



RESEARCH ARTICLE

A GLOBAL PERSPECTIVE OF CO₂ CAPTURE – AN OVERVIEW

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ABSTRACT

There is growing concern that anthropogenic carbon dioxide (CO₂) emissions are contributing to global climate change. Therefore, it is critical to develop technologies to mitigate this problem. The high capture cost and energy consumption would be a major barrier for implementing CO₂ capture mitigation methods. Alternative technologies may offer improvement. This paper reviews several separation technologies applicable to CO₂ capture from flue gas, and discusses improvement opportunities and research needs. The main object of the reviewers is to synthesize the novel chitosan based ionic liquid, as a potential solvent for post-combustion CO₂ capture. To enhance the absorption performance of CO₂ capture.

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INTRODUCTION

Carbon dioxide removal is a form of carbon capture, and it is a part of a potential process of carbon sequestration. It removes carbon dioxide from large point sources, such as fossil fuel fired power stations. It is regarded as greenhouse gas remediation, which is a branch of geoengineering. Some commentators regard air capture as a form of carbon capture and storage, Carbon dioxide removal methods are notably supported by IPCC (Intergovernmental Panel on Climate Change) chief Rajendra Pachauri (Pagnamenta, 2009). Air capture is not generally seen as an attractive alternative to capture at large, point source emitters (such as power plants), as it is likely to be more efficient and cheaper to capture and store carbon dioxide from more concentrated streams. There are, however, some advantages of air capture as it removes the need for CO₂ piping to transport the gas to underground storage sites, and allows the use of renewable energy and optimal storage sites.

It is particularly effective at dealing with small point sources such as domestic heating systems and vehicle exhausts, where piping of exhaust gases is impractical (Lenton & Vaughan, 2009). The mitigation effectiveness of air capture is limited by societal investment, land use, and availability of geologic reservoirs. These reservoirs are estimated to be sufficient to sequester all anthropogenically generated CO₂ (LiveScience, 2007). As it is extremely distinct that the greenhouse effect is caused by mankind, sturdy measures must be taken to find solutions to conflict or slow global warming. Today, most researchers are confident about the carbon dioxide mitigation

by carbon capture and storage is a feasible option to significantly reduce the level of carbon dioxide emissions. One of the most promising processes is post-combustion, the capturing of CO₂ after the combustion process (Zeman & Lackner, 2004). One advantage of this process is the potential for retrofit to existing power plants without changing the existing processes. This circumstance will result in high availability, flexibility and reliability of the power plant operation which will prevail Carbon Capture and Storage (CCS) in the beginning (Hallvard et al., 2011). CO₂ scrubbing or absorption is the most predominantly proposed post-combustion capture (PCC) process, since it is the most developed process of all CCS options that has been used for decades for applications, such as in the chemical industry.

In most CO₂ scrubbing applications, CO₂ is captured from the process stream by absorption into a solvent and subsequently released from the solvent by stripping by changing the temperature. By condensing the water out of the flue gas, highly concentrated CO₂ can be gained and stored underground after compression. By determination of the total costs of capture, transport and storage it is evident, that the capturing of CO₂ is the most cost intensive stage (Bates et al, 2002). Hence significant scientific effort has been directed to find more effective process technologies by the identification or development of new solvents. Increasing the performance is directly linked with lowering operating costs, which opens an enormous saving potential. In the past, much research has been completed in the development of new amines or blend of amines, but also ammonia, amino acids and potassium carbonate. More recently ionic liquids have started to be considered as potential candidates. Ionic liquids (ILs) are very promising for capturing CO₂ because they have some potential

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advantages in comparison to other solvents such as monoethanolamine (MEA) based solutions. Ionic liquids are salts with a melting temperature below the boiling point of water (Blanchard et al., 1999). Most ionic liquids have an organic cation and an inorganic anion. Pure ionic liquids contain only ions and no solvent (e.g. water) is necessary. A major advantage of ILs is that the properties can be selected on the basis of the choice of anion, cation or other substituents. This flexibility leads to innumerable different structures and variation possibilities. Concern over the increased concentration of CO₂ in the atmosphere and its effect on global climate change has increased the awareness and investigation for reducing CO₂ emissions. Most of the methods for mitigation require CO₂ in a concentrated form, while the CO₂ from fossil fueled power plants is mixed with nitrogen, water vapor, oxygen, and other impurities and has low concentrations (15 % for coal fired power plants, and less than 10 % for natural gas fired plants). Therefore, capturing CO₂ from flue gas is an important common link.

