



Advances in CO₂ Capture using Absorption and Adsorption Technology: A Review

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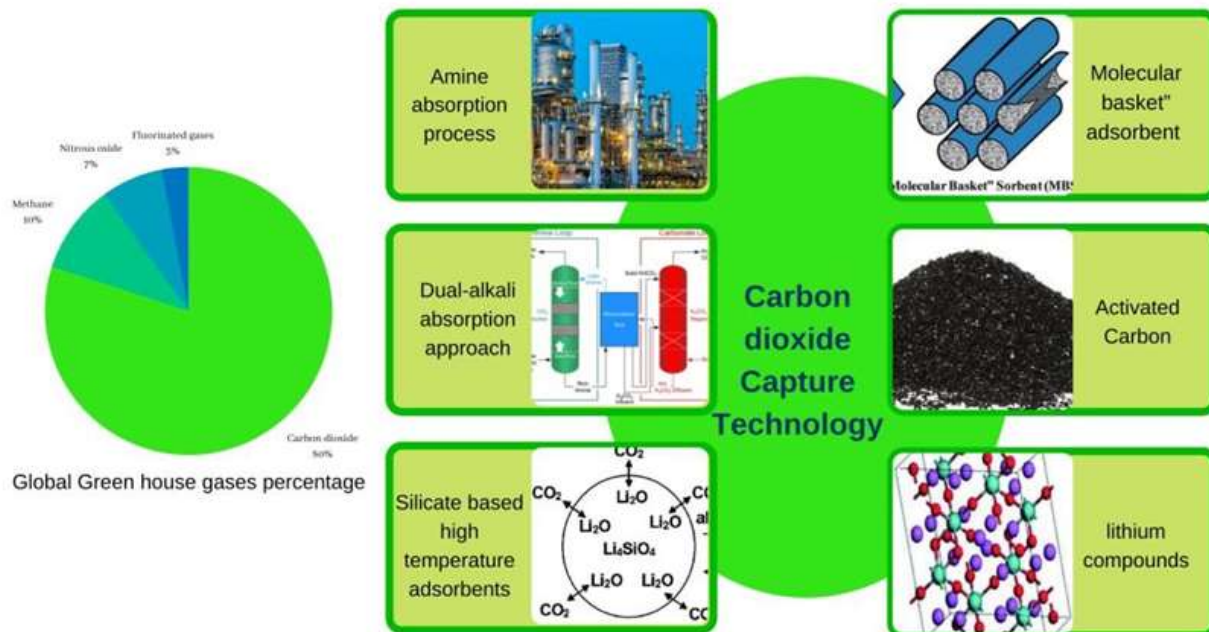
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ABSTRACT: The CO₂ capture from flue gas which is emitted from the power plants, is one of the important way to reduce greenhouse gases. The flue gas from coal-fired power stations can be removed by Absorption process using solvents and by Adsorption processes using solid sorbents. This article reviews the progress made in CO₂ separation completely. A variety of promising sorbents such amine, ammonium, activated carbonaceous materials, microporous/ mesoporous lithium based silicates, carbonates for the removal of CO₂ from the flue gas streams have been reviewed. A wide ranging review and analysis of the literature of CO₂ has been carried out to update the recent progress. A comparison of different sorbents efficiency is made with current research efforts to progress of carbon capture.

KEYWORDS: CO₂ capture, Amine absorption, Activated carbon, Li based adsorbents, Ammonium absorption.

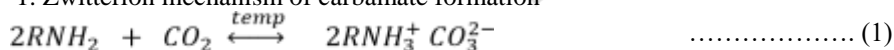
GRAPHICAL ABSTRACT:



