



Assessment of a membrane contactor process for pre-combustion CO₂ capture by modelling and integrated process simulation

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ABSTRACT

A membrane contactor process for pre-combustion CO₂ capture from shifted synthesis gas originated from IGCC power plant is assessed from the technical and economical point of views. The process is designed as pressure swing absorption and desorption in a closed loop. The design basis for process simulation were synthesis gas containing CO₂ and H₂ only, and the CO₂ capture efficiency was fixed to 90%. The CO₂ gas was absorbed in ionic liquid [bmim][TCM] inside a hydrophobic, porous hollow fibre membrane contactor. One-dimensional mathematical model of membrane contactor developed in MATLAB was integrated to the process simulation software (HYSYS) through Cape-Open simulation compiler. The energy evaluation of this process revealed that compressors are the most energy demanding process equipment. The specific energy requirement for this process is estimated 0.75 MJ/kg CO₂. A parametric study was also performed to analyse the effect of CO₂ concentration in feed gas and liquid to gas ratio. The capital cost investment and total operating costs of CO₂ capture unit were also evaluated. The capital investment required for capturing 0.14 M ton CO₂/year including CO₂ compression is 47.4 M \$, and the operating cost per year is 9.04 M \$. The membrane absorber contributed about 39% to total investment cost. The specific cost of this capture unit is calculated to be 87 \$/ton CO₂.

1. Introduction

Greenhouse gas emissions are imposing great threat to increase the climate temperature. Carbon capture and storage (CCS) is addressing this challenge in order to mitigate the CO₂ emissions. The widely known strategies for reducing CO₂ emissions are pre-combustion or post-combustion CO₂ capture, or through oxy-fuel combustion. Post-combustion has been investigated the most among the above-mentioned three approaches due to retrofitting to existing power plants. However, post-combustion CO₂ capture offers many challenges, equipment corrosion, low CO₂ partial pressures, and pressurization of gas stream to storage site are a few to mention here. Pre-combustion CO₂ capture involves the mitigation of CO₂ before the fossil fuel is burned to produce power (Thambimuthu et al., 2005) and can be applied to both coal/natural gas fired power plants. The benefits of pre-combustion CO₂ capture encompass the less energy exhaustive process, high gas volumes, high gas pressure as driving force, less regeneration energy requirement, and more prominently the generation of another important fuel gas, hydrogen (H₂). The energy generation demand is typically 10–16%, which is approximately half of that in the case of post-combustion CO₂ capture (Susta and LP, 2007). Nevertheless, this technology is still under scrutiny for different physical and chemical

absorbents and a CCS plant based on it is yet on launching pad.

The pre-combustion CO₂ capture from an Integrated Gasification Combined Cycle (IGCC) plant is comparatively an efficient technology to reduce emissions, but the overall cost of CO₂ capturing plant is potentially higher. There are some other challenges such as operational cost of the CO₂ capture plant, lack of sufficient experimental data and operational activities. Moreover, the IGCC power plant with CO₂ capture also tends to reduce the overall efficiency of the power plant. In order to combat this challenge, more energy efficient solvents and processes should be investigated and the absorption equipment be optimized.

There are, in general, two types of absorbents namely chemical absorbents and physical absorbents. Chemical absorbents react with CO₂ gas and enrich the mass transfer. Aqueous amine, carbonate-bicarbonate, hydroxide solutions are preferably employed on pilot and industrial scale, and in membrane contactor processes (Zhao et al., 2016), but high regeneration energy associated with it increases the Capex of the industrial plant. On the other hand, physical solvents absorb CO₂ as pressure of the gas increases. Generally, high pressures are favourable for physical solvents (Figuroa et al., 2008). For CO₂ absorption, physical solvents are favoured in case of high pressure and low temperatures and when large gas volumes are available at high

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pressures (Dave et al., 2016; Dymont, 2015). The commercially available physical absorbents include polypropylene carbonate (Kohl and Nielsen, 2018), Methanol (Rectisol) (Knapp, 1968; Gatti et al., 2014), N-methyl-2-pyrrolidone (Purisol), Dimethylether polyethylene glycol (Selexol) (Damen et al., 2011; Tsunatu et al., 2015), and water. The physical absorbents exhibit stripping of acid gas by pressure swing, which reduces the energy penalty as in case of chemical absorbents, where the regeneration of absorbents demands high energy. The selection of an ideal physical absorbent could be based on CO₂ solubility and viscosity of the absorbent. The vapour pressure of the selected absorbent should also be as low as possible in order to circumvent the entrainment and solvent losses in regeneration step. The commercial physical solvents as mentioned above grab a few disadvantages such as reduced mass transfer (Selexol), validity upto low temperature operation (Rectisol), volatile solvent (Purisol), and economically not adequate to achieve high product gas purity. Ionic liquids are investigated as physical absorbent in recent years for CO₂ capture due to their task specific nature. The ionic liquids could be alternative promising physical absorbents due to less energy demand during stripping process as these exhibit insignificant low vapour pressures. Ionic liquids are also reported to have significant CO₂ solubility. The surface tension of ionic liquid with respect to membrane material should be tested first before its final selection. The only challenge in its implementation arises due to high viscosity. In this work ionic liquid Butyl-3-methylimidazolium tricyanomethanide ([Bmim][TCM]) was used as a CO₂ absorbent due to its high thermal stability, moderate viscosity and high CO₂ absorption capacity. More detailed information on the selection of the ionic liquid and the validation of the mathematical models by experimental data of the same ionic liquid can be found in our prior publications (Dai et al., 2016a; Dai et al., 2016b; Dai and Deng, 2016; Usman et al., 2017).

