
Membrane separation	Review about polymeric membranes to separate CO ₂ /N ₂ gas mixtures.	Powell and Qiao (2006)
	Comparison of dense polymeric membranes with amine absorption for CO ₂ capture in post-combustion situation.	Favre (2007)
	Evaluation of the cost reduction of CO ₂ capture from flue gases using membrane separation under vacuum conditions.	Ho et al. (2008a)
	Performance evaluation of selective membrane with mobile and fixed amine carriers to capture CO ₂ from industrial gas mixtures.	Huang et al. (2008)
	Description of the mass and energy balance equations and simulation of the membrane performance in different operating conditions.	Zhao et al. (2008)

brane permeance (Xomeritakis et al., 2009). Table 3 presents some relevant issues about membrane separation studies concerning the CO₂ capture.

3.4. Cryogenic distillation

Cryogenic distillation is an air separation process, where gaseous components of a mixture are separated by condensation. The cryogenic temperatures are obtained by a closed-cycle operated refrigeration system consisting of a compressor, a Joule–Thompson valve (JTV), multi-stage heat exchangers and expanders. Cold traps (filters) of different cryogenic temperatures are built into the system to condense air components of different condensation temperatures (Pfaff and Kather, 2009).

The currently applied technologies for CO₂ capture remove this gas at ambient pressures. Thus, the CO₂ should be compressed to levels needed for transport and storage. Cryogenic method can capture CO₂ in a liquid form, being easier to transport and storage or send for enhanced oil recovery fields (Hart and Gnanendran, 2009; ZareNezhad and Hosseinpour, 2009). The CryoCell® process, developed by Cool Energy Ltd (Australia), was tested in collaboration with industrial partners (Hart and Gnanendran, 2009). This process eliminates the water consumption, usage of chemicals and corrosion related issues.

The cryogenic process can be also applied as a post-combustion CO₂ capture process (Khuo and Tan, 2006; Tuinier et al., 2010). Tuinier et al. (2010) proposed a cryogenic process using dynamically operated packed beds. CO₂ and H₂O are separated from flue gas on the basis of differences in dew and sublimation points. The main advantage of this process is the simultaneous separation of CO₂ and H₂O from flue gas, avoiding the use of solvents and high pressures. The principal concern of this methodology is that the water content in feed stream of cooling units should be minimized to prevent the plugging by ice or the high rise of the pressure drop during the operation. The main cost of this methodology is the refrigeration. It can be avoided when exploiting the cold duty available at liquefied natural gas (LNG) regasification sites. Currently, LNG is being regasified using sea water or by using water baths which are heated by burning fuel gas.

Another application to cryogenic distillation is the separation of acid gases from natural gas. The increase of the world energy demand leads to the production of natural gas from fields that were not economically attractive until now (ones that present high content of acid gases). Thus, the separation of these compounds from methane represents a new challenge, which is equivalent to the one for separation of CO₂ from flue gases. Northrop and Valencia (2009) proposed a technology involving a single-step process with this aim.

This technology is able to separate methane with no limitation in the amount of CO₂ and H₂S in the feed gas. The acid gas components are discharged as a high pressure liquid stream, providing a more economic transport and sequestration (geo-sequestration or enhanced oil recovery purposes). Table 4 presents some relevant issues about cryogenic separation studies concerning the CO₂ capture.

4. CO₂ transport

The transport of CO₂ is a mature technology as the technical requirements are similar to those applied to other gases transport. Commonly, the CO₂ is not stored in the same place where it is captured. Thus, it should be transported and, depending on the distance between the two places (point of capture to storage site), this could be done by pipeline, ship or tanker trucks. To transport large volumes of CO₂, the pipelines are considered to be the most cost-effective and reliable method (McCoy and Rubin, 2008; Haugen et al., 2009). However, in some situations or locations, CO₂ transport by ship may be economically attractive, particularly when the CO₂ has to be moved over large distances or overseas. Vandeginste and Piessens (2008) published a review about the CO₂ pipeline transportation and revealed that the pipeline diameter is the crucial parameter for cost estimation of this transport method. Additionally, quantitative risk assessment for CO₂ pipeline transportation was evaluated in several studies, some of them in the context of CCS projects (Chrysostomidis et al., 2009; Koornneef et al., 2009; Koornneef et al., 2010b).

