

Application of Different Materials for Carbon Capture

Rundong Tian*

Guanghua Cambridge International School, Shanghai 201319, China

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Abstract: Human activities have led to increasing CO₂ emissions, mainly coming from factories and plants. The increasing concentration of carbon dioxide (CO₂) in the atmospheric environment has become a severe concern of the world due to the existing and potential consequences. Different carbon capture technologies have been developed and used to remove CO₂ from the atmosphere. In addition, carbon capture can also solve certain environmental pollution problems, by directly reducing CO₂ discharge from anthropogenic sources. The research focuses on carbon capture techniques and corresponding materials (sorbents). The current situations are stated and the significance of carbon capture is enhanced, while also giving brief introductions on several sorbents. The amine method, which is conventional and the most frequently used, is discussed first. Afterwards, porous materials including zeolites, activated carbons (AC) and metal-organic frameworks (MOFs) are discussed, mainly focusing on the sorption mechanism, advantages, drawbacks and possible corresponding solutions, specific examples and outlook. These porous materials hold great potential as solid adsorbents for carbon capture, likely replacing conventional amines in the future. Further improvements and evaluations will enable porous materials to be used in industrial applications.

1 INTRODUCTION

The issue of global warming has attracted great attention since last century. Human beings have already experienced the severe aftermath of global warming and relevant climate changes, such as sea-level rising, more frequent extreme weathers, increased heat and drought and flooding. The increase in concentration of greenhouse gases like carbon dioxide (CO₂) and methane present in atmosphere contributes to global warming. The global industrial development has led to increased CO₂ emissions, mainly from anthropogenic sources such as burning of fossil fuels. For example, global CO₂ emissions reached 34.9 Gt in 2021 (Liu, 2022), leading to a dramatic increase in the concentration of CO₂ in the atmosphere. Possible solutions to the issue include replacing conventional energy sources by cleaner and more sustainable resources as well as carbon capture and storage processes. The complete treatment of carbon dioxide includes capturing CO₂ from various sources, regeneration of sorbents, storage and transport of CO₂ and finally the conversion of CO₂ into harmless or even valuable products.

There are three common carbon capture technologies. The post-combustion carbon capture belongs to the most studied and applied one, being considered the most in existing conventional power units, such as amine method. The amine method is through the use of aqueous alkanolamine absorbents. Such chemical absorption carbon capture method has been used for years in industry and still exists to be the most recognizable one. Since the absorption process involves chemical reactions, relatively large amount of heat is released, where carbamate or bicarbonate species are formed in the absorption reactions, depending on the species of alkanolamine used (Sumida, 2012). The amine method holds several advantages, such as high process efficiency. Moreover, they are commercially available and improvements on the method have been made over time. However, the existing amine methods do have various problems and limitations. Such capture process is very energy-consuming, specifically the regeneration of absorbents, leading to high energy requirements and costs. Furthermore, amine solutions tend to be corrosive against vessels, and also possess low thermal stability, shown by decomposition under heating.

Due to the energy-intensive nature of current processes, new technologies are required to reduce energy requirements. Porous materials have been studied, aiming to replace aqueous absorbents, as they have the potential to reduce the energy costs as well as increasing the efficiency in carbon capture processes. Zeolites are microporous aluminosilicate minerals. As solids, zeolites have much lower heat capacity, thereby leading to lower energy penalty for regeneration. In addition, zeolites have a well-developed structural chemistry, which could lead to optimized zeolites for carbon capture processes. Moreover, zeolites possess unified micropores, resulting in rapid adsorption at relatively low pressures. Zeolites are also relatively cheap to produce on a large scale which makes them preferred in industrial applications. However, zeolites' main limitation is the hydrophilic nature, so water vapor would compete with CO₂ for adsorption sites and the porous materials will get saturated with water vapor eventually, leading to lower adsorption capacity for CO₂ over time. They also undergo large enthalpy of adsorption, which leads to high temperature required for desorption of guest molecules. Activated carbons (AC) are porous carbon materials produced from carbonaceous source materials. Compared to zeolites, activated carbons are hydrophobic, therefore the issues from water vapor are not main concerns. They can be used to lower uptake at low pressure compared to adsorbents such as zeolites, hence they are more suitable for high-pressure applications (Sumida, 2012). Metal-organic frameworks (MOFs) have been widely studied and engineered for various applications in recent years, including carbon capture processes. The MOFs is a kind of porous coordination polymer, consists of metal clusters and organic ligands in a crystalline structure. It holds many advantages that makes it suitable for carbon capture process, including a precise control over structures that could be achieved through synthesis and shaping, high pore volumes and surface areas, tunable pore sizes by reticular chemistry, as well as post-synthetic modifications optimizing MOFs for different industrial environments of carbon capture processes (Trickett, 2017). However, challenges do exist for MOFs in various carbon capture processes, specifically their low mechanical, thermochemical and hydro-chemical stability, as well as relatively low density due to the trade-off between pore volume and density.

