

Solvent Development for Post-Combustion CO₂ Capture: Recent Development and Opportunities

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Abstract. Chemical absorption is widely regarded as the most promising technology for post-combustion CO₂ capture from large industrial emission sources with CO₂ separation from natural gas using aqueous amine solvent system having been applied since the 1930s. The use of monoethanolamine (MEA) in CO₂ absorption system possesses several drawbacks, such as high regeneration energy, high solvent loss, and high corrosion tendency. Various solvents have been developed for post-combustion CO₂ capture application including the development of aqueous solvents and phase-change solvents. Some of these alternate solvents have been reported to have better solvent properties, which could improve the CO₂ absorption system performance. This paper reviews key parameters involved in the design improvement of several chemical absorption process systems. In addition, some novel solvent systems are also discussed, for example encapsulated solvents systems. Some of the key solvent parameters that affect the capture performance, such as heat of reaction, absorption rate, solvent working capacity, solvent concentration, and solvent stability, are discussed in this paper, particularly in relation to the economic viability of the capture process. In addition, some guidelines for the future solvent development are discussed.

1 Introduction

A CO₂ reduction scheme that is gaining growing interest is Carbon Capture and Storage (CCS) [1]. Within the CCS process chain, CO₂ capture is the costliest stage and therefore it is important to develop the technologies that can reduce costs. Among all CO₂ capture methods, post-combustion CO₂ capture using chemical absorption has been recognized as the most commercially ready technology. The concept of this technology has been applied, albeit at different feed gas sources, in natural gas industry since the 1930s, where CO₂ is absorbed using aqueous amine solvent system [2]. Many researchers suggested that other CO₂ removal methods, such as membranes and adsorption, are not likely to be competitive because of compression work [2]. The application of physical absorbents in post-combustion CO₂ capture is likely to be more limited than that of chemical absorbents because of the low CO₂ partial pressure in the flue gas [3]. The future development of chemical absorption will be the focus of this paper. This paper aims to review the key parameters involved in the

design improvement of several chemical absorption process systems. In particular, the recent updates on solvent development are presented for two solvent classes (aqueous solvents and phase-change solvents). In addition, some novel solvent systems are also discussed, for example encapsulated solvents systems.

2 Post-combustion CO₂ Capture using Chemical Absorption

In CO₂ capture using chemical absorption, weak chemical bonds between CO₂ from emission gases and a solvent solution is formed. Heat is then typically provided in the regeneration column in order to reverse this reaction. A simplified schematic of the post-combustion CO₂ capture process using chemical absorption is shown in Figure 1. Various solvents have been developed for post-combustion CO₂ capture application in order to improve the performance of the absorption system compared to MEA [4]. Typically, these alternate solvents have better solvent properties..

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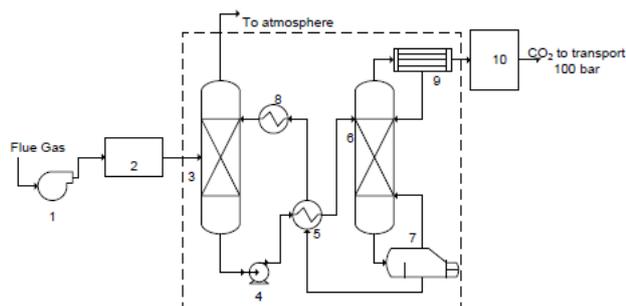


Fig. 1. Simplified process flow diagram for CO₂ capture process using chemical absorption (1) Blower, (2) Pre-treatment, (3) Absorber, (4) Solvent pump, (5) Cross heat exchanger, (6) Stripper, (7) Reboiler, (8) Lean solvent cooler, (9) Condenser, (10) Post-treatment (dehydration and CO₂ transport compressor)

Some of the key solvent parameters that affect performance are:

