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CARBON DIOXIDE SEPARATION TECHNOLOGIES

TECHNOLOGIE SEPARACJI DWUTLENKU WĘGLA

CO_2 emission from combustion fossil fuels is considered as the primary factor in the global warming. Different methods for separation CO_2 from combustion flue gases are extensively used across the world. The aim of this study is to analyze the most important technological solutions of CO_2 separation. For this reason chemical absorption, physical absorption, adsorption approach, membrane filtration and cryogenic process were researched. Concluding, selection of the right method for carbon dioxide capture separation is a complex issue and a range of technological and economic factors should be taken into consideration prior to application on the industrial scale.

Keywords: CO_2 capture, membrane technology, absorption, cryogenic separation

Emisja CO_2 do atmosfery pochodząca ze spalania paliw kopalnych jest uważana za główny czynnik globalnego ocieplenia. Różne metody oddzielania CO_2 od gazów spalinowych są szeroko stosowane na całym świecie. Celem niniejszego artykułu jest analiza najważniejszych technologicznych rozwiązań separacji CO_2. W tym celu przeanalizowano następujące metody: absorpcja chemiczna, absorpcja fizyczna, adsorpcja, filtracja membranowa i proces kriogeniczny. Podsumowując, wybór właściwej metody separacji dwutlenku węgla pod kątem wychwytu dwutlenku węgla jest złożonym zagadnieniem, a przed zastosowaniem na skalę przemysłową należy wziąć pod uwagę szereg czynników, w tym technologiczne i ekonomiczne.

Słowa kluczowe: wychwyt CO_2, technologia membran, absorpcja, separacja kriogeniczna

1. Introduction

CO_2 sequestration and CO_2-EOR (Enhanced Oil Recovery) enables neutralization of enormous amounts of that gas from large industrial sources of emission (Czarnota et al., 2017). The millions tons of CO_2 are produced in: power plants, thermal-electric power stations, steel plants, cement plants, chemical plants (refineries, ammonia, ethylene oxide, hydrogen production plants),

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488

cars and others. An important issue here is a diversified concentration of CO_2 in flue and industrial gases. In most cases it is low and amounts are between a few to several % of the gas volume. A clean stream is only emitted in some industrial processes. It is one of the factors holding back geological sequestration of carbon dioxide today. Therefore, firstly the underground storage of CO_2 near emission centers of that gas should be taken into consideration (Darde et al., 2010). High costs of implementing the carbon capture and storage (CCS) installation are connected with first stage, that is the capture of CO_2 from the stream of gases, especially when the concentration of CO_2 is not very high. The costs of the capture amount to 60-70% of the entire cost of sequestration. CO_2 storage is considered as a technically proven technology, however, deployment can be delayed by uncertainties and challenges associated with estimation of storage capacity, tracking and monitoring of CO_2 flow after injection, injection-induced seismicity and standardization of storage evaluation criteria (Aminu et al., 2017). Exhaust gases in energetics, depending on the type of the fuel, consist from 3 to 15% of CO_2. Modern technologies of energy production enable to limit the amount of CO_2 entered into the atmosphere with simultaneous increase of its share in the flue gas (Zhao et al., 2013). In the power production sector, three main technologies of CO_2 capture can impair this increase: post-combustion CO_2 capture, pre-combustion CO_2 capture and oxy-fuel combustion. In the post-combustion process CO_2 is captured from the power plant's exhaust gas after process of combustion. For the pre-combustion separation method, the fossil fuel is pre-treated before combustion. As a result of gasification (for coal) or reforming (for natural gas) a syngas is produced. The mixture of CO and H_2 then undergo water gas shift reaction with steam where the CO gas is converted to CO_2 and later separated in the capture subsystem. The CO_2 level in combustion flue gas is lower (i.e. 7-14% for coal-fired and as low as 4% for gas-fired) comparing with the shifted syngas (>20%) from the pre-combustion process (Leung et al., 2014). As a result, the energy penalty and associated costs for the capture unit are higher for the post-combustion process. For both processes (pre- combustion and post-combustion) similar CO_2 separation technologies can be applied to isolate the CO_2 from the flue/fuel gas stream (Jansen et al., 2015). The detailed division of separation techniques is presented in Fig. 1.
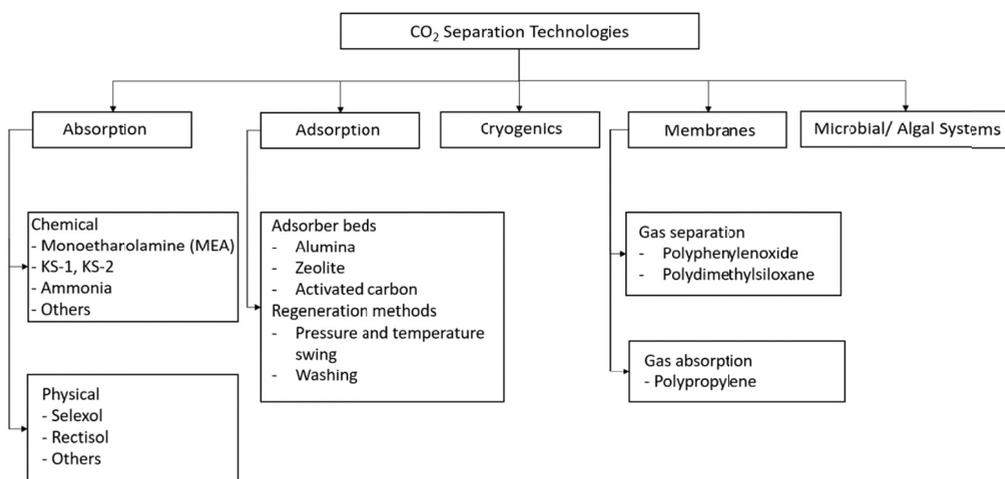

