



Carbon capture by absorption – Path covered and ahead



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ABSTRACT

Global warming and associated climate change has resulted in serious efforts towards reducing greenhouse gas emissions, primarily carbon dioxide through carbon capture. There are various technical options in pre- and post-combustion modes available viz., adsorption, absorption, membrane separation, chemical looping combustion with and without oxygen uncoupling and cryogenic separations. Among all these, absorption technology which could be deployed as a post-combustion option to be integrated with power plant, has been commercialized with amines as solvents long back. But there is a long way to go to improve this process in terms of economic viability due to large regeneration costs involved and to make it more environmental friendly with minimum toxic and corrosion problems due to solvents. In this review, absorption based carbon capture has been reviewed comprehensively and critically in terms of various aspects like solvents and their synthesis protocols, performance analysis of different solvent systems, contactors, kinetics and thermodynamics, modeling and simulation studies etc. Every section has been discussed in terms of the trends and developments observed including the contemporary status besides citing future challenges and prospects to improve the technology.

1. Introduction

The impact of global warming due to greenhouse gas emissions, primarily carbon dioxide is a serious concern all over the world. Efforts are being made continuously to limit the degree of future climate changes due to the increase in greenhouse gas emissions. The rise in temperature would eventually lead to the rise in sea level, if not controlled, increase the probability of occurrence of storms and floods. The International Panel of Climate Change (IPCC) estimates that by the year 2100, the CO₂ content in the atmosphere shall reach 570 ppmv, the sea level will increase by 3.8 m and the global mean temperature will rise by 2 °C with major consequences on the environment [1–4]. With the continuous rise in world population, demand for energy was expected to shoot up. Burning of fossil fuels (coal, petroleum and natural gas) continues to cater to the energy demand which accounts for massive CO₂ emissions into the atmosphere, thus leading to significant increase in global warming. In order to reduce this emissions significantly, carbon capture (CC) has been identified as the key solution and hence under the focus of scientists, technologists and environmentalists worldwide [5].

The three major modes employed in CC were pre-combustion, oxyfuel combustion (OFC) and post combustion. Pre-combustion was

preferred at relatively higher pressures and concentrations of CO₂. Post combustion options were expensive but could be used for low concentrations of CO₂ and could be integrated with the existing power plants for effective CC. A comparative study between pre- and post-combustion technologies was done in integration with a coal combustion plant [6]. To achieve CC efficiency of 90%, L/G ratio was found as 2.55 mol solvent/mol syngas and 1.13 mol solvent/mol syngas, solvent regeneration heat was found to be 2.18 GJ/tCO₂ and 2.8 GJ/tCO₂ for pre- and post-combustion respectively. Oxy fuel combustion involves blowing pure oxygen over the fuel for complete combustion and producing relatively pure steam and CO₂. Out of the above three modes, post combustion carbon capture was widely used. The different technologies of post-combustion CC were absorption (chemical and physical), adsorption, cryogenic separation, membrane separation and chemical looping combustion.

In adsorption, liquid or gas adheres onto solid adsorbents which could later be regenerated by the application of temperature, vacuum or pressure swing operations. Activated carbon, metal oxides, alumina, phosphates and zeolites [7–11], metal organic frameworks [12–15], micro-porous carbons and polymers [16–20] and modified amines [21–23] were some of the different adsorbents reported to be used in this process. Membrane separation of carbon dioxide from flue gases

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depends on the difference in selectivity and permeability. Some of the membranes like organics consisting of polymers and inorganics consisting of zeolite, ceramic, metals, carbon etc. were used for the separation. It also acts as a contacting unit for the solvent and the gases which move in counter-current fashion and CO₂ gets absorbed selectively on the matrix of the membrane [8,24,25]. Oxyfuel combustion (OFC) is the latest technology which has not yet been commercialized on large scale. Pure oxygen ensures minimal production of undesirable gases like NO_x, resulting in the increased selectivity for CO₂. Cryogenic separation involves compression and cooling of the gas mixture with CO₂ at various levels to advance phase change of CO₂ along with other constituents which could later be separated by distillation [26]. This process wouldn't require chemical absorbents; could be carried out at atmospheric pressure and was compatible with both the pre- and post-combustion modes of CC. The major drawbacks of this technology include the drop of heat transfer rate due to the built-up of thick CO₂ layers, plugging caused due to the solidification of CO₂ and ice formation [25,27,28]. In CLC, the oxygen required for combustion was obtained from metal oxides to avoid direct contact between fuel and air. No additional separation step was required in this process and formation of NO_x was also minimum, leading to enhanced CC efficiency. But it is yet to be commercialized to the industry level [26]. Combining oxy-combustion with post-combustion CC could also potentially reduce the overall energy consumption by 25% when compared to an oxy-combustion process [29].