The following sections of this research will discuss current and prospective sorbents for carbon capture. Specifically, amine-based solvents in chemical-absorption-based carbon capture process,

zeolites, AC and MOFs in physical-adsorption-based process and outlook for porous materials in the field of CO₂ capture are given. And this research will only concentrate on the CO₂-capturing step and regeneration of sorbents, since the regeneration requirements are essential for determining the viability of specific sorbents in industrial applications

2 AMINES-BASED SOLVENTS

Aqueous amines are mainly considered for post-combustion CO₂ capturing, which is the only carbon capture technique demonstrated at full commercial scale at present. Amines are conventional solvents, and still the mostly used in power plants. Different amines, such as monoethanolamine (MEA), diethanolamine (DEA), methyl diethanolamine (MDEA) and ammonia (NH₃), present different properties, leading to different performances and energy requirements in carbon capture processes (Romeo, 2020). The chemical reaction between amines and CO₂ is mainly because the lone pair electrons of nitrogen, which act as Lewis bases, or nucleophiles. The lone pair electrons attack the partially-positive carbon atoms in CO₂. And with participation of water molecules, carbamate species are formed while bicarbonate species are formed when tertiary amines are used. Moreover, the usage of primary and secondary amines can be used to cause different kinetics of reactions, as the formation of carbamate species is generally faster than the formation of bicarbonate species. Evaluations have been made for different amines (Romeo, 2020). Primary amines appeared to possess the greatest capabilities of capture CO₂ when used singly, since they possess the highest reaction enthalpies among all aqueous amines. High reaction enthalpies lead to the favorable reaction kinetics. Hence a high degree of CO₂ purity, high efficiency and fast absorption rates of the capturing process might be achieved. However, trade-off exists between reaction enthalpies, kinetics and regeneration penalty, since high reaction enthalpies would generally lead to high regeneration temperature and energy requirements. Contrarily, secondary amines possess less energy penalty as lower regeneration temperature. Furthermore, unstable carbamates formed in the reaction would also lead to lower regeneration energy requirements since backward reaction would be favored more, and also lead to less favored absorption kinetics and lower speed of capturing.

The technology of aqueous amines is mature, and preferred for industrial applications among other

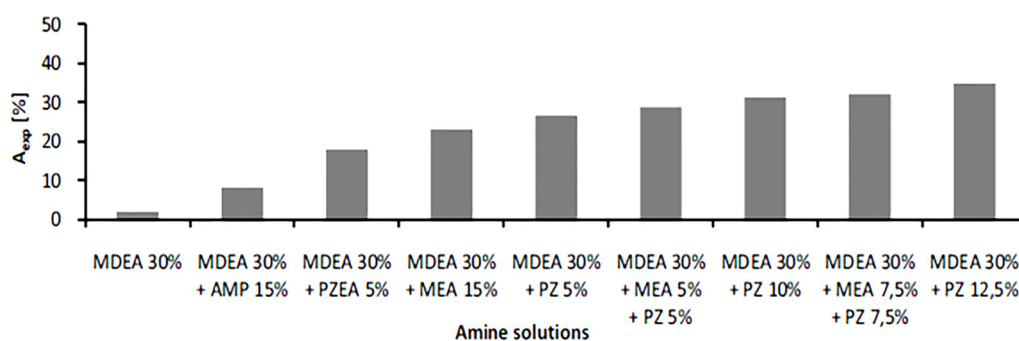


Figure 1: Experimental results of CO₂ absorption using different amine solutions (Dubois, 2011).