The current commercial operations for capturing CO₂ from flue gas use a chemical absorption method with Monoethanol Amine (MEA) as the sorbent. An estimate by Bergman et al using data from an AB Lummus Crest brochure indicated the cost of capturing a ton of CO₂ including removing impurities and compressing CO₂ to a supercritical pressure. While compression is energy intensive and expensive component, the capture part of MEA method actually uses more energy and costs more than compression. The high capture cost is a major barrier for implementing near term carbon sequestration methods such as injecting CO₂ for enhanced oil recovery (Zeman & Lackner, 2004). The other major shortcoming of the current technology is its high-energy consumption. The amine based method uses a large amount of low-pressure steam for sorbent regeneration, and the total power output is significantly reduced. It could lead to ' 20% reduction in electricity production for a power plant (107MW/854MW) as a study by Booras and Smelser indicated (Bergman et al, 1997). Following is a preliminary analysis of several separation technologies applicable to CO₂ capture, new power generation configurations that may simplify CO₂ capture, as well as a discussion of some improvement opportunities. Chemical absorption of acid gases, such as CO₂, H₂S or SO₂ has been widely used in the industrial process.

At present, one of the effective methods for the uptake of CO₂ usually employs aqueous solutions of alkanolamine or their mixtures, including monoethanolamine (MEA), diethanolamine (DEA), and N-methyldiethanolamine (MDEA). However, the use of alkanolamine presents several concerns (Brennecke et al, 2002). Corrosion of amines system limits the concentration of amine in the solution and causes extra cost; (A) amines are prone to degrade under high temperature especially in the regeneration process; (B) oxidative degradation of amines occurs in the presence of oxygen; (C) amines are volatile and lost into the gas stream. The loss of absorbents not only produces extra waste streams but also leads to additional absorbent cost. It should be noted that among the amines usually used, MDEA has lower volatility, thermal stability, less alkaline, higher CO₂ load (up to 1.0 mol CO₂/mol amine) and less regeneration cost, though the absorption of CO₂ into MDEA is quite slow since MDEA just promotes CO₂ hydrolysis to form bicarbonate. Therefore,

in the industrial application, aqueous solution of MDEA is usually mixed with activators of fast reactivity, including MEA, piperazine (PZ), and DEA (Cheng, 2000). Reducing CO₂ emissions for addressing climate change concerns is becoming increasingly important as the CO₂ concentration in the atmosphere has increased rapidly since the industrial revolution. Many mitigation methods, including CO₂ sequestration and novel CO₂ utilization, are currently under investigation. Most of these processes require CO₂ in a concentrated form. However the CO₂ from large sources such as fossil fueled power plants is mixed with nitrogen, water vapor, oxygen, and other impurities (Crosthwaite et al, 2005). A typical CO₂ concentration from coal fired power plants is around 15 %, while CO₂ concentrations for natural gas fired plants are less than 10 %. Therefore, capturing CO₂ from flue gas is an important common link for many mitigation methods.

The current commercial operations for capturing CO₂ from flue gas use a chemical absorption method. The cost of capturing a ton of CO₂ including removing impurities and compressing CO₂ to supercritical pressure using existing MEA technology would be in the order of \$40, and the power output would be significantly reduced by the energy consumption in capturing and compressing CO₂. The high capture cost and energy consumption would be a major barrier for implementing CO₂ mitigation methods. Alternative technologies may offer improvement (Daiguji and Hihara, 1999). Ionic liquids (ILs) are organic salts that form stable liquid below 100 °C or even at room temperature (room temperature ionic liquids, RTILs). For the removal of acidic gases, compared with alkanolamines, RTILs have negligible volatility and significant thermal stability that avoids loss of absorbents. First time Brennecke's group reported that, CO₂ was highly soluble in [bmim][PF₆] (1-butyl-3-methylimidazoliumhexafluoro phosphate), reaching a mole fraction of 0.6 at 8MPa. Since 0.2MPa was probably the lowest limit of the Henry's law constant for the physical absorption of CO₂ in ILs at 298K when pressure of CO₂ was below ambient pressure, the solubility of CO₂ in conventional ILs was only up to 0.035 mol fraction. By introducing functional groups that could enhance the CO₂ absorption into the anion or cation, functionalized ionic liquids are expected to have significant performance in the uptake of CO₂ (Dupont, 2004).