Fig. 1. Classification of separation techniques available for the removal of CO_2
in post-combustion stage (Wang et al., 2011)

In industrial practice, the degree of the use of particular techniques of separation differs, owing to required instrument solutions, effectiveness and costs. Currently, post-combustion CO_2 capture by amines-based chemical absorption is the most popular technology to reduce CO_2 emissions. The most commonly used technologies of CO_2 separation can be divided according to the physical and chemical nature of the process into the following categories: chemical absorption, physical absorption, adsorption, membrane filtration and cryogenic processes. The aim of this study is to discuss the most important technological solutions of CO_2 separation.

2. Chemical absorption

Chemical absorption consists in the moving of carbon dioxide from gas phase to liquid phase, and then in the liquid phase a chemical reaction occurs, resulting in the absorption balance being moved towards the product, that is, carbon dioxide in a bound form. As absorbents, most commonly used are monoetanoloamine (MEA), dietanoloamine (DEA), methylodietyloamine (MDEA) and trietanoloamine (TEA) (Shakerian et al., 2015). However, more amines were tested in process of absorption with a chemical reaction of CO_2, e.g. diisopropylamine (DIPA), 2-amino-2-methyl-1-propanol (AMP), methyl-1,3-propanediamine (MAPA) (Cuccia et al., 2018). The particular amines differ significantly when it comes to their physical and chemical characteristics and also sorbent characteristics. A choice of a right sorbent is difficult, owing to complex amine characteristics. For example, the enthalpies of reaction and evaporation decrease from primary to tertiary amines hence the energy required for solvent regeneration is lower for tertiary amines. The reaction rate of primary amines is higher than of tertiary amines which results in a decreased circulation of primary amines in the system. Low purchase costs and the availability of monoethanolamine (MEA) makes it the most popular sorbent, despite some obvious limitations. Mostly dilute solutions of amines are used in the process, with the concentration no higher than 20 wt%. The absorption process is carried out in a temperature between 20 and 50°C, the process of desorption (regeneration) in the temperature between 110 and 130°C (Kim et al., 2013). Fig. 2 presents a typical installation scheme with the use of amine absorption.
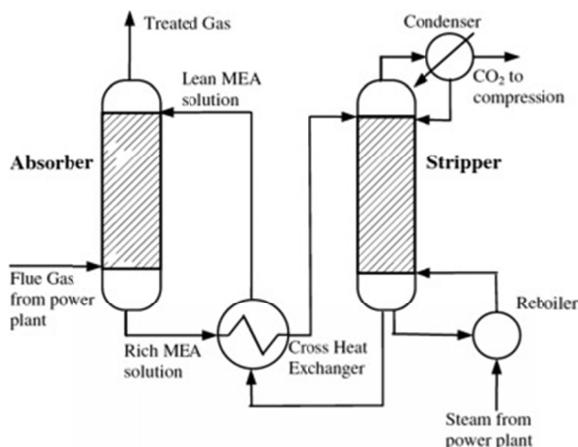