sorbents, due to high CO₂ affinity possessed by amines (KUROPKA, 2011). MEA, as a primary amine, holds merits of high chemical reactivity against CO₂ as well as high reaction enthalpies, leading to high absorption rates. While MDEA, as a tertiary amine, ensures the appreciable absorption capacity, therefore the combination of the two different amines leads to excellent performance of CO₂ capture. Another specific example is KM-CDR technology, which uses KS-1™ instead of MEA, achieving higher energy-efficiency (Kadono, 2013). Moreover, KS-1™ tends to be less corrosive and more resistant against O₂ degradation compared with MEA. Recent research has focused on mixing amines with other solvents, which refers to solvent blends. By combining types properly, a better absorption performance can be achieved, specifically producing solvents with both high absorption rates and absorption capacities. For instance, a higher absorption rate is achieved by mixing MEA with a little PZ (Vega, 2018). In Fig. 1, it is evidently demonstrated that CO₂ absorption increases significantly by using various solvent blends, compared to singly using 30 wt.% MDEA, due to activation phenomenon.

Although the amine method is well-developed and used in carbon capture plants worldwide, several drawbacks and challenges have always existed for aqueous amines, most significantly the energy penalty of the regeneration process. This issue is caused by various factors. The chemisorption nature and high reaction enthalpies lead to high energy requirements of backward reaction, referring to the desorption process. The amines are corrosive toward the vessels, so the concentration of amine species has to be limited. A lower concentration of amines in the solution means a larger volume of water, which has relatively high heat capacities among all liquids. A larger volume of water is required to be heated to regenerate the sorbents, causing high energy requirements and therefore severe energy penalty.

Furthermore, the solutions are unstable toward heating, therefore the temperature available for full regeneration is limited, leading to incomplete desorption of carbon dioxide from the solutions. Amines will also decompose under heating, leading to poorer performance over time and shorter life time of such sorbents.

There are several ways to deal with the drawbacks and reduce massive energy penalty, including replacing conventional amines with less corrosive and more thermally-stable ones, decreasing the stripper temperature to achieve lower corrosion rate, as well as discovering ways to increase the concentration of amines (reducing volume of water) in solution. NH₃ is considered as a potential alternative to MEA because of its low-cost, less corrosive and less likely to degrade under heat, hence greater stability compared with MEA. Moreover, it holds relatively high CO₂ absorption capacity which could lead to better process efficiency. Most excitingly, NH₃ can react with NO₂ and SO₂ in flue gas, forming ammonium sulfates and ammonium nitrates, hence not only removing impurities, but also directly producing usable and useful fertilizers. This property of NH₃ is able to greatly reduce the costs in solvent regeneration, CO₂ compression and storage. They are far less corrosive compared to conventional amines like MEA, resulting in green and sustainable chemistry. Amino-functionalized ionic liquids have been designed and absorption capacity is highly enhanced since one amine is able to bind with two carbon dioxide molecules (Luo, 2019), compared to the 1:1 or 2:1 stoichiometry of conventional amines. Overall, different aqueous amines should be examined multidimensionally before being used in CO₂ capture processes. Besides absorption capacities and rates, other key parameters include corrosiveness, resistance towards O₂ degradation, thermal stability, tolerance to impurities, recovery in regeneration, process efficiency and energy efficiency in cycle.