CO₂ Capture

Capturing CO₂ from the flue gas is essentially a gas separation process. Several separation methods, including Chemical Absorption, Physical Absorption / Adsorption, Membrane Technologies, and Cryogenic Separation, can achieve such a goal. The judging criteria's are

- Capture effectiveness,
- Process economy,
- Energy consumption,
- Other technical and operational issues.

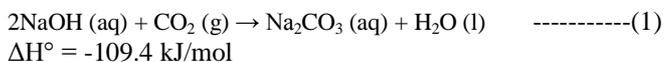
Other measures such as modifying of power generation configurations by using CO₂ Combustion mode and oxygen blown Gasification Combined Cycle could alter the flue gas composition and pressure that could help the CO₂ capture. Such strategy has some benefit but also raises new technical

challenges (Hiller et al, 1989). Fossil fuel is still the most important and energy source with almost no other alternate. On the other hand, it is also the major source for green house gases that are assumed to cause global warming. The over all efficiency of a power plant is significantly reduced if CO₂ is captured from flue gas. So far the only reliable methods for reducing CO₂ emissions are:

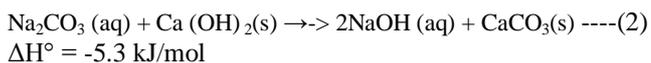
1. A decrease of fuel consumption
2. Increase in process efficiency
3. Switch to lower carbon content fuels e.g. Natural gas instead of Coal.
4. Enhance the natural 'sinks' for CO₂, e.g. forests, soil and the ocean, which drawdown CO₂ from the atmosphere.
5. Use energy sources with very low or neutral CO₂ emissions, such as renewable energy e.g. biomass, wind or nuclear energy. (Lepaumier et al, 2009)

Existing technologies for CO₂ capture are: Scrubbing

Various scrubbing processes have been proposed to remove CO₂ from the air, or from flue gases. These usually involve using a variant of the Kraft process. Scrubbing processes may be based on sodium hydroxide (Kishimoto et al, 2009. Reddy et al, 2008). The CO₂ is absorbed into solution, transferred to lime via a process called causticization and released in a kiln. With some modifications to the existing processes, mainly an oxygen-fired kiln, the end result is a concentrated stream of CO₂ ready for storage or use in fuels. An alternative to this thermo-chemical process is an electrical one in which an electrical voltage is applied across the carbonate solution to release the CO₂. While simpler, the electrical process consumes more energy as it splits water at the same time. It also depends on electricity and so unless the electricity is renewable, the CO₂ produced during electricity production has to be taken into account (Hessen, 2010). The early incarnations of air capture used electricity as the energy source and therefore depended on carbon-free sources. A thermal air capture system uses heat that can be generated on-site, reducing the inefficiencies associated with producing electricity, but of course it still needs a source of (carbon-free) heat. Concentrated solar power is an example of such a source (Lepaumier et al, 2009). Zeman and Lackner outlined a specific method of air capture (Erga et al, 1995). First, CO₂ is absorbed by an alkaline NaOH solution to produce dissolved sodium carbonate. The absorption reaction is a gas liquid reaction, strongly exothermic, (equation No.1)



The carbonate ion is removed from the solution by reaction with calcium hydroxide (Ca (OH)₂), which results in the precipitation of calcite (CaCO₃). The causticization reaction is a mildly exothermic, aqueous reaction that occurs in an emulsion of calcium hydroxide (equation No.2)

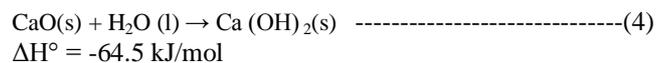


Causticization is performed ubiquitously in the pulp and paper industry and readily transfers 94% of the carbonate ions from the sodium to the calcium cation (Freguia & Rochelle, 2003).

Subsequently, the calcium carbonate precipitate is filtered from solution and thermally decomposed to produce gaseous CO₂. The calcination reaction is the only endothermic reaction in the process and is shown (equation No.3).



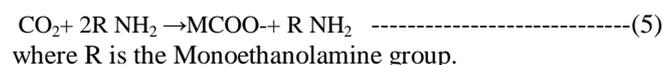
The thermal decomposition of calcite is performed in a lime kiln fired with oxygen in order to avoid an additional gas separation step. Hydration of the lime (CaO) completes the cycle. Lime hydration is an exothermic reaction that can be performed with water or steam. Using water, it is a liquid/solid reaction as shown (equation No.4).