- Heat of reaction - The use of solvents with a low heat of reaction may decrease the regeneration energy. However, the reduction in regeneration energy for solvents with a low heat of reaction can only be achieved if it is not coupled with high water evaporation in the stripper [5].
- Absorption rate - By formulating solvents with a high absorption rate, the dimensions of the absorber decrease, reducing the capital cost [6].
- Solvent working capacity - The difference between the solvent rich and lean loading is commonly defined as solvent working capacity. Improvement in this property would decrease the solvent circulation rate and potentially reduce the regeneration energy [7-8]. A higher solvent capacity also reduces the dimensions of the absorber, heat exchangers, pumps, and piping, thus lowering the capital cost [8].
- Solvent concentration - High solvent concentrations translate to a low solvent circulation rate, and hence it only requires relatively small dimensions of heat exchangers, absorber, stripper, and pipelines. This also leads to a decrease in the regeneration energy because of the lower amount of water that goes into the stripper [7-8].
- Solvent stability - Solvent degradation accounts for 15 – 25 % of total solvent losses for the MEA system. By formulating a solvent with better solvent stability, the operational cost for solvent make-up cost decreases [9].

Table 1 summarizes the key solvent properties of various solvent groups reported in literature based on their molecular structure. The typical ranges of values for the heat of reaction, absorption rate, and solvent capacity are also given. For the amine-based solvents, the values of absorption capacity and absorption rate are gathered from a study by Puxty et al. [10]. For other solvent groups, the level of information of solvent properties is not as extensive as amine-based solvents.

As a consequence, the values presented in Table 1 for amino acid salts, ionic liquids, and deep eutectic solvents may not encompass all solvents within these solvent groups, but are representative of the most prevalent solvents for these groups.

3 Aqueous Solvents

In most industrial applications, only two phases are involved in the CO₂ absorption, the gas phase made up of the gas/gases to be recovered and the solvent liquid phase. In the following section, the key characteristics of different groups of aqueous solvents are discussed.

3.1. Primary Amines

As shown in Table 1, there are various solvents belonging to the primary amines group including MEA, DGA, and EDA. The most widely used is MEA. MEA was first applied and widely developed to capture CO₂ from feed gases with low partial pressure of CO₂ as it possesses fast reaction rate and high working capacity compared to other solvent groups. MEA has several advantages over other amine-based solvents [9, 11-12], such as high reaction rate, high absorption capacity on the mass basis (because of low molecular weight), good thermal stability, and cheap.

However, MEA owns deficiencies such as high energy penalty to regenerate the solvent, corrosive, and high solvent make-up rate [4]. MEA has a high heat of reaction (82 kJ/mole), thus it requires high regeneration duty in the stripper. This is due to the formation of the stable carbamate ion (MEACOO⁻). The regeneration energy of the generic MEA 30 %-wt. is reported to be between 4.1 to 4.4 MJ/kg CO₂ [8, 13-15]. The high level of solvent loss in an MEA system is due to the vaporization at the absorber. The other major issue regarding the solvent loss is the degradation of solvent, which has several causes (oxidative and thermal reactions). Make-up solvent is required to compensate for the loss that occurs in the process. Some operating strategies that have been implemented to address the issue of solvent loss include limiting the solvent concentration while corrosion effects are dealt with by ensuring proper equipment material selection and utilizing mild operating conditions [8].

3.2 Secondary and Tertiary Amines

CO₂ separation from natural gas using secondary and tertiary amines, such as DEA and MDEA, have been commonly applied commercially for decades. DEA and MDEA have a lower heat of reaction and a higher working capacity compared to MEA and thus require lower regeneration energy. The drawback of these solvents is a much slower absorption rate compared to MEA. Aqueous solvents consisting entirely of secondary and tertiary amines are unlikely to be applied in post-combustion CO₂ capture applications.