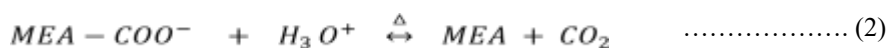
I. INTRODUCTION

The rapid population growth leads to rise the industrialization and utilization of fuels. Due to increasing fuel usage, the amount of CO₂ liberated into the environment also upsurge (Kim et al., 2008). The elevated energy demands have begun the severe rise in fossil fuel (coal, petroleum, and natural gas) combustion. This huge utilization of fuel liberated approximately 45% of CO₂ and signifies over half of the global greenhouse gas emission. It causes the sharp increase of CO₂ in the atmosphere by 25 % over the past century (Sattari, Ramazani, Aghahosseini, & Aroua, 2021) (Carapellucci & Milazzo, 2003). The CO₂ buildup in the atmosphere is one of the significant global greenhouse which is the reason for global warming, acidic rain, sea level raising and also causes for climate transformation. This changes leads droughts, floods and ecosystem destruction including species extinction, global and regional food insecurity, constraints on human activities followed by adverse influences on future generations (Kim et al., 2008). Generally, we have three ways to reduce total CO₂ emission into the atmosphere, (i) reduce the high energy utilization, (ii) reduce the carbon intensity, (iii) enhance the sequestration of CO₂ present in the atmosphere. The first way can be achieved by using the energy efficiently. The second one, requires switching to using non-fossil fuels such as hydrogen and other renewable energy sources. The third way can done by develop the technologies to capture and sequester more CO₂ (Fauth, Frommell, Hoffman, Reasbeck, & Pennline, 2005). To enhance the capturing of CO₂, the designed process should be accelerate to the fixation of carbon in lithosphere and biosphere. The CO₂ fixation can be carried out by the enhancement of natural sinking process, such as forestation, ocean fertilization and mineral carbonation, artificial CO₂ sequestration, such as injection into geological formations and ocean (H. Yang et al., 2008). The selection of efficient CO₂ sequestering technology is depends numerous parameters such as pressure and temperature of CO₂ resources, partial concentration of CO₂, concentrations of impurities and ecological aspects of system (Yu, Huang, & Tan, 2012). This paper mainly reviews recent progress of CO₂ separation and capture from a gas mixture from the chemical conversion point of view. Absorption technology using solvent such as amine, ammonium and dual alkali process of CO₂ capture has been discussed. Moreover, Adsorption process using adsorbents like silicates, molecular basket, activated carbon and Li based salts has been discussed with removal efficiency of CO₂.

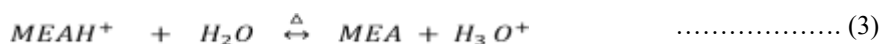
1. Zwitterion mechanism of carbamate formation



2. Zwitterion mechanism of carbamate breakdown with protons:



3. Amine H⁺ deprotonation process:



However, this benefit is dependent on whether the chemical stability of the solvent can be maintained (Idem et al., 2006). Even MEA have many advantages, it has some disadvantages also. The loading capacity of carbon dioxide by MEA process is low (Idem et al., 2006). It's expensive due to high equipment corrosion rate and high energy consumption during absorbent regeneration. The amine degradation takes place by SO₂, NO₂, HCl, HF, and oxygen in flue gas which induce a high absorbent makeup rate (Fauth et al., 2005; Resnik, Yeh, & Pennline, 2004).

II. AMINE ABSORPTION PROCESS

The MEA (Monoethanolamine) absorption process is one of the widely studied CO₂ absorption systems in industry. Recently, various aspects of MEA absorption have been studied, such as modifying the absorbents, improving the process design or optimizing the process operation (Samanta, Zhao, Shimizu, Sarkar, & Gupta, 2012; Chunfeng Song et al., 2017). Largely, Natural gas producing Industries uses MEA to absorb CO₂. Commercial MEA absorption processes have implemented to remove CO₂ from combustion flue gas stream (Gholidoust, Atkinson, & Hashisho, 2017; Oyenekan & Rochelle, 2006; Rahimi, Riahi, & Abbasi, 2020). Recently, amine impregnated carbon and nano particles showed better adsorption nature of CO₂ from flue gas (Gholidoust et al., 2017; Goff & Rochelle, 2006; Rahimi et al., 2020). The MEA in solution reacts with CO₂ in the gas stream to form MEA carbamate and protonated amine. Apart from MEA, diethanolamine (DEA) and methyl diethanolamine (MDEA) are often used as absorbents. The majority of the CO₂ captured by this mechanism will result in the formation of bicarbonate in the liquid amine capture system. In aqueous medium, there is a requirement of two mol-amine with one mol CO₂ for the formation of stable bicarbonate compounds resulting in the capture of CO₂. This process is denoted in equation (1). The CO₂-rich MEA solution can be regenerated with liberation of pure CO₂. It is represented in the following equations (2) & (3) and both the processes are endothermic (Shi, Naami, Idem, & Tontiwachwuthikul, 2014) (Stewart and Hessami, 2005). This process is generally expensive as it requires large equipment size, intensive energy input and endothermic process. The large heat duty for solvent regeneration constitutes up to 70% of the total operating costs in a CO₂ capture plant (Nguyen & Allinson, 2002) (Shi et al., 2014).