Fig. 2. Flow diagram of chemical absorption process for post-combustion carbon capture (Wang et al., 2015)

3. Physical absorption

Rectisol is a low-temperature physical CO_2 absorption process with the use of methanol. Originally, the process was developed for synthesis gas wash from sour components in plants such as coal to liquids as well as coal to substitute natural gas and integrated gasification combined cycle. The first Rectisol installation was built during the 1950s by Lurgi at the Sasol-Secunda CTL plant in South Africa, currently there are 150 units operating worldwide (Gatti et al., 2015). Methanol has a number of advantages in comparison with other solvents. Rectisol wash allows to remove both H_2S, COS and CO_2 as well as other impurities like HCN and NH_3. Methanol has high selectivity towards sour components. Other advantages of methanol are low viscosity, low-corrosivity, high thermal stability, high chemical stability and non-foaming tendency. The Rectisol process is flexible and can be configured any specific upstream syngas condition as well as downstream product specification. It allows to reach a wide range of H_2S and CO_2 separation levels. The limitation of methanol is its toxicity and high vapor pressure which enforces the process to be carried out at low temperatures. On the other hand, the solubility of gases increases significantly as the temperature decreases. Low operating temperatures (usually between –40°C and –62°C) in the methanol process result in the lowest required circulation rate, and lowest net power requirement. Most of the solvent can be regenerated by a simple pressure letdown or by inert gas stripping. Depending on the needs, Rectisol can be carried out according to the scheme proposed by Lurgi or Linde. In Fig. 3. the typical Rectisol unit is presented. The systems differ when it comes to the ways of regeneration of the solvent and the quantity of absorbers. First stage in Lurgi's scheme consists of a pre-washing column for water, hydrocarbons and heavier contaminants removal. The syngas flows to the absorption column in which two chilled methanol streams wash the CO_2, H_2S and COS out. The bottom of the column is externally refrigerated with an ammonia refrigeration cycle. The CO_2-rich liquid methanol is then regenerated by flashing to atmospheric or sub-atmospheric pressure. The small fraction of methanol is regenerated by distilling and then introduced in the top of absorber. Sol-
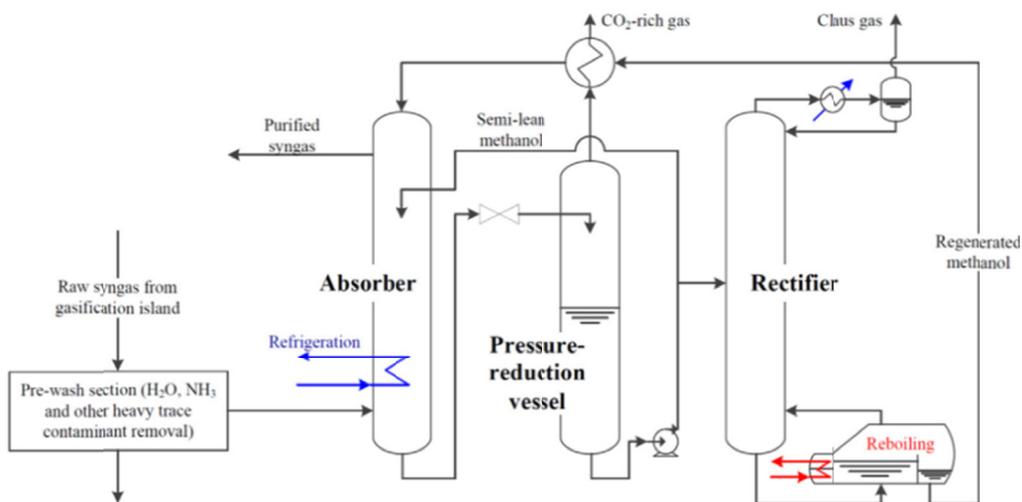

Fig. 3. Lurgi patented Rectisol® (Gatti et al., 2015)

vent regeneration mainly by pressure reduction, is simple and inexpensive. The disadvantage of this solution is high content of H_2S in CO_2-rich gas (up to 1.5% of H_2S) hence the process is unsuitable for direct CCS (Gatti et al., 2015).