3 POROUS MATERIALS

Since aqueous absorbents such as amines require high energy input for regeneration, as the so-called energy penalty, sorbents with higher energy efficiency are considered for carbon capture processes. The main kind of solid adsorbents is porous materials, capturing CO₂ by either physical adsorption or chemical adsorption. As solids, porous materials have significantly lower heat capacity than aqueous absorbents which involve large volume of water. Adsorption, in contrast with absorption, refers to the enrichment of adsorbates (guest molecules) on the internal or external surface of adsorbents. Herein, three common porous materials will be discussed in the article, zeolites, AC and MOFs, though have different chemical compositions and structures, all uptake CO₂ gas molecules by physical adsorption generally. Key parameters to evaluate solid adsorbents with best CO₂ capture performance include adsorption capacities, adsorption kinetics (rates of adsorption and desorption) and selectivity towards CO₂. Moreover, since the solid adsorbents are to be used in practical applications, other industrial and engineering evaluations need to be made. Stability under specific environments and conditions is highly important, specifically towards water vapor and moisture as well as other impurities in gas mixtures. Thermal stability and mechanical stability also greatly affect the life cycle of adsorbents and their performance over time, since they might frequently undergo high temperature and high pressure. Due to the presence of pores, the density of porous materials is relatively low, which is not positive. Low density of sorbents leads to lower volumetric uptake of gas than expected, hence an optimized sorbent for industrial application should have a balanced pore volume and density. Engineering evaluations should also include the costs in various processes, such as large-scale production, transportation and regeneration of adsorbents.

3.1 Zeolites

Zeolites belongs to microporous crystalline materials that consist of silicon, aluminum and oxygen, and the porous framework can accommodate various cations, such as Na⁺, Ca²⁺ and Li⁺. Zeolites are initially explored as natural minerals, but have been studied and manufactured artificially for decades as adsorbents and catalysts. Zeolites hold a well-developed surface chemistry, as their porous structures and chemical compositions can be precisely altered, leading to different properties for

various industrial applications. Moreover, they are produced on a large scale commercially and are relatively cheap among all solid adsorbents.

Both chemical and structural factors determine the adsorption capacity to zeolites. Because of the charges on cations and the charges induced by cations within the structures, a diverse of different gas molecules can be adsorbed in different amounts. Zeolites with low and high Si/Al ratios are categorized into X and Y respectively, while different Si/Al ratios lead to different adsorption capacities (Férey, 2008). Adsorption capacities can also be increased by substitution of cations (Walton, 2006), resulting in higher charge densities and stronger attractions. Adsorption capacities of zeolites are determined by pore structures as well, larger specific surface area and pore volume will likely promote adsorption capacities. The CO₂/N₂ separation selectivity of zeolites is affected by K/Na ratio, as CO₂ and N₂ uptake might vary significantly between low and high K/Na ratios. Furthermore, selectivity towards CO₂ is also affected by pore size (pore diameter). Different pore sizes lead to different diffusion rates and selectivity. Trade-off exists between diffusion rates and selectivity, as larger pore sizes generally result in faster diffusion rates while lower selectivity towards CO₂, and vice versa.

Zeolites hold advantages including high porosity, uniform pore size and distribution, and most significantly high capacities. Good CO₂ capture performance is achieved by strong interactions and molecular sieving effect. Moreover, low Si/Al ratio is preferred due to greater aluminum content, which leads to stronger basicity, hence better CO₂ capture. Zeolites are mainly used for low-pressure adsorption due to their microporous nature which limits the uptake at high pressure, as the pores will be saturated. Zeolite 13X has been one of the most studied and synthesized zeolites, with outstanding adsorption capacities. Moreover, zeolite 13X can hold high contents of alkali- and alkali-earth cations such as sodium cations, which contributes to the strong interactions with guest molecules, enhancing the adsorption capacities as well. There have been intense researches on cation exchange, aiming to improve adsorption capacities by introducing cations with higher charge densities into the structure. Cations with smaller ionic radii and higher ionic charges are generally preferred. Kongnoo et al. further enhanced the adsorption capacity of zeolite 13X by acid activation in the preparation from palm oil mill fly ash (Kongnoo, 2017), which promoted the adsorption capacity of zeolite 13X by 22% compared with the initial (unactivated) zeolite 13X. Type 13X zeolite