- Physical solvent e.g. Selexol
 - Based on Henry's Law (Temperature & Pressure dependent with absorption occurring at low temperature and high pressure)
 - Applicable for high CO₂ concentration (CO₂ partial pressure >525 kPa).
- Chemical solvent e.g. Mono-ethanol amine (MEA)
 - Depends on acid base neutralization reaction
 - Applicable for low to moderate CO₂ partial pressure (3.5 to 21.0 kPa)

Chemical absorption

Chemical absorption uses the different reactivities of various gases with sorbents to separate them. The reactions need be reversible so that the spent sorbent can be regenerated. For separating CO₂ from flue gas, chemical absorption appears appropriate because CO₂ is acidic and the majority of the rest of flue gas, nitrogen, is not. CO₂ can be absorbed by many basic sorbents including alkali carbonate, aqueous ammonia, and alkanolamines. The attention needs to be paid is how the sorbents can be regenerated. The binding between sorbent molecules and CO₂ generally is strong and this offers a fast and effective removal of most of CO₂ in one stage of absorption. However, the strong binding between CO₂ and the sorbent molecules is also one of the causes for high regeneration energy requirement (Hiller et al, 1989). Second concern is the control of impurities and minor components in the flue gas including SO₂ oxygen, etc. that may degrade the sorbents. These components have to be removed before the gas enters the absorber, or treated with appropriate measures. Lastly, because many sorbents are corrosive, only diluted solutions (around 18% for MEA) are used.



In addition to the regenerator, a reclaiming operation is conducted periodically to recover amine by decomposing heat stable salts and to dispose of degradation products. It is the most suitable method for the separation of CO₂ from exhaust gases, when carbon dioxide has a low concentration (5-15% by volume) in a gaseous stream at atmospheric pressure.

The separation process of carbon dioxide by chemical absorption consists of two steps:

1. The absorption of CO₂ by chemical solvents at a low temperature (40-65°C)
2. The recovery of CO₂ from chemical solvents by using low-grade heat (a temperature in the range of 100-150°C), usually extracted from power plants.

Absorption reaction



$$P_{\text{CO}_2} = K \frac{\text{MEAH}^+ \text{MEACOO}^-}{\text{MEA}} \frac{1}{1 - 2 \frac{\text{CO}_2 \text{ Loading}}{\text{Total MEA}}}$$

In the MEA process CO₂ from the cooled power plant exhaust gas reacts with aqueous solution of MEA in a contacting device, usually an absorption tower at a pressure slightly above the ambient pressure and at a temperature depending on the power plant upstream. First, they are compressed to about 1.3 bars, to overcome pressure drops within the system, and cooled to nearly 50°C. Then, the gases go to the absorption column where the carbon dioxide binds to the solvent chemically. Most of the CO₂ is thus removed from the exhaust gas that is released to the atmosphere and the rich solution (i.e. the solution containing the absorbed CO₂) flows to a lean/rich heat exchanger: here the hot lean solution, coming from the stripper column (regenerator), cools itself giving out its heat to the rich solution, which then goes to the regenerator. Here the solvent is regenerated by heat where the chemical bonds are decomposed thermally. The CO₂ and water vapour leaving the stripper is next cooled and essentially pure CO₂ leaves the separation plant for further treatment, in this case compression and drying (Kim & Svendsen, 2010). There are two energy requirements for this process:

1. Regeneration heat for the solvent: According to literature the required heat is 4 MJ per kg of recovered carbon dioxide, which is provided by the low pressure steam from the power plant. Assuming the temperature for the regeneration of the solvent about 150°C so that the extraction of the steam takes place at about 5 bars. The steam provides its latent heat of condensation and returns into the power plant as saturated liquid at the same pressure as its extraction. Thus, the amount of steam for the regeneration can be determined by the quantity of CO₂ recovered and by the characteristics (temperature and pressure) of the extracted steam.
2. Energy for the compression of the flue gases and for the pumping of the amine solution through the removal plant: According to literature, the required energy is 0.11 MJ per kg of CO₂ recovered. This amount of energy is subtracted from the total plant work to obtain the network.

Avoiding oxidation

Following steps can be adapted to control the rate of degradation in the oxidizing environment of a flue gas.