Linde's scheme (Fig. 4) allows to produce selectively a CO_2-rich stream (mostly for either EOR or urea production purposes). In this configuration, the absorber is intercooled to remove part of the heat of absorption of CO_2 and features an intermediate extraction of almost half of the solvent flow rate at the exit of the CO_2 removal section of the column (Gatti et al., 2015). In the H_2S section at the base of the column, the already CO_2-saturated methanol removes the H_2S from the flowing raw syngas. The liquids from the bottom of the absorber go through a pre-flash to an H_2S enrichment column where most of the CO_2 is released. The remainder CO_2 is desorbed in a N_2 stripper. Pure CO_2 (98.6% purity) consists of 86.6% of the inlet of this flowsheet.
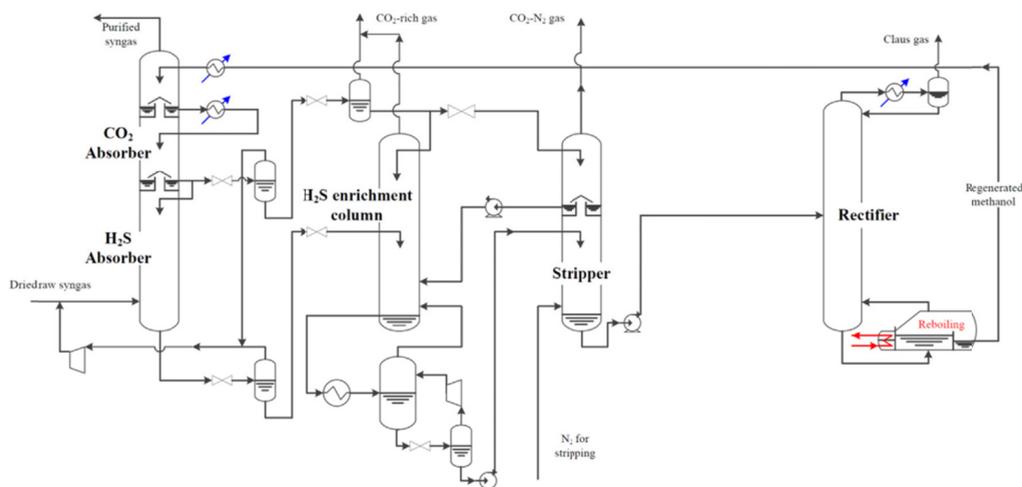

Fig. 4. Linde patented Rectisol (Gatti et al., 2015)

Recently both Linde and Lurgi have proposed flowsheets for the selective removal of H_2S and CO_2 where CO_2 is released at different pressures in two sequential desorption columns MP and LP (medium and low pressure) (replacing the original H_2S enrichment section). LP column is more complex then N_2 stripper combining CO_2 desorption by flashing and heating with H_2S reabsorption via methanol recirculation. In Fig. 5. Linde Rectisol scheme for CCS is presented.

Currently, the Great Plains Synfuels Plant, North Dakota, US is the largest carbon dioxide capture project from a coal gasification plant. Its Rectisol unit working since 2000 captured over 20 million metric tons of CO_2 for EOR (Weyburn, Canada) to date (Huxley, 2006).

4. Adsorption

Adsorption is a method used in the industry mainly to purify and recover gases, that is, the separation of air, recovery and purification of hydrogen, separation of CO_2. The method consist in condensation of vapor and gases on the surface of a solid adsorbent as a result of molecular
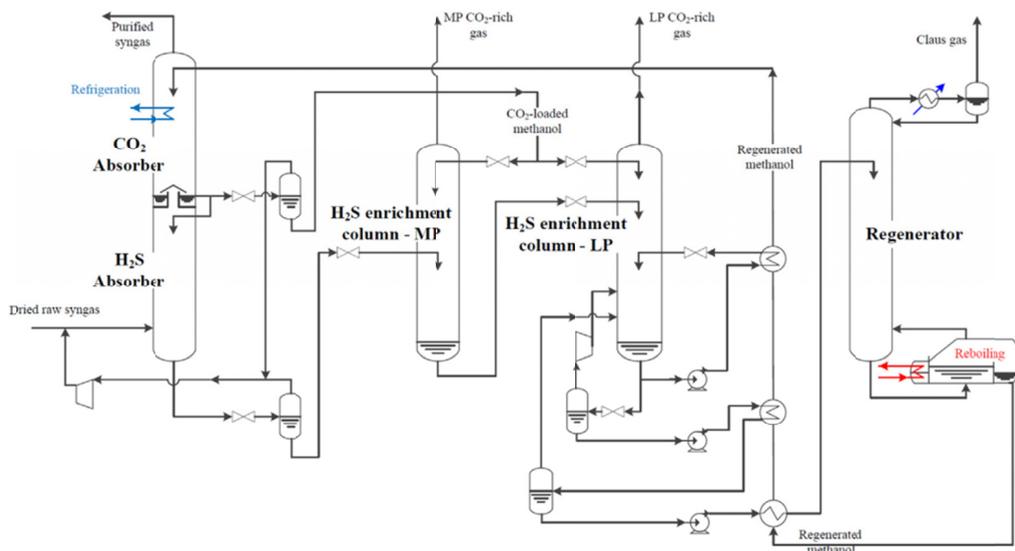
Fig. 5. Linde Rectisol scheme for CCS (Gatti et al., 2015)