Table 1. Solvent properties of various solvent groups

Solvent group	Examples	Working capacity (moles CO ₂ /mole solvent)	Absorption rate (min ⁻¹)	Heat of reaction (MJ/kg CO ₂)	Solvent class
Primary amines	Monoethanolamine (MEA) Ethylenediamine (EDA)	0.15-0.69	0.003-0.032	70-85	Aqueous
Secondary amines	Diethanolamine (DEA) Diisopropanolamine (DIPA)	0.10-1.0	0.004-0.031	65-75	Aqueous
Tertiary amines	Methyldiethanolamine (MDEA) Triethanolamine (TEA)	0.07-1.1	0.001-0.008	50-65	Aqueous
Hindered amines	Aminomethylpropanol (AMP) 2-piperidineethanol (2-PE)	0.25-1.0	0.003-0.02	50-75	Aqueous
Polyamines	Piperazine (PZ) Hydroxyethylpiperazine (HEP)	0.50-1.8	0.003-0.3	66-78	Aqueous
Alkali carbonates	Potassium carbonate (K ₂ CO ₃) Sodium carbonate (Na ₂ CO ₃)	0.4-1.0	0.001-0.002	30-50	Aqueous and phase-change
Ammonia	Ammonia (NH ₃)	0.35	<0.007	27	Aqueous and phase-change
Amino acid salts	Potassium taurate Potassium sarcosine	~0.1-1.0	0.003-0.02	60-80	Aqueous and phase-change
Ionic Liquids	([bmim]-PF ₆) ([bmim]-BF ₄)	<0.5	<0.001	15-100	Aqueous and phase-change
Deep eutectic solvents	Choline chloride (ChCl)/urea Choline chloride (ChCl)/MEA	<0.2	<0.001	15-100	Aqueous

The absorption rate and kinetics of DEA solution have been reported in a study by Rinker et al. [16] and Cadours et al. [17], while the kinetics for MDEA solution have been investigated in a study by Ko and Li [18]. The other possible drawback of using DEA or MDEA in post-combustion CO₂ capture applications is that they are less suitable for applications where the partial pressure of CO₂ is low (less than 100 kPa) [19]. The rate promoter addition (such as MEA, piperazine, and carbonic anhydrase) or blending with other solvents have been reported to be able to lessen this deficiency [19-22].

3.3 Hindered Amines

Hindered amine compounds are commonly recognized when the nitrogen atom of the amine molecule adds a bulky substitute group. The bulkier substitutes attached to the amine backbone lead to a lower rate of carbamate formation and hence the bicarbonate formation will then be the main reaction mechanism [23-24]. Furthermore, as the reaction mechanism in hindered amines is dominated by bicarbonate formation, the heat of reaction of hindered amines is lower than MEA. A proprietary hindered amine, trademarked as KS-1, has been developed by Mitsubishi Heavy Industries (MHI) in 1990 [25]. The CO₂ capture process using KS-1 is similar to the MEA (Figure 1), except that it incorporates a heat recovery system and absorption inter-cooling. It was reported that the regeneration energy obtained during testing was 2.53 MJ/kg CO₂, which is up to 40 % less than MEA [26]. KS-1 has a larger loading capacity, thus the circulation rate of KS-1 is 40 % lower than MEA.

3.4. Polyamine

The most widely known polyamine solvent for CO₂ absorption is piperazine (PZ), which has been

commercially developed as a rate promoter for various solvent types, such as MDEA and potassium carbonate. When PZ is used as a promoter, the concentration used is low, between 0.5 and 2.5 m PZ, because PZ is not highly soluble. The application of CO₂ capture using aqueous concentrated PZ has attracted increasing interest since the early 2000's. It is reported that in order to preserve a liquid solution without precipitation at ambient temperature for 8 m PZ, a CO₂ loading of approximately 0.25 mole CO₂ per mole alkalinity is necessary [27]. Under these conditions, the working capacity of 8 m PZ (0.90 mole CO₂ per kg PZ+H₂O) is twice that of 7 m MEA (0.43 mole CO₂ per kg MEA+H₂O), while the rate of CO₂ absorption into 8 m PZ is up to triple the rate of 7 m MEA. The drawback of PZ is that it forms nitrosamines, a class of carcinogenic compounds, at faster rates compared to MEA [28]. This could hinder the use of PZ in CO₂ capture applications because nitrosamines can be released from the absorber units through aerosol formation and accidental spills [28].