Several millions of tonnes of CO₂ are already transported by pipelines, most of it being transported to enhanced oil recovery (EOR) fields. Pipelines linking several industrial regions can be shared, allowing the greatest emission reductions for the lower cost. A computer tool for economic analysis was developed within EU-funded GeoCapacity project to evaluate the CO₂ transportation systems based on low-cost pipeline networks to connect sources of CO₂ and storage reservoirs (Kazmierczak et al., 2009). Additionally, an engineering-economical model was proposed to evaluate the cost per ton of transporting CO₂ for a range of CO₂ flow rates, over a range of distances in the United States (McCoy and Rubin, 2008). Atmospheric dispersion of carbon dioxide after sublimation from a dry ice bank is of concern when dealing with safety criteria for the transportation of carbon dioxide in Carbon Sequestration projects. The outturn of this paper confirms that cold gaseous CO₂ could result in human safety problems for particular conditions (Mazzoldi et al., 2008; Chrysostomidis et al., 2009).

Before the transport, the CO₂ stream is conditioned to remove impurities and compressed into supercritical form. Captured CO₂ may contain impurities such as water vapour,

Table 4 – CO₂ capture based on cryogenic distillation.

Process	Relevant issues	Reference
Cryogenic distillation	Evaluation of the impact of the air separation technology (high temperature of membranes or cryogenic distillation) on the efficiency of oxyfuel steam power plant.	Pfaff and Kather (2009)
	Presentation of a pilot plant for CO ₂ cryogenic capture (CryoCell®) from natural gas and basic economical comparison with the amine absorption process.	Hart and Gnanendran (2009)
	Experimental and modelling study of post-combustion CO ₂ capture based on cryogenic CO ₂ freeze-out in dynamically operated packed beds.	Tuinier et al. (2010)
	Performance evaluation of a hybrid system composed by a membrane and cryogenic process to separate oxygen from air for oxyfuel combustion.	Burdyny and Struchtrup (2010)

H₂S, N₂, methane, O₂ and hydrocarbons. The water should be reduced to a lower percentage, as it reacts with CO₂ and other acidic compounds to form acids, which are corrosive (Koorneef et al., 2010b). The CO₂ transport in supercritical form (at pressures ranging 80–150 bar, CO₂ behaves as a compressible liquid with a density of about 900 kgm⁻³) is more efficient, because of the lower density of gaseous CO₂ and relatively high pressure drops per unit of length (McCoy and Rubin, 2008; Haugen et al., 2009). Applying pressures higher than CO₂ critical pressure, temperature fluctuations along the pipeline will not result in the formation of gaseous CO₂ and the difficulties encountered with two phases flow. The energy requirement for the conditioning processes will depend on the composition and pressure of the CO₂-rich stream and the selected transport process and is typically between 90 and 120 kWh/ton-CO₂ (Aspelund and Jordal, 2007). In ship transport most of the volatiles must be removed in order to avoid too cold temperatures and dry ice formation in the liquid CO₂. For pipeline transport, removal is not necessarily required; however, it makes sense from an economic point of view.

5. CO₂ storage

CO₂ sequestration refers the long-term CO₂ storage to reduce the emissions of CO₂ to atmosphere. Their principles are: (i) storage must be safe; (ii) the environmental impact should be minimal; (iii) storage must be verifiable; and (iv) storage liability is indefinite (Lackner and Brennan, 2009). Based on IPCC (2005), the storage options are grouped in: geological storage, ocean storage or mineralization. The last process consists on the conversion of CO₂ to solid inorganic carbonates using chemical reactions (similar to natural weathering). This process offers an opportunity of permanent and safe storage of CO₂ during a long period (Allen and Brent, 2010). The main disadvantage is the high cost. The ocean storage consists on the CO₂ injection at great depths where it dissolves or forms hydrates or heavier-than water plumes that sinks at the bottom of the ocean. This process accelerates the transfer of CO₂ to the ocean that occurs naturally with an estimated rate of 2 Gton/year (Khoo and Tan, 2006). The ocean is considered to be the largest store of CO₂. It is estimated that the ocean contains 40,000 Gton of carbon, contrasting with 750 Gton in the atmosphere and 2200 Gton in the terrestrial biosphere. Several techniques were tested to perform the CO₂ transfer to the ocean: (i) vertical injection; (ii) inclined pipe; (iii) pipe towed by ship; and (iv) dry ice (Khoo and Tan, 2006). However, the increase of CO₂ concentration in the ocean can have serious consequences in marine life. CO₂ leads to the ocean acidification, affecting the growth rate of corals. Therefore, the CO₂ geological sequestration is considered the most viable option (Celia and Nordbotten, 2009; van der Zwaan and Smekens,