Moreover, mixed amines have been shown to maximize the desirable qualities of the individual amines. The mixed amines solutions have low regeneration costs when compare to that of individual tertiary amines. There are reports of substantial reduction in energy requirements, and modest reduction in circulation rates for amine blends on the corresponding single amine system of similar total amine concentration (R. Wang et al., 2021). A large heat-duty reduction can be achieved by using a mixed MEA/MDEA solution in its place of a single MEA solution in an industrial environment of a CO₂ capture plant.

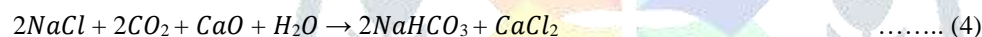
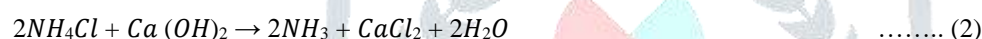
III. AMMONIUM ABSORPTION PROCESS

Aqueous ammonia is used as good carbon dioxide sorbent in industries and it has the huge capacity of multi-component control. In this absorption process flue gas need to preprocess since to avoid oxidizing sulfur dioxide and nitric oxide to form sulfur trioxide and nitrogen oxide respectively (Gholidoust et al., 2017; Kim et al., 2008; Lamas Galdo, Rodriguez García, & Rebolledo Lorenzo, 2021; Resnik et al., 2004). Generally, the flue gas reacts with aqueous ammonia in a wet scrubber and it converted in to ammonium bicarbonate and ammonium carbonate. So it requires huge

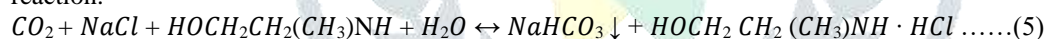
heat energy for the regeneration of ammonium from enriched yield. This process produces major by-products such as ammonium sulfate, ammonium nitrate, ammonium bicarbonate. This ammonium sulfate and ammonium nitrate are best known fertilizers which can be used but ammonium bicarbonate is unreliable so we can thermally decompose it to recycle ammonium (Kim et al., 2008). When compared to MEA process this ammonium absorption method saved energy up to 60 percent and MEA showed 1.5- 2 times greater efficiency of CO₂ absorption (Lamas Galdo et al., 2021; Puxty, Rowland, & Attalla, 2010).

IV. DUAL-ALKALI ABSORPTION APPROACH

Recently, the dual alkali solvent (DAS) system has been employed for CO₂ capture and separation systems. Unlike traditional solvent-based CO₂ capture systems, DAS system consists of two aqueous phases and the same solvent is used for both CO₂ absorption and stripping. The concept of the DAS technology has brought a new outlook to solve the challenges of the solventbased CO₂ capture technologies, and made it possible to largely reduce the CO₂ capture cost (Li et al., 2017). Huang, et.al has been suggested an alternative approach for the absorption of CO₂. According to his proposal the exhausted CO₂ could be converted into carbonate salts that can be safely returned to the environment. The natural method of CO₂ sequestration is weathering of alkaline rocks. The CO₂ absorption by rock weathering is one of the most promising measures for CO₂ problem from the present experimental and



Methylaminoethanol (MAE) is opted as the effective primary alkali in the mentioned study by the following reaction:



CO₂ absorption of MEA (0.5 mol CO₂ /mol-MEA) is lesser than that of MAE (0.75 mol CO₂ /mol-MAE). The maximum CO₂ absorption capacity of an amine is 1.0 mol-CO₂ /mol-amine if the reaction product is bicarbonate and 0.5 mol-CO₂ if the reaction product is carbamate. To regenerate the primary alkali MAE the group of researchers had not identified the secondary alkali (Näfe, 2014).