3.5 Alkali Carbonate

A hot potassium carbonate process has been used for CO₂ absorption from natural gas and ammonia synthesis since the 1960's. In this process, the CO₂ is captured at high pressure and the concentration of the potassium carbonate is limited to 20-30 %-wt. to prevent solid precipitation. Alkali-carbonate solvents have several benefits compared to other solvents including low heat of reaction and low solvent cost [5, 29-31]. The CO₂ is absorbed through bicarbonate formation, which leads to the significantly lower heat of reaction than that of MEA.

Endo et al. (2011) reported that the regeneration energy of an aqueous potassium carbonate system is 4.9 MJ/kg CO₂, which is 10-20 % higher than that of MEA. Although potassium carbonate has a lower heat of reaction than MEA, the regeneration energy is

Figure 2 shows the simplified process flow diagram of CO₂ absorption using phase-change solvent. The most commonly used solvents for this process are based on three molecular structure groups, namely amino acid salts, alkali carbonates and ammonia.

4.1. Amino Acid Salts

Amino acid salts are one of the phase-change solvents currently under development for CO₂ capture. Some of the attractive characteristics of amino acid salts are high solvent working capacity, relatively fast reaction kinetics, good stability towards degradation, favorable heat of reaction, and low environmental impact [49]. Kumar et al. reported that amino acid salts show negligible volatility because of the ionic nature [50].

The CO₂ solubility in various amino acid salts has been reported in recent studies [50-54]. Portugal et al. (2009) reported that the CO₂ absorption capacities of potassium glycinate and potassium threonate solutions are within the same order of magnitude as that of MEA 30 %-wt. Ma'mun and Kim [53] reported that the CO₂ absorption capacity of potassium sarcosine is 34 % higher than that of MEA. In a recent study by [49], two processes to enhance the absorption and desorption process of a precipitating amino acid solvent system was introduced (DECAB and DECAB Plus). The addition of a solid-liquid separator between the absorber and the stripper may result in a stripper feed stream with a higher solid fraction. Therefore, the pH of the rich solution changes and it consequently enhances the hydrolysis of the carbamate species producing amino acid salts and bicarbonate. This translates to a lower heat of desorption required in the stripper. Sanchez-Fernandez et al. [53] reported that the regeneration energy of DECAB and DECAB Plus are 3.3 and 2.5 MJ/kg CO₂, which are approximately 15 to 40 % lower than MEA 30 %-wt.

4.2 Alkali Salts

Recently, the use of high concentration potassium carbonates for post-combustion CO₂ capture has been investigated. At this concentration range (30-40 %), the solubility limit is exceeded hence bicarbonate precipitates are formed [31]. The precipitating alkali carbonate systems have the same advantages as those of aqueous alkali carbonates, such as low heat of reaction, low solvent cost, negligible solvent loss, and a possibility to exclude FGD and SCR [31].

The performance of "UNO Mk 3", a proprietary precipitating potassium carbonate solvent developed by the Cooperative Research Centre for Greenhouse Gas Technologies (CO2CRC), has been reported by [31]. The solution contains potassium carbonate ranging from 35 to 50 %-wt. The CO₂ loading in the solution was reported to be between 0.3 to 0.68 and the regeneration energy of this system was reported to be 3.6 MJ/kg CO₂ [31]. UNO Mk 3 has been evaluated in a pilot plant at The University of Melbourne and at the Hazelwood Power Station [55-56].