2009; Yang et al., 2010a). The geological storage options are: oil and gas reservoirs (depleted, in combination with EOR or in combination with enhanced gas recovery); saline aquifers; and unminable coal seams (in combination with enhanced coal bed methane recovery). The oil and gas reservoirs have already been used, due to the recovery of fossil fuels (Solomon et al., 2008). The requirements for geological storage are: (i) adequate porosity and thickness (storage capacity) and permeability (injectivity); (ii) a satisfactory sealing caprock; and (iii) a stable geological environment to avoid compromising the integrity of the storage site (Solomon et al., 2008). The saline aquifers attracted the attention of many researchers, due to their high storage capacity, higher than those from other geological storage options (IPCC, 2005; Eccles et al., 2009). Michael et al. (2010) and Yang et al. (2010a) presented complete reviews of the CO₂ storage in saline aquifers. However, the technical risks and economical feasibility are rarely mentioned. These aspects were studied by Xie and Economides (2009), Mathias et al. (2009) and Singleton et al. (2009). The main concern of CO₂ storage is the leakage of this gas to the atmosphere, which could render CCS ineffective as climate change reduction option (Celia and Nordbotten, 2009). The CO₂ leakage has several consequences (Bachu, 2008; Allen and Brent, 2010): (i) asphyxiation (CO₂ is dangerous for human health for concentrations greater than 0.5–1.5% v/v, where the atmospheric concentration is 0.038% v/v); (ii) death of low-lying and small animals living in low-level enclosed areas where the CO₂ can accumulate (as it is heavier than air); and (iii) change of the water pH and affecting the marine life (if the leakage occurs in the bottom of the ocean). Moreover, the injection of CO₂ in geological formations can acidify the potable groundwater presented nearby, which causes the dissolution of heavy (toxic) metals (van der Zwaan and Smekens, 2009). This effect reduces the quality of the drinking water obtained from these sources. Table 5 presents some relevant issues about studies concerning the CO₂ storage.

6. CCS projects

Steenveeldt et al. (2006) presented a list of the international research projects on CCS. The description of some of them (EU projects) is presented by Krooss and May (2006). A compilation of the recent published works concerning CCS projects is presented in Table 6.

7. Future trends

The costs associated with all parts of CCS should be reduced through intensive research and demonstration. For application of CCS at large-scale, substantial research is required in several areas, mainly in capture technologies (60–80% of the

Table 5 – CO₂ storage publications.

Process	Relevant issues	Reference
Geological storage	Assessment of the economic efficiency of CO ₂ sequestration and analysis of the optimal timing and extent of CO ₂ sequestration.	Keller et al. (2008)
	Presentation of a review of CO ₂ sequestration in saline aquifers.	Yang et al. (2010a)
	Analytical and numerical analysis of the effect of coupling of geochemical reactions with convective mixing of dissolved carbon dioxide during geological storage.	Ennis-King and Paterson (2007)
	Analysis of the economic and climatic implications of the large-scale use of CCS to achieve climate change control target, considering CO ₂ leakage.	van der Zwaan and Gerlagh (2009)
	Presentation of the current knowledge of health, safety and environmental risks of geological CO ₂ storage.	Damen et al. (2006a)
Oceanic storage	Estimation of air-sea fluxes of anthropogenic CO ₂ and investigation of the oceanic transport of this gas.	Fletcher et al. (2006)
	Evaluation of the environmental impacts of direct CO ₂ injection in the ocean.	Israelsson et al. (2010)
	Discussion of CO ₂ storage in geological formations and in oceans	Strak and Wardencki (2007)
Mineralisation	Development of a numerical model to evaluate the CO ₂ diffusion, when it is injected in the ocean.	Jeong et al. (2008)
	Investigation of the precipitation of magnesium carbonates from magnesium-rich solutions, prepared from serpentinite.	Teir et al. (2007)
	Evaluation of the energy economy of a two or three-stage gas–solid process for magnesium silicate carbonation.	Zevenhoven et al. (2008)
	Assessment of CO ₂ sequestration costs by aqueous Ca silicate carbonation	Huijgen et al. (2007)
	Evaluation of the stability of the mineral carbonation product against acid rain.	Allen and Brent (2010)