V. SILICATE BASED HIGH TEMPERATURE ADSORBENTS:

The specially designed molecular sieves/ molecular basket of silicates based solid adsorbents shows high removal of CO₂ at high temperature. Due to high porosity and surface area. By chemically treated the molecular sieve surface improves the CO₂ adsorption. Many research activities aiming to improve the efficiency of CO₂ adsorption on silicates based adsorbents. The mesoporous Si-MCM-41 was functionalized with loadings of tetraethylenepentamine (TEPA) improves the performance of CO₂ adsorption. The capacity of adsorption increases with increasing loading of TEPA. The TEPA-functionalized Si-MCM-41 shows the CO₂ adsorption capacity of 54.65 mg/g at 25 °C and 1 bar. The adsorption capacity of 50 wt % TEPA-Si-MCM-41 increases with increasing temperature from 25 to 75 °C with the highest CO₂. The maximum adsorption capacity of 70.41 mg/g reported at 75 °C and 1 bar (Ahmed, Ramli, Yusup, & Farooq, 2017). The Mesoporous silica substrates which is

energy evaluation (Kojima, Nagamine, Ueno, & Uemiya, 1997).

In the Solvay process, ammonia utilizes as a catalyst to support the reaction of CO₂ with sodium chloride for the production of sodium carbonate. The reaction was achieved by saturating brine with ammonia, and then with carbon dioxide. The reaction of the process mentioned in equation (1). Sodium Carbonate (product) is obtained by heating sodium bicarbonate. The ammonia is retrieved by reacting ammonium chloride with lime (secondary alkali), here limestone is the source of lime. This reaction is represented in equation (2). It's evident that there are drawbacks to applying this process to capture carbon dioxide from flue gas which is the mixture of gases produced by the burning of fuel or other materials in power stations and industrial plants and is extracted via channel. The reclamation of ammonia with the use of lime stone made the process is ineffective. This is mainly because of the consumption of limestone, production of CO₂ and intensive energy needs during calcination and for the process of purification. It's seen that for every two moles of CO₂ captured, one mole of CO₂ is released from the calcination of limestone which is shown by the following reaction equation (3&4)(Huang et al., 2001):

functionalized with N-[3-(trimethoxysilyl) propyl] diethylenetriamine to form diethylenetriamine[propyl(silyl)]- (DT-) functionalized hybrid products more suitable for CO₂ adsorption. Higher temperature led to reduced adsorption capacities but higher heats of adsorption (H_{ads}) of CO₂, thought to be due to the reduced role of weak physisorption sites (Knowles, Delaney, & Chaffee, 2006). Contrasted with seethed silica upheld Polyethylenimine(PEI) sorbent, the PEI /APTES-FS sorbent can accomplish a CO₂ limit as high as 130.4 mg-CO₂ /g-sorbent, a 44% expansion from 90.4 mg-CO₂ /g-sorbent over PEI/FS. The amine productivity of PEI for CO₂ sorption was additionally improved by more than 25%. Besides, both the CO₂ sorption and desorption energy were extraordinarily improved by 1.23 and 1.61 times (X. Wang, Fujii, & Song, 2020). Fumed silica based molecular basket sorbent (MBS) showed the highest CO₂ sorption capacity due to its unique interstitial pores formed by the accumulation of fine particles (L. Zhang, Wang, Fujii, Yang, & Song, 2017). The fig 2.1 represents the schematics reaction between amines and CO₂ within an adsorbent environment. Both amine absorption and CO₂ adsorption process have same capacity 0.5 mol CO₂ / mol when it is surfacebound amine without presence of water, the capacity is 1.0 mol Carbon dioxide/ mol went it is surface-bound amine with the presence of water. Some mesoporous silica tend to be more attractive due to porosity and surface functional group and it helps in facilitating the capture of CO₂ (Kojima et al., 1997)(Y. Yang et al., 2021)(Ahmed,

Ramli, & Yusup, 2017)(Y. Zhang, Yu, Louis, & Wang, 2018).