forces and uses the differences in sorption capacity of particular components of the separated gas mixture on a given adsorbent (Ben-Mansour et al., 2016). Among the adsorbents, the most popular are zeolite and carbon adsorbents, which constitute an adsorption bed when filling the adsorption tower. For the process to be carried out continuously, at least two adsorption towers are required. In practice multi-bed systems are used which benefits the effectiveness of the process. CO_2 is adsorbed through physical or chemical sorption. In the process of physical adsorption a van der Waals interaction occurs between the adsorbent and adsorbate (as a result of the dispersive forces or dipole interactions). Van der Waals forces are weak, but long range interactions. The energy released during physical adsorption is of the same order as enthalpy condensation. The process generally takes place rapidly and reversibly. In the process of chemisorption the molecules of CO_2 fuse with the surface to form chemical bonds (usually covalent). Development of chemical adsorbents aims to improve selectivity towards CO_2 and ability to operate at high temperatures. In the case of physical adsorbents, there is a tendency to increase the surface area and selectivity. The type of adsorbent plays a crucial role. The key parameter is the high selectivity for the adsorption of CO_2 relative to N_2 adsorption and a high capacity of the adsorbent and the associated high surface area. Because of high flow rates, a considerable mechanical strength of the adsorbent and the form not causing excessive pressure drops is required. The resistance of the adsorbent for so-called poisoning, that is, irreversible adsorption leading to deactivation of the adsorbent and a necessity to replace it, is also important. By far, numerous adsorbents were developed which meet the process conditions, among them zeolites, activated carbons, silica, aluminum oxide. Recently, there are also proposals for the use of adsorbents with the ability of chemisorption of CO_2, among others, aluminosilicates impregnated with amines, a so-called network of metal oxides.

5. Membrane technology

A membrane is a selective barrier, separating ingredients from the flue gas stream through the membrane to the permeate at various velocities. Depending on the membrane structure, the following separation mechanisms can be distinguished (Narębska, 1997):

- sieve effect,
- separation arising from differences of components solubility and diffusion speed in the membrane, this separation mechanism dominates in compact membranes (dense) or containing micropores only, wherein in the case the latter, the various compounds transport speed is a result of Knudsen's diffusion and surface diffusion in micropores (non-organic membranes),
- separation concerning various ions migration speed through ion-exchange membranes,
- separation with selective and reversible exchange reaction of the specific compounds with other active groups of polymer membrane skeleton. The membrane separation processes occur in the so-called membrane modules assuring a flow of separated mixture parallel or perpendicularly towards the membrane surface. In order to limit membrane surface blockage by active deposit, the feeding solution movement should be turbulent, preferably directed along the membrane surface. Flow directions of the feeding solution and permeate are perpendicular (cross-flow filtration), the so-called retentate flows out of the module (apart from the permeate). Such membrane modules are applied mainly in the ultrafiltration, nanofiltration and reverse osmosis. The so-called dead end flow filtration is usually used in microfiltration processes. In Fig. 6 dead-end flow filtration and cross flow filtration schemes are presented.
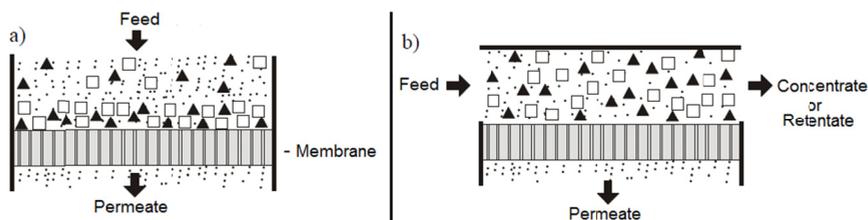

Fig. 6. a) Dead-end flow filtration scheme b) cross flow filtration scheme (Narębska, 1997)