4.3 Other Phase-Change Solvents

One example of a carbonate-based solvent is the chilled ammonia process (CAP), which is licensed by Alstom and was installed on a plant at Mountaineer to capture 100,000 tons of CO₂ per year. This project was a co-operation with American Electric Power [57]. Key advantages of CAP include low regeneration energy, good resistance to solvent degradation, high desorption pressure and high solvent working capacity compared to MEA [33, 58-60]. The main drawbacks of this system are the need for refrigeration to support the low-temperature absorption process and the high solvent volatility. An ammonia washing system is typically installed to recover vaporized ammonia at the outlet of the absorber, which contributes to the capital costs of the CAP system. It has also been reported that CAP has a low absorption rate, about 1.5 to 2 times lower than MEA [61]. There are five sections in the CO₂ capture system using CAP: flue gas cooling, CO₂ absorption into solution, NH₃ stripping from the vent gas, CO₂ regeneration, and CO₂ compression [12].

5 Encapsulated Solvent

Encapsulated solvents are one of the new processes currently under development for CO₂ capture [62-65]. In the encapsulated solvent system, the solvent fluid is enclosed in a thin membrane capsule typically 100-600 μm in diameter [65]. The capsule shell can be made from polymers with typical wall thickness of 10-30 μm [65]. The working solvent is isolated within the capsule while the gas is absorbed through the capsule wall. Thus, the membrane capsule needs to be thin enough in order to prevent excessive mass transfer resistance, but also strong enough to maintain its integrity and stability.

While any class of solvents can be used within the encapsulated solvent system, for phase change solvents, encapsulated solvent systems can overcome some of the processing challenges of the traditional process design. If conventional absorption equipment such as a packed column is used, the precipitation of solids in the absorption process raises concerns of plugging [49]. However, if CO₂ absorption is operated in a spray tower to prevent plugging, the capital cost for the system may be uneconomic. The use of encapsulated solvents overcomes this as the precipitates are isolated in the capsule, thus preventing plugging. The other advantage of an encapsulated solvent is that it has significantly higher surface areas compared to conventional packing [63, 65]. Compared to a conventional absorption system, an encapsulated solvent system is likely to possess a drawback in performing heat recovery between the rich and lean sorbent streams. As observed in a solid adsorbent system, exchanging the heat between solid streams is not as straightforward as exchanging heat between liquid streams [66-68].

At the current stage of encapsulated solvent development, the optimal capsule properties such as the capsule diameter, capsule thickness, membrane capsule permeability, and so forth, have not been formulated.

Different process configurations including fixed-bed circulated fluidized bed, and bubbling fluidized bed columns have been proposed as possible equipment for use as the absorber and the regenerator [69]. However, this has not been examined in detail.

6 Discussion

Aqueous Solvents

Raksajati et al. [70] discussed the parameters that have the largest effect on the CO₂ capture cost by using aqueous solvent. The authors reported that solvent stability to SO_x and NO_x, solvent working capacity, solvent concentration, and heat of reaction are the most important variables that affect the CO₂ capture cost significantly. On the other hand, the solvent loss had a relatively small impact on the CO₂ capture cost. Furthermore, in terms of process design improvement, it was found that reduction of capture cost could be achieved by employing novel heat transfer configuration to reduce the temperature difference across stripper, a high solvent regenerator pressure system, advanced column packing with high surface area, and alternate cheaper materials for process vessels.

The costs estimation of CO₂ capture by using aqueous solvent was estimated to be in the range of US\$62-US\$80 per metric ton of CO₂ avoided and US\$44-US\$59 per metric ton of CO₂ avoided for capture plant with and without FGD and SCR, respectively [70]. Based on the parameters that are important for aqueous processes, it was reported that capture cost can be reduced significantly for solvents that have good stability to SO_x and NO_x, high working capacity (> 0.35 moles of CO₂/mole of solvent), high concentration (> 50 wt. %), and fast (comparable absorption rate with MEA). This result may suggest that future generations of CO₂ capture processes may be likely to involve a phase change during CO₂ absorption that have higher solvent capacity.