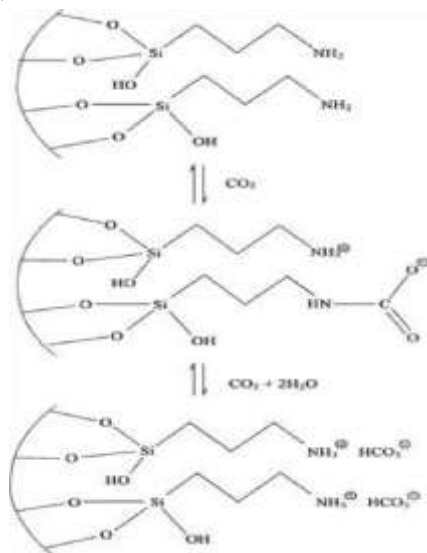


Fig 2.1 Schematic reaction between amines & CO₂

VI. “MOLECULAR BASKET” ADSORBENT FOR CO₂ SEPARATION

Molecular basket adsorbents are based on mesoporous molecular sieves that show high sorption of CO₂. Zhang et al., 2017 studied the influences of silica supported and Polyethylene glycol (PEG) additive on the adsorption efficiency of molecular basket sorbent (MBS) for CO₂ capture. The MBS consisting of polyethylenimine and with one of the following additives supports: SBA-15 (2-D structure), TUD-1 (3-D sponge-like structure) and fumed silica HS-5 (3D disordered structure). Zhang synthesized series of SiO₂-based MBS has been prepared by using SBA-15, TUD-1 and HS-5 supports which have different pore structures and pore properties. The results suggested that the pore properties of a support including 3D pore structure, pore size and pore volume may not be directly related to the CO₂ sorption performance of MBS sample, while these properties after PEI loading play a more important role in the CO₂ sorption capacity. Ahmed, Ramli, Yusup, et al., 2017 studied CO₂ adsorption performance on mesoporous Si-MCM-41 was functionalized with different loadings of tetraethylenepentamine (TEPA) using a gravimetric technique. These results showed the best CO₂ adsorption of 70.41 mg/g at 75 °C which determines the greater potential to be applied for CO₂ capture from the flue gases of power plants.

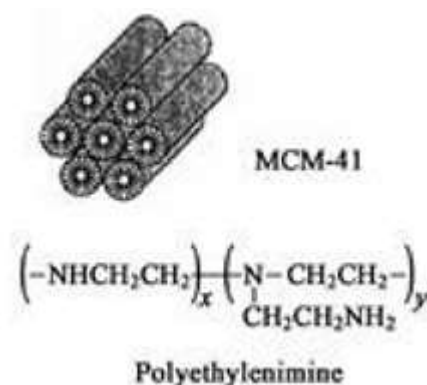


Figure 2.2. Structure of MCM-41 loaded with polyethylenimine (PEI) (Source: Chunshan Song, 2006)

The MCM-41 (mesoporous zeolite), mesoporous molecular sieve MCM-41 loaded with polyethylenimine (PEI) (Figure 2.2), Si-MCM-41 with loadings of polyethylenimine (PEI) and aminosilane-grafted mesoporous silicas were utilized as adsorbents for the capture of CO₂. These research articles results clearly demonstrated the greater efficiency of CO₂ adsorption. It confirms that molecular basket adsorbents is best suitable for power plants. (Ahmed, Ramli, & Yusup, 2017; Chunshan Song, 2006; Tiwari, Goel, Bhunia, & Bajpai, 2017; Xu, Song, Miller, & Scaroni, 2005; Yoshitake, Yokoi, & Tatsumi, 2002)

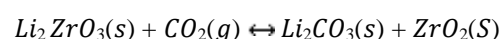
VII. ADSORPTION BY ACTIVATED CARBON

The activated carbon (AC) is low cost and has high specific surface area, high pore structures and easy regeneration. It displays AC as an excellent potential adsorbent for removal of CO₂(Boujibar, Ghamouss, Ghosh, Achak, & Chafik, 2021; He et al., 2021; Ngoy, Wagner, Riboldi, & Bolland, 2014). In addition, activated carbon can be derived from various agricultural waste materials (Saravanakumar, Muthukumar, & Selvaraju, 2019; Sud, Mahajan, & Kaur, 2008). Recently, cheap agricultural and forestry wastes such as bamboo waste scraps, coconut shell (de Oliveira Cruz, Gomes, Tonetti, & Figueiredo, 2019), seed powder of *prosopis juliflora* (Jayaram & Prasad, 2009), waste potato residue, sugarcane bagasse (Guo et al., 2020) and spent coffee have been confirmed as the potential feedstock for AC preparation. Generally, activated carbon preparation consists of two-steps. The first step is to pyrolyze the mixture of raw materials and the second step is activation processes (He et al., 2021).