- Minimize contact time with dissolved O₂
- Minimize dissolved metals, Nox
- Add chelators or free radical scavengers

- Use solvents that oxidize slower
 - Hindered amines with tertiary or quaternary C's
 - Tertiary amines
 - K₂CO₃

Physical Adsorption/Adsorption

- In which a solid absorbent (such as activated carbon, zeolites) is passed through the gas stream, and the CO₂ is held on the surface of the particles by (non-chemical) surface forces. Once collected, the particles are heated, releasing (desorbing) the CO₂.
- In pressure swing adsorption (PSA), the gas mixture flows through a packed bed of adsorbent at elevated pressure until the adsorption of the desired gas approaches equilibrium conditions at the bed exit. The bed is then regenerated by stopping the feed mixture and reducing the pressure.
- In temperature swing adsorption (TSA), the adsorbent is regenerated by raising its temperature.
- Adsorption is not yet considered attractive for large-scale separation of CO₂ from flue gas because the capacity and CO₂ selectivity of available adsorbents is low. However, it may be successful in combination with another capture technology.
- Electric Swing Adsorption (ESA) (Lepaumier et al, 2009).

In physical absorption, the gas CO₂ molecules are dissolved in a liquid solvent, and no chemical reaction takes place. The binding between the CO₂ molecules and solvent molecules, being either Van der Waals type or electrostatic, is weaker than that of chemical bonds in chemisorption. The amount of gas absorbed is linearly proportional to its partial pressure (Henry's law). The curves show linear relations between the partial pressure and CO₂ loading for physical sorbents, (Mimura et al, 1997) and nonlinear relations for chemical sorbents. Thus the physical absorption is more effective when the partial pressure of the gas to be absorbed is high. The amount of gas absorbed also depends on the temperature. The lower the temperature and more gas is absorbed. Typical sorbents for CO₂ are methanol, N-methyl-2-pyrrolidone, polyethylene glycol dimethyl ether, propylene carbonate and sulfolane (Hallvard et al, 2011). Desorption can be achieved either by lowering pressure as in the pressure swing absorption (PSA), or raising the temperature as in the temperature swing absorption (TSA). Physical absorption has been used in gas production processes (Zeman & Lackner, 2004) to separate CO₂ from hydrogen and CO.

These processes include: Rectisol that uses methanol as solvent, Selexol that uses dimethyl ether of polyethylene glycol (DMPEG), Sepsolv that uses n-oligoethylene glycol methyl isopropyl ethers (WE), Purisol that uses N-methyl-2-pyrrolidone (TMp), and Gasolan that uses N-methylcaprolactam (NMC). In physical adsorption, gas is adsorbed on the solid surface by a Van der Waals force. Most important adsorbents are activated carbon, zeolite, silica gel, and aluminum oxide. The separation is based on the difference in gas molecule sizes (Steric Effect), or different binding forces between gas species and the adsorbent (Equilibrium Effect or Kinetic Effect (LiveScience, 2007)). Like physical absorption, two types of processes: Pressure Swing Adsorption and Temperature Swing Adsorption are used.

Because the gas molecules are attached on the solid surface and form mono or multi-layers in physical adsorption, the gas loading capacity could be lower than in physical absorption, (Chakma A, 1997) even though many adsorbents have large surface area per unit volume. Because of the large volume of CO₂ in the flue gas, it appears physical adsorption might not be an effective and economical solution for separating CO₂ from flue gas. The other limit in using physical adsorption for this purpose is the low gas selectivity of available sorbents (Hallvard et al, 2011). However in combining with other capture methods, physical adsorption may become attractive. Such applications include membrane technologies. Membrane Technologies Many membranes are made with similar materials used for physical adsorbents, because the physical quality, such as porous structure and selective gas affinity, are also useful for making membranes. Membranes are microscopic sieves (Falk Pendersen and Dannstrom, 1997). Under an applied pressure some molecules will pass through the micro pores in membranes, and some molecules will be stopped.

The driving forces for gas separation using membranes are generally hydrostatic pressure and concentration gradient. There has been some work done in electrically driven separation of gases via ion conduction. Two types of membrane technologies can be used for separating CO₂ from other gases: gas separation membranes and gas absorption membranes. In using gas separation membranes a hydrostatic pressure is applied and the difference in permeability of gas species leads to separation of these gases (Arwnwilas and Tontiwachwuthikul, 1997). Although using separation membranes to separate CO₂ from light hydrocarbons has met with considerable success in the petroleum, natural gas and chemical industries, this technology may not be preferable to separate CO₂ from flue gas because of the large volume of the flue gas and the compression energy requirement. A study shows that a good separation using a two-stage system would cost twice much of the conventional amine separation processes (Hallvard et al, 2011). In using gas absorption membranes, a liquid sorbent is used to carry away CO₂ molecules that diffuse through the membranes, and no high hydrostatic pressure is required.