total cost of CCS chain): enhanced performance of solvents, improved membrane systems, efficient chemical looping processes. Concerning the CO₂ capture, all technologies (post-combustion, pre-combustion and oxy-fuel combustion) have advantages and disadvantages. The initial application of a CCS technology to large CO₂ emission sources will always require additional incentives to reduce the investment effort. This can be performed by the payment of the avoided emissions to atmosphere. The needed research directions in CO₂ capture are: (i) improvement of chemical and physical sorbents; (ii) improvement ion-transport and other membranes and integration in the power processes; and (iii) eliminating

the gaps of the scale-up from laboratory to pilot scales (CSLF, 2010).

With respect to CO₂ transport and storage, it is expected less potential reduction costs. Additionally, the effects of impurities in CO₂ transport should be studied. Response procedures should be developed in advance for the possibility of CO₂ pipeline accidents. However, research is required about safety aspects and capacity understanding (CSLF, 2010). The safety could be the greatest barrier for the CCS implementation. The public should be convinced that this technology can store CO₂ for long periods with low risk probability for human health and environment.

Table 6 – Studies concerning CCS projects.

Project	Focus areas	References
CANMET Energy Technology Centre (CETC) R&D Oxy-fuel Combustion for CO ₂ Capture	CO ₂ capture	Lu et al. (2008), Manovic and Anthony (2008), Manovic et al. (2008a), Manovic et al. (2008b), Manovic et al. (2009a), Manovic et al. (2009b), Zanganeh et al. (2009)
CO ₂ Capture Project – Phase II (CCP2)	CO ₂ capture	Abad et al. (2007), Gayan et al. (2008), Gayan et al. (2009), Jansen et al. (2009), Wright et al. (2009), van Selow et al. (2009), de Diego et al. (2009), Dueso et al. (2009), Abad et al. (2010)
CO ₂ SINK	CO ₂ storage: geological storage	Yordkayhun et al. (2007), Yordkayhun et al. (2009), Kopp et al. (2009c), Kopp et al. (2009b), Kopp et al. (2009a), Giese et al. (2009), Freifeld et al. (2009), Schilling et al. (2009), Prevedel et al. (2009), Förster et al. (2009), Kopp et al. (2010), Morozova et al. (2010), Zettlitzer et al. (2010)
EU CASTOR	CO ₂ capture: absorption	Abu-Zahra et al. (2007), Notz et al. (2007), Tobiesen et al. (2008), Dugas et al. (2009), Kim et al. (2009), Knudsen et al. (2009), Mangalapally et al. (2009), Kim and Svendsen (2010), Notz et al. (2010)
EU CO2GEONET	CO ₂ storage: geological storage	Pauwels et al. (2007), Annunziatellis et al. (2008), Bateson et al. (2008), Beaubien et al. (2008), Annunziatellis et al. (2009), Arts et al. (2009), Holt et al. (2009), Krüger et al. (2009), Maul et al. (2009), West et al. (2009) and Oppermann et al. (2010)
EU DYNAMIS	CO ₂ capture: pre-combustion capture	Abu-Zahra et al. (2009), Bouvart and Prieur (2009), Hetland (2009), Hetland et al. (2009b), Cormos et al. (2010)
US DOE Carbon Sequestration Program	CO ₂ capture and storage	Litynski et al. (2006a), Litynski et al. (2006b), Kutchko et al. (2007), Litynski et al. (2008), Figueroa et al. (2008), Kutchko et al. (2008), Kutchko et al. (2009), Litynski et al. (2009), Israelsson et al. (2010)
COACH	CO ₂ capture and storage	Hetland and Christensen (2008), Hetland (2009), Hetland et al. (2009a), Vincent et al. (2009), Fang et al. (2009b), Yan et al. (2008), Yan et al. (2009)
EU GeoCapacity	CO ₂ storage	Vangkilde-Pedersen et al. (2009), Hatziyannis et al. (2009), Martínez et al. (2009), Chen et al. (2009, 2010)
CATO	CO ₂ capture and storage	Damen et al. (2006b), Damen et al. (2007, 2009), van den Broek et al. (2008), Koornneef et al. (2008, 2010a, 2010b), van Alphen et al. (2010a, 2010b)

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