Guo et al. studied the CO₂ capture behavior of AC which is derived from sugarcane bagasse. He investigated various carbon adsorbents which are derived from the NaOH activation methods. It showed outstanding static CO₂ uptake 4.28 mmol CO₂/g at 25 °C and 1 bar and great dynamic CO₂ adsorption capacity 1.31 mmol CO₂/g at 60 °C and 10% CO₂. Further, the authors conclude, the NaOH-activated carbon adsorbent has a high specific surface area (1149m²/g) and excessive pore volume (1.73 cm³/g). So it influences the performance of the adsorbent with excellent efficiency for CO₂ adsorption and separation applications. Moreover, Gholidoust et al. studied the enhancement of CO₂ adsorption via amine impregnated activated carbon from oil sands coke. The best activated carbon sample showed 5.63 mmol CO₂/g adsorbent capacity. The adsorption capacity decreases only 14% after 15 cycles of adsorption- desorption, which confirms that this activated carbon can be utilized for multi-cycle processes. Nowadays, high porous activated carbon can be synthesized by an eco-friendly process, making AC being a good potential adsorbent for CO₂ capture and separation (Selvaraju, Kartini, & Bakar, 2018).

VIII. CO₂ ADSORBENTS BASED ON LITHIUM COMPOUNDS

A satisfactory CO₂ sorption using lithium zirconate (Li₂ZrO₃) has been investigated by Peltzer, Mùnera, Cornaglia, & Strumendo, 2018. The following reaction illustrates the technology based on the chemical reaction used Li₂ZrO₃ to capture CO₂.



In the temperature range between 450 °C – 590 °C, this reaction becomes reversible. The direction can be easily reversed by a simple temperature swing approach. This adsorbent can be reversed for various cycles. The eutectic carbonate is formed such as Li_2CO_3 and K_2CO_3 , this can accelerate the CO_2 absorption reaction. There were a number of binary and ternary eutectic salt modified lithium zirconate sorbents identified and they were evaluated for higher temperature CO_2 capture noticeably. The CO_2 uptake rate and CO_2 sorption capacity improved due to the combination of binary alkali carbonate, binary alkali/alkaline earth carbonate, ternary alkali carbonate and ternary alkali carbonate/halide (Fauth et al., 2005).

Y. Zhang et al. has studied Lithium silicate (Li_4SiO_4) showed higher CO_2 adsorption capacity when compared to lithium zirconate. Lithium silicate adsorbs CO_2 below 720 °C and releases CO_2 above 720 °C by the following mechanism:



Incline features like rapid absorption, large capacity, wide range of temperature and concentration of CO_2 and good stability make this compound a strong adsorbent for developing commercially competitive CO_2 adsorbents (Izquierdo et al., 2018). Additionally, lithium-borate oxide (Li_3BO_3) also used as high capacity CO_2 adsorbent operative over the intermediate temperature range of 500 to 650 °C. This adsorbent depicted high CO_2 uptake capacity (e.g., 11.3 mmol g^{-1} at 520 °C) with excellent cyclic regenerability in the presence of alkali-metal nitrite salts as a reaction facilitator. The superior CO_2 adsorption is recognized to the dissociative formation of lithium carbonate (Li_2CO_3) and lithium borates ($Li_6B_4O_9$, $LiBO_2$ and $Li_2B_4O_7$) during the reaction of Li_3BO_3 with CO_2 (Harada & Hatton, 2017). So, the Lithium based adsorbents are considered as a capable superior for the CO_2 capturing adsorbent from power plants, flue gases, etc. But, its industrial applications are limited since the cost of raw materials are high (Y. Yang et al., 2021; Y. Zhang et al., 2018).

IX. CONCLUSIONS

The usage of fossil fuels is the primary source of CO_2 emission. The reduction in utilization of energy is also one of the ways to reduce CO_2 emissions. The CO_2 from combustion flue gas and other industrial effluents can be captured by various technical methods other than geological injecting and direct ocean dump, forestation and ocean fertilization. This study clearly depicts Amine absorption process is established way but involves many steps for high removal capacity. Similarly, Ammonium absorption process and dual alkali absorption approach also shows considerable removal of CO_2 . Adsorption is another option to capture the CO_2 while it also has some demerits such as efficiency decrease after exhausted the surface areas. Even the solid adsorbents such as Activated carbon, Li compounds and Molecular basket type adsorbents shows higher efficiency towards capture of CO_2 .

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