In this technology, the membranes serve as an interface between the feed gas and liquid sorbent. The membranes can be made in the forms of parallel hollow fibers, and feed gas and sorbent are moving on the opposite sides of the fibers (Feron and Iansen, 1997). This configuration provides significantly greater liquid-gas contact area than packed absorbers, and can reduce the size, and possibly the cost of the absorber (Tokuda et al, 1997). Additional benefits include the independent control of gas and liquid flows, and minimization of flooding, channeling, or foaming of liquid sorbent in the absorber (Hallvard et al, 2011). This configuration was under investigation by Norwegian researchers as a choice for offshore operation where the space and weight of the absorber are important factors. Considerable research is still required to address issues such as how to deal with possible clogging of hollow fibers by fine particles in the flue gas.

Membrane Systems

- Porous or semi-porous structure, through which some chemical species permeate more easily than others.

- Gas Separation Membranes :- differences in physical or chemical interactions between gases and a membrane material, causing one component to pass through the membrane faster than another (porous inorganic: palladium membranes, polymeric membranes and zeolites)
- Multiple stages and/or recycle of one of the streams are necessary which leads to increased complexity, energy consumption and costs.
- Gas absorption Membranes
- Solvent assisted membranes demonstrated (Atkins & dePaula, 2006).

Cryogenic fractionation

- This involves the compression of the gas stream, and cooling it to a temperature low enough to allow separation by distillation. The resulting liquid CO₂ may then be removed for disposal
- Used for high CO₂ concentrations (typically >90%).
- High energy required for refrigeration.
- Some components such as water have to be removed before the gas stream is cooled, to avoid blockages
- Applicable for high-pressure gases, such as in pre-combustion capture processes or oxygen-fired combustion (Lepaumier et al, 2009).

Cryogenic separation of gas mixtures uses the difference in boiling points of various gas species to separate them. All gas species have a distinctive boiling temperature, the cryogenic method -56.8^oC and 31.6^oC respectively, between these temperatures, CO₂ can be liquefied by compression and cooling (Bergman et al, 1997). The major disadvantage of cryogenic method is the high-energy consumption and costs associated with gas compression and cooling. For CO₂ sequestration, liquid CO₂ is required in many cases, and the work spent on liquefying CO₂ is not wasted. However, since the concentration of CO₂ in flue gas is about 15 % or less, the energy used to compress the rest 85% of flue gas is substantial. A simple calculation for the energy requirement for liquefying CO₂ by isothermally compressing the flue gas near the critical temperature to 74 bars would spend about 30% total power output in compressing 85% of the remaining gases, and this is about 50% more than MEA process (Booras & Smelser, 1991). Methods to recover that amount of compression energy spent on 85% gas components could lower total energy consumption. Generally speaking, unless novel process schemes can be developed, cryogenic separation is an energy intensive operation (Bergman et al, 1997).

The amount of CO₂ that can be removed from the exhaust depends on the size of the absorption unit and the concentration of CO₂ in the exhaust. For a standard plant the economical recovery limit is approximately 85% for 3% CO₂ in the exhaust and 90-92% for 8%. A key requirement is to limit losses of solvent both as 'carry-over' in the flue gas and as heat-stable salts. There are a few facilities in which amines are used to capture CO₂ from flue gas streams. Improved solvents could reduce energy requirement by as much as 40 % compared to conventional MEA solvents. There is considerable interest in the use of sterically hindered amines, which are claimed to have good absorption and desorption characteristics (Hessen, 2010). Therefore the reviewers are

exploring on the synthesis of the novel chitosan based ionic liquid, as a potential solvent for post-combustion CO₂ capture.

Conclusions

The present chitosan based ionic liquid, is not necessarily the best choice for the post-combustion CO₂ capture. The method presented here with a particular ionic liquid is merely a demonstration for such applications. However, it is shown that the present ionic liquid can affect the gaseous selectivity and future work will explore more optimal ionic liquid choices.

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