The process under assessment in this work is for coal fired power plants. According to literature, a coal fired power plant costs 27–39 \$/ton of CO₂ while natural gas combined cycle plant costs 48–102\$/ton (I.E.A.G.G. Programme, 2006). The cost of CO₂ capture plant integrated to IGCC power plant depend on different factors including place, utilities, choice of separation method. The thermal regeneration of absorbent, absorbent pumping, heating/cooling of liquid and gas streams contribute to added auxiliary cost. Power, heating and cooling energy penalties are comparatively lower in case of Selexol unit for CO₂ absorption than Rectisol and MDEA processes (Cormos, 2011). Dave et al. (Dave et al., 2016) described the process design of CO₂ capture from syngas using DMEPEG as solvent in a packed tower and resulted in 90.4% CO₂ absorption and 89% solvent saturation. The also studied the co-absorbed hydrogen recovery from the solvent and reported to be 55.7% based on process design. The process design of IGCC power plant and CO₂ recovery using Selexol solvent was reported in (Doctor et al., 1993). The CO₂ recovery process helped to control CO₂ emissions by 90.9% at the cost of reduction in net electric power by 145MW. In another study, the 90% CO₂ removal by Selexol solvent accounted for 5–7% reduction in LHV (Thermal efficiency) (Chiesa and Consonni, 1999).

Pressure-swing membrane absorption and desorption process in a closed loop is employed in the present scenario for process simulation studies. The general flow scheme of a pre-combustion CO₂ capture process by means of pressure swing absorption and stripping is represented in Fig. 1.

The absorption of CO₂ in solvent is carried out in an absorber for pre-combustion process. The CO₂-rich solvent is passed through the pressure reduction valve and is fed to the flash tank to separate CO₂ from the rich solvent. After heat exchange with the lean solvent, the rich solvent is fed to the top of the stripper. The depressurization in stripper causes the stripping of CO₂ from loaded solvent. The lean solvent is pumped again to the absorber to complete the continuous process.

Here, a membrane contactor based pressure swing absorption and desorption process for pre-combustion CO₂ capture is designed by

employing an ionic liquid as absorbent. Energy and economic evaluation has been performed for this proposed process. Compared with the process presented in Fig. 1, in the membrane absorption process the conventional packed column is replaced with hydrophobic hollow membrane contactor as absorber unit. The desorption of CO₂ is carried out by flash separators and pressure reduction valves. The ionic liquid [bmim][TCM] absorbs the CO₂ in the absorber which is quite promising solvent. The energy and cost analyses are estimated in order to predict the overall performance of this process and are compared with other CO₂ capture processes that involved physical absorbent in conventional packed column.

2. The proposed process concept

2.1. CO₂ capture in IGCC power plant

Fig. 2 represents the generic layout of an IGCC power plant with CO₂ capture. The coal is converted into carbon monoxide and hydrogen after the gasification process. The hot syngas exiting the gasifier at a temperature of 1100–1500 °C (Scholes et al., 2010) is cooled down and high pressure or low-pressure saturated steam is produced because water is being used as cooling agent. The solid particles contained in the syngas after gasification are removed by water scrubber. The syngas is cooled down to ambient temperature after water wash scrubber.

The sulphur contents which are present in syngas as H₂S, are removed from the syngas in order to avoid the corrosion of the gas turbine and pollutant emissions. The removed hydrogen sulphide is sent to Claus unit to get elemental sulphur. After H₂S removal, the syngas is fed to the CO₂ capture unit. The IGCC power generation efficiency ranges between 40 and 46% (thermal energy efficiency).

2.2. Process flow diagram

Fig. 3a demonstrates the pressure swing absorption-desorption cyclic membrane contactor process for CO₂ separation from shifted syngas. The shifted syngas after particulate and H₂S removal is being considered as feed gas for this process simulation. The feed gas, containing 45% CO₂ and 55% H₂, is passed through the membrane absorber where CO₂ is absorbed in ionic liquid [bmim][TCM] at 20 bar and 50 °C. It was assumed that only CO₂ will be absorbed in ionic liquid [bmim][TCM] and whole amount of H₂ gas leaves the absorber as the solubility of H₂ in the ionic liquid are reported (Kumelan et al., 2006; Lei et al., 2014) remarkably low. The CO₂-rich absorbent leaves the membrane absorber at 20 bar and the pressure of this stream was reduced by introducing it to the pressure-reducing valve. The pressure drop of 11.8 bar occurs in the valve and CO₂-rich absorbent is sent to the flash separator 1 where part of CO₂ is stripped off. The flash separator 1 operates at 7.5 bar pressure. The semi-lean absorbent from flash separator 1 is further introduced to the pressure reducing valve followed by flash separator 2. The flash separator 2 is operated at atmospheric pressure (1 bar). The lean absorbent is at low pressure after flash separator 2 and pressure is increased to 20 bar by booster pump. Heat exchanger is installed after the pump to bring the temperature of the solvent to 50 °C. The stripped CO₂ from both the flash separators is mixed and compressed to 75 bar as shown in Fig. 3b. Membrane absorber and flash separators accomplish the pressure swing in this process.

2.3. Property models of liquid, gas and membrane

2.3.1. Property model of the gas

The shifted syngas is a mixture of carbon dioxide and hydrogen along with some other gases in traces. The gas stream assumed here is after the removal of H₂S and other impurities. To simplify the process simulation, only carbon dioxide and hydrogen are considered in the shifted syngas. The fluid package used in HYSYS is Peng-Robinson EOS,

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