To date, most of the developments for aqueous solvents have focused on amines. However, this presents a difficulty if developments continue to be based on amines, as the degradation of amines due to acid gas impurities will make the possibility of excluding the FGD and SCR very challenging. Continued development of amine solvents would require a novel approach, for example integrating the CO₂ and acid gas impurities removal, such that has been applied in the Cansolv system in Boundary Dam Project [71]. Alternatively, future development of aqueous solvents could focus on developing non-amine solvents [34].

Phase-Change Solvents

Raksajati et al. [48, 72] studied the techno-economic analysis of the carbon capture using phase-change solvents. The authors estimated that the capture costs using phase-change solvents were US\$52–108 per tonne CO₂ avoided if the absorber can be operated in a packed column, in comparison to the cost of MEA 30% wt solvent at US\$88 per tonne CO₂ avoided. Furthermore, in terms of process design improvement, the use of the additional solid-liquid separator increases the capital

costs. However, the separator may reduce the heat duty for the solvents, due to a lower heat of absorption duty and water vaporization duty, or the lower sensible heat duty and water vaporization duty in the stripper [48].

For phase-change solvents, potassium carbonate, ammonia and potassium taurate are at the most advanced stage of development within this solvent class, being at various stages of demonstration field. Potassium carbonate is the main compound used in UNO Technology's UNO MK 3 solvent processes [31], ammonia is the compound used in Alstom's Chilled Ammonia Process (CAP) [58], and potassium taurate is one of the amino acid salt solvents under development by Siemens Energy and TNO [49, 73].

The carbonate phase-change solvents currently tested at demonstration stage do not require FGD and SCR pre-treatment. This is because the SO_x and NO_x are neutralised by alkali hydroxides resulting in sulphate and nitrate salts. For potassium taurate, the behavior of the solvent with SO_x and NO_x has not been reported in published studies. For phase-change solvents, one of the challenges identified is the need to improve absorber designs. This is because current process technologies require the use of spray towers to avoid plugging. During the pilot testing of the Chilled Ammonia Process (CAP) by Alstom power, it was revealed that managing solids in a packed column was unfeasible because of equipment clogging resulting in difficulties in plant operation and control [46]. Therefore, the use of a novel absorber or packing design to allow precipitation during absorption and regeneration is needed to advance the development of phase-change solvents, such as the proposal by CSIRO for their ammonia solvents [34]. Another important process improvement required for this solvent class is the use of internal heat recovery [48].

Encapsulated Solvents

Encapsulated solvents are still in the early development phase and have only been demonstrated at lab scale. Sodium carbonate was identified as an excellent solvent in an encapsulated solvent system following an evaluation of a broad range of solvents by Stolaroff and Bourcier [64] and Vericella et al. [65]. To date, there is no published study that reports the solvent degradation for an encapsulated solvent system, which needs to be investigated in future research. There has also been minimal research of process and equipment designs for encapsulated solvent systems. It is also crucial to improve the current process and equipment designs, especially by developing a novel CFB absorber, such as a multistage counter-current CFB absorber, in order to enhance the equilibrium conditions in the absorber.

Raksajati et al. [74] presented the techno-economic assessment of encapsulated solvent for CO₂ capture using two configurations: (1) multiple fixed bed columns and (2) a circulating fluidized-bed absorber and a bubbling fluidized-bed regenerator. The authors reported that the capture costs using this process were 1.6-2 times more expensive than the conventional MEA solvent system, which is due to the extra membrane resistance in the encapsulated system which increases the regeneration energy required, the higher equipment cost,

and the higher capital cost. Further improvement of encapsulated solvent may involve the use of a suitable heat recovery scheme within the process, the use of novel absorber and/or regenerator column designs, the use of solvents encased in very thin capsules, and the use of new solvents with more favourable properties [74].

7 Conclusion

This paper reviews the development and the key parameters involved in the design improvement of several chemical absorption process systems. These processes include chemical absorption using aqueous solvents, phase-change solvents, and encapsulated solvents systems. In addition, some guidelines for the future development has also been presented.

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