The value of the permeability coefficient depends in porous membranes on the pore size distribution and tortuosity, and in compact membranes – on individual capability of diffusing components in the membrane. In the case of reactive membranes, an additional factor is chemical reaction kinetics. Selectivity of the membrane towards two ingredients i and j is a measure indicating practical separation capabilities of these ingredients. These values are usually specified in accordance with the following formula, defining the so-called separation coefficient, β_{ij} (Narębska, 1997):

$$\beta_{ij} = \frac{c_i^{II} / c_j^{II}}{c_i^{I} / c_j^{I}} \tag{1}$$

where: c_i^{I} and c_j^{I} are concentrations or partial pressures of i and j components in the feeding solution, while c_i^{II} and c_j^{II} apply to the permeate.

A variety of membranes have been considered for flue gas CO_2 capture. The non-organic membranes currently available in the market for the CO_2 separation include different types and are given in Table 1.
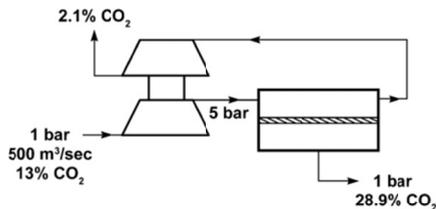
TABLE 1

Comparison of different materials for large-scale CO_2 capture (Abanades et al., 2015)

Membrane type	Structure	Application	Advantages/cost
Silica	Amine functionalized silicas coated on a α-alumina or γ-alumina support; also anisotropic metal oxide silicas	CO_2/H_2 and CO_2/N_2	High selectivity / medium to high cost
Zeolites	Thin zeolite layer deposited on a support such as porous α-alumina or stainless steel with well defined pore structure	CO_2/CH_4	Very high selectivity and high permeability along with good thermal, mechanical, chemical stability /high cost
Carbon molecular sieve	Porous random networks typically produced by pyrolysis of thermosetting polymers under controlled conditions	CO_2/CH_4	Good thermal and chemical stability/ high to very high cost
Metal	Palladium and alloys in dense or composite formats (Pd on tantalum or vanadium support film)	H_2 separation	High selectivity, high temperature tolerance / very high cost
Ceramic	Anisotropic perovskites and fluorites	O_2/N_2	High selectivity, high temperature tolerance, lower energy consumption than cryogenic separation / high to very high cost

The major problem preventing adoption of membrane systems for CO_2 treatment are the scale of the process and the large and energy-consuming compression equipment needed. A key issue is the pressure difference across the membrane. For one-stage separations, compression of the flue gas can be provided either by compressing the feed gas or drawing a vacuum on the permeate. Compression is preferred due to lower capital cost of compression equipment and smaller membrane area needed. (Merkel et al., 2010) compared two one-stage membrane processes treating a simplified model flue gas from a 600 MWe power plant (13% CO_2, 87% N_2, no O_2, no H_2O). A turboexpander allows to recover a portion of the energy when feed compression is used but still more than 20% of the energy production of the power plant is here consumed. The vacuum process requires less energy which is needed to pump the much smaller volume of gas that permeates the membrane. On the other hand, large-scale industrial vacuum equipment is not commercially available. Both designs produce the same separation but for feed compression less membrane area is needed. In Fig. 7 a comparison of feed compression versus permeate vacuum for single-step membrane process is given.

Both cross-flow and counter-flow modules are likely to be used for CCS purposes. (Merkel et al., 2010) proved that counter-flow operation is clearly superior: the permeate CO_2 concentration increases from 29% to 41%, the membrane area required is reduced by 38%, and the power consumption decreases from 56 MW to 46 MW – an 18% energy saving. However problems with greater complexity, flow distribution and permeate-side pressure drops associated with counter-flow modules limit their application. Apart from these technical issues a single-stage
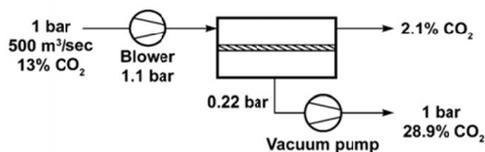
Fig. 7. A comparison of feed compression versus permeate vacuum for single-step membrane process
(Merkel et al., 2010)

membrane process cannot capture more than 90% of the CO_2 in flue gas. To achieve the desired CO_2 recovery and purity a multi-stage membrane design is required.