Absorption CC, the main focus of this paper is a technical option that could be used in both pre- and post-combustion modes of CC and this technology has been commercialized too. Absorption could be both physical and chemical. Physical absorption depends on solubility of CO₂ in the solvent and was preferred at higher pressures [30]. Solvents like rectisol, selexol etc. have been reported to be used as physical absorbents [27]. However the concentration of CO₂ in flue gases from coal fired power plants was reported to be upto 15% [31–34] at low pressure conditions, hence, chemical absorption was preferred owing to higher absorption capacity at low CO₂ partial pressure [35,36]. A typical chemical absorption system consists of three components i.e. solvent, absorber and stripper. Flue gases from different CO₂ sources like coal power plants come under contact with lean solution in the counter-current manner in the absorber. CO₂ was then absorbed by the solvents, leading to a lower CO₂ concentration in the flue gas. The solvent rich CO₂ was then regenerated in the stripper. The regenerated lean solution was returned back to the absorber and compressed CO₂ was collected at the top of the stripper and transported. Chemical absorption has been the most successful technology that has been commercialized from many years but it is yet to be scaled up in CO₂ recovery in power plants. It could be easily fitted with the existing power plants. However, less CO₂ loading capacity, elevated equipment size and corrosion rate, energy penalties during absorbent regeneration were few major drawbacks of chemical absorption which could be resolved by improvement in absorbents and operations involved in the process [37]. Use of aqueous alkanolamines was the most commercially successful and widely practiced technique in chemical absorption based CC [38]. Monoethanolamine (MEA) was one of the earliest alkanolamines used for CC. It has a high reaction rate, good absorption capacity and was cheap. However some of the major drawbacks were high energy penalties for regeneration of solvent, degradation in oxidizing environment, and corrosive effects [7–11,39–43]. Other alkanolamines such as diethanolamine (DEA) have also been used for absorption but they suffered from similar limitations. Researchers have also proposed the use of hybrid solvents like methanol with DEA or MEA [44,45] and N-methyl-2-pyrrolidone (NMP) with diethylene glycol (DEG) and MEA [46]. These could perform better than the single solvents due to their enhanced diffusivity and solubility. Combination of MEA and DEA has been reported to give some success in improving carbon loading capacity, withstanding corrosion and reduction in energy penalties during regeneration [11,43,47–51]. The use of sterically-

hindered amines like 2-amino-2-methyl-1-propanol (AMP) were also reported in literature because of good absorption capacity, degradation and corrosion resistance, lower energy penalties and higher selectivity [43,52–55]. Combinations of AMP with DEA and DEA with MDEA were also reported [56,57].

Piperazine (PZ)/cyclic diamines were used because of good absorption capacity, higher resistance towards corrosion and degradation and lower regeneration costs [26]. Dash et al. [58] reported that 90% CC could be achieved using 22 wt% AMP and 8 wt% PZ [58–62]. Use of aqueous potassium carbonate (K₂CO₃) promoted with inorganic salts like arsenates, borates, silicates and vandates; organics like amines and their derivatives, alkaline amino acids like arginine and biological enzymes like carbonic anhydrase were also described as effective absorbents due to reduced enthalpy, degradation and cost requirements [63–72]. K₂CO₃ with MgO as support was reported to attain a 99.4% efficiency of CO₂ capture [73]. Ionic liquids integrated with functional groups like amino acids exhibited good CO₂ absorption ability, lower energy penalties for regeneration, higher selectivity and higher thermal and chemical stabilities [74–76]. The use of ammonia as solvent for CC has been reported to have higher rate of reaction with lower regeneration energy than traditional MEA solvent [77–79].

To enhance CC with minimal energy penalty, extensive research has been done and reported for the development of efficient gas-liquid contactor configurations, solvent systems and stripper configurations [26]. The central idea was to maximize the surface area and mass transfer for the processes of absorption and desorption. Different absorber configurations like packed bed (PB), bubble column, spray column, rotating packed bed (RPB) and tray towers were employed. Some of the retrofit options like use of multiple columns, vapor recompression and heat integration in stripping section, use of split flows and matrix stripping were reported to improve the efficiency [80–87].

In this review article, the importance of carbon capture and the various technical options available in achieving the same so as to mitigate a very grave concern of global warming have been initially discussed in the preceding the introduction section. In the sections that follow, the developmental trends on following aspects related to absorption based carbon capture were discussed thoroughly besides citing future challenges and prospects in each of them. Every section has been summarized by a table that captures the overall findings.

- Solvents-classification, synthesis protocols and the comparison of their performance in CC.
- Promoters employed along with solvents to enhance their performance.
- Contactors employed in absorption based CC-type, scale, critical design parameters, merits and demerits of each of them.
- Modeling studies done to analyze, validate and predict the technology under more rigorous conditions. Various models were discussed in terms of their governing equations, assumptions, performance, advantages and limitations.
- Thermodynamic and kinetic aspects of the process employing various solvent systems were presented and discussed as these were very vital in contactor design and scale-up.

2. Absorbents and their performance

2.1. Absorbents and their classification

An absorbent employed in the CC process could be classified as two types: (a) *chemical* where chemical reactions occur to separate carbon dioxide and (b) *physical* where separation occurs by physical methods with mass transfer occurring at the gas-liquid interface. It depends on the solubility of gases, surface area of the interface and operating conditions like temperature, pressure. Fig. 1 highlights the classes of absorbents used for CC. The chemical or reactive absorbents involve

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