Universal Oil Product's (UOP's) membrane systems have been in commercial use for more than 20 years in the natural gas and petroleum refining industries. At present in the world more than 80 membrane units have been installed by UOP (Dortmundt, 1999). The two largest CO_2 removal membrane systems in the world are the Separex TM units installed in Qadirpur and Kadanwari, in Pakistan. For example, the Kadanwari system is a two-stage unit designed to treat $5.946 \cdot 10^6 \, m^3/d$ of feed gas at 90 bar and the CO_2 content is reduced from 12 to less than 3%. In Fig. 8. a comparison of cross-flow module versus counter-flow module is illustrated (Merkel et al., 2010).

To sum up, the membrane separation processes can be performed in a continuous way at ambient temperatures, as a result of which it is possible to use them to separate substances which cannot support conditions of other processes, e.g. distillation or crystallization. Furthermore, it is not necessary to use additional substances which pose a threat to the environment. The next strong points are inconsiderable demand for energy and relatively low installation and material costs. Finally, wide range of available membranes and simple methods of modification allow an easy selection of membranes to satisfy particular needs. However, when using membranes, some obstacles are likely to occur. To be more specific, on membrane surfaces concentration polarization processes may be observed and in micro, ultra, nanofiltration processes membrane surfaces are blocked by concentrated and frequently gelled mass of contained ingredients, the so-called active deposit. This decreases the selectivity and filtration efficiency, and makes it necessary to replace or regenerate membranes.

6. Cryogenic CO_2 capture

Cryogenic CO_2 Capture (CCC) uses phase change to separate CO_2 and other pollutants from exhaust or process gases. The simplified process scheme for CCC is presented in Fig. 9. The CO_2-rich stream is cooled to such a low temperature (about -140°C) that it desublimates.
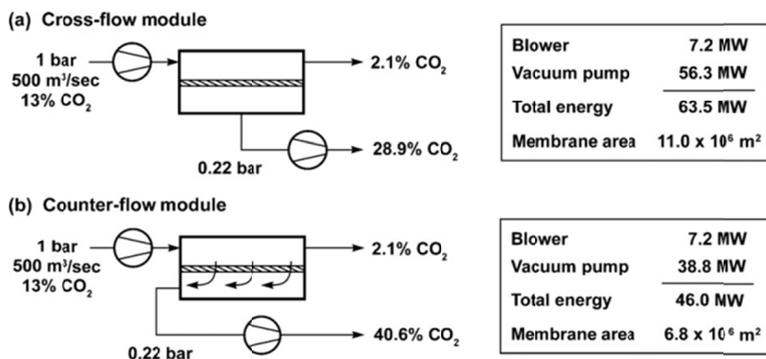
496

Fig. 8. A comparison of cross-flow module versus counter-flow module (Merkel et al., 2010)

The solid CO_2 is separated from the remaining gas, pressurized, melted, and delivered at pipeline pressure. The gas that remains after the CO_2 and other pollutants have been removed is nearly pure nitrogen, and can be safely released to the atmosphere. At present CCC is considered as an unrealistic candidate for post-combustion CO_2 capture due to expected high cooling costs (Tuinier et al., 2011). The novel cryogenic capture technology based on the dynamic operation of packed beds which may be economically feasible. A process cycle consists of three consecutive steps: a cooling, capture and recovery step. A fixed bed is cooled down to temperatures below −120°C using cold energy of Liquefied Natural Gas (LNG). A hot flue gas is cooled down by the refrigerated packed bed resulting in H_2O condensation at the packing surface. The further cooling down leads to CO_2 desublimation extracting the cold from the packing until again an
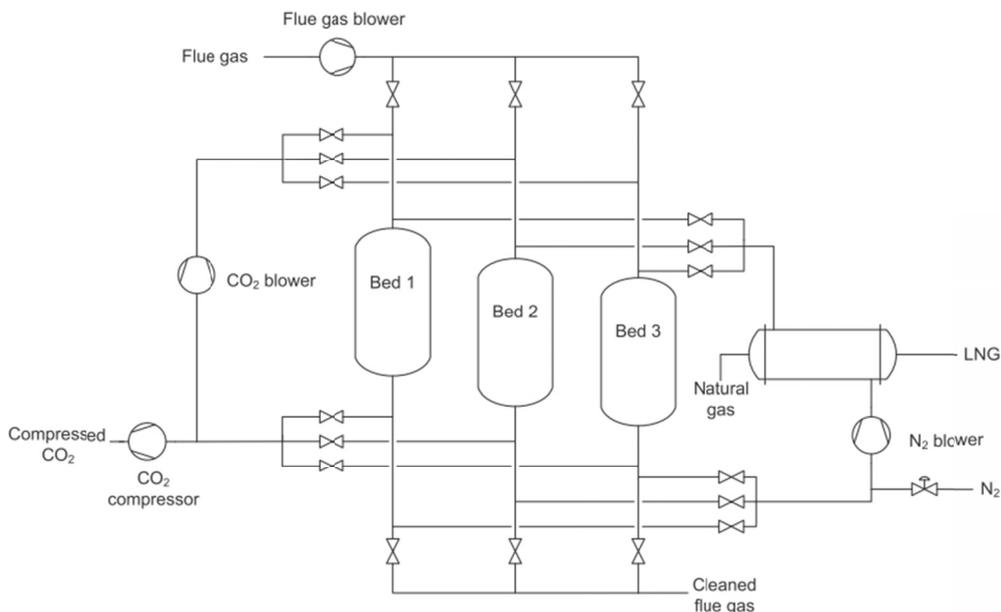

Fig. 9. Simplified process scheme for cryogenic CO_2 capture (Tuinier et al., 2011)

equilibrium is reached and a front of sublimating CO_2 will move through the bed. The bed has to be switched to the recovery step, when CO_2 breakthrough is observed at the outlet (Tuinier et al., 2011; Wang et al., 2011). During recovery stage the hot zone at the inlet will be moved through the bed, causing the zones with condensed H_2O and desublimated CO_2 to continue moving through the bed and obtaining pure CO_2 at the outlet. To achieve continuous operation of the process, it is required to operate all three steps in parallel in three beds. The cryogenic technology shows that deep CO_2 removal can be obtained, for example when cooling to –150°C, the CO_2 removal efficiency is more than 99.9%. From a practical point of view the cryogenic concept could be the preferred option only when is integrated into LNG regasification or/and cryogenic air separation. Cryogenic nitrogen removal processes for natural gas also offers an attractive opportunity for liquefied CO_2 production as a side stream (Knapik et al., 2018).

7. Summary

Choosing the right method of carbon capture is a complex issue, a range of technological and economic factors should be taken into consideration, including feed gas compositions and conditions, the desired purity of the treated gas, plant costs (capital and operating), commercial experience, and process reliability and operational flexibility. On the other hand, a large number of working CO_2 capture installations and relatively simple mechanisms of operation of individual units allows to formulate broad guidelines for the choice of the appropriate method of capturing. The technical problems related to CO_2 separation are well-recognized and in the case of most processes properly solved. A considerable progress with regard to apparatus and material solutions allows separation of CO_2 in any scale, and it is possible to strictly control the product parameters. A high level of technological readiness is proved by numerous industrial systems operating for the purposes of the food industry or chemical industry. Similarly, numerous pilot systems operating for the purpose of the heat and power stations herald a high level of CO_2 recovery from flue gases and high purity of the final product. Certainly, there are still difficulties related to intensifying the heat exchange and working medium losses, but such difficulties can be easily eliminated thanks to experiences related to other chemical systems. In the European industry, there is no demand for large amounts of CO_2. Actually, the entire investment can be considered profitable if produced CO_2 is used for EOR methods or CO_2 capture is associated with natural gas purification. Further development of CO_2 separation methods is dependent upon political and economic conditions. This review shows that technologies available on the market are sufficiently effective, however, the biggest obstacle pertaining to implementing CO_2 capture on the wide scale are economic issues with capital and operating expenditures. Merkel et al. (2010) performed a fundamental study to the economics for membrane technology capture, handling flue gas of 602 kg/s including 12.9 vol.% of CO_2. In their work an air sweep was applied and fed to the boiler. This option outperformed variant of generating driving force for permeation by a vacuum on the permeate side. Tuinier et al. (2011) showed that for the amine absorption with steam generation, the avoidance costs are equal to 133.4 \$/ton and for the cryogenic concept including refrigeration agent generation, price is 126.5 \$/ton. Membrane technology has comparable cost of 120.9 \$/ton, however, expenses can be substantially reduced when steam or LNG is available for free or low cost, then amine or cryogenic approaches could be considered as preferable, since CO_2 avoided costs can be reduced to 54.5 \$/ton and 52.8 \$/ton, respectively. These technological solutions could become a major candidates for capturing CO_2 from flue gas.

498

Acknowledgement

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