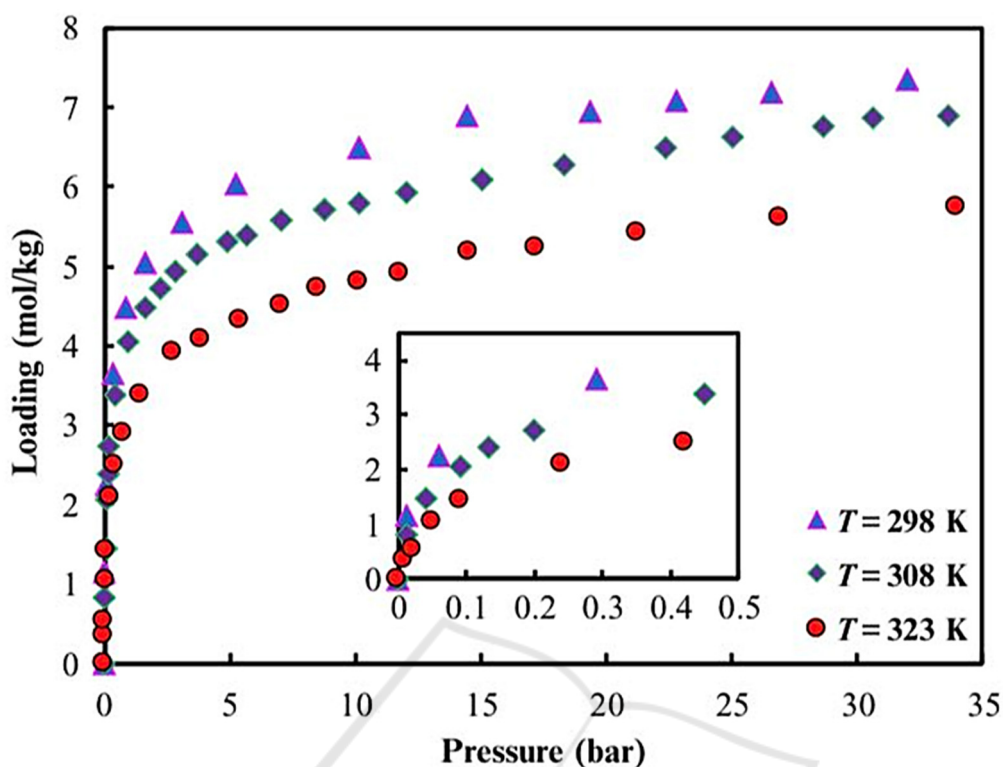


Figure 2: Adsorption isotherm of zeolite 13X for CO₂ (Cavenati, 2004).

manufactured by Ceca has gravimetric capacity of 2.05 mol/kg at temperature 298 K and pressure of 0.1 bar (Cavenati, 2004). From Fig. 2, most of the adsorption process takes place at low pressure as shown by a steep rise on the curve. Moreover, the results show that CO₂ adsorption capacity for 13X zeolite at room temperature (298 K) is significantly higher than that at elevated temperatures.

The major drawback of zeolites is their hydrophilic nature which leads to lower adsorption capacities and selectivity toward CO₂ when moisture and other impurities are present. Due to the cations and induced charges within the porous structures of zeolites, various polar molecules are favorably adsorbed, including H₂O, NO_x, SO_x and H₂S, which compete with CO₂ molecules for adsorption sites. Moreover, such characteristics also lead to regeneration penalty, as higher regeneration temperatures are required in order to remove water molecules adsorbed in the pores. Besides higher energy requirements, the recovery of capacities and other properties after desorption is also an issue, as some zeolites lose evident adsorption capacities after regeneration process under high temperature, which will generate greater costs, since the adsorbents have

to be replaced more frequently. One solution to the issue is by incorporating amine functional groups into the mesopores of zeolites. After amine grafting, interactions with CO₂ molecules tend to be chemisorption, involving chemical reactions between CO₂ molecules and amines, hence less affected by H₂O molecules. Moreover, adsorption capacity is enhanced significantly by amine impregnation, and water molecules can even promote the uptake, according to experimental data. Overall, evaluations of zeolites for industrial applications should not only be conducted under ideal CO₂/N₂ or CO₂/CH₄ gas mixtures, but also include all the substances that appear in practical working environments, such as water vapor, moisture and other impurities.

3.2 Activated Carbon

Activated carbon (AC) is another kind of conventional solid adsorbent, used for gas storage, air purification, solvent recovery and water purification.

The AC is produced from various carbonaceous source materials, such as coconut husk, bamboo, wood and coal, which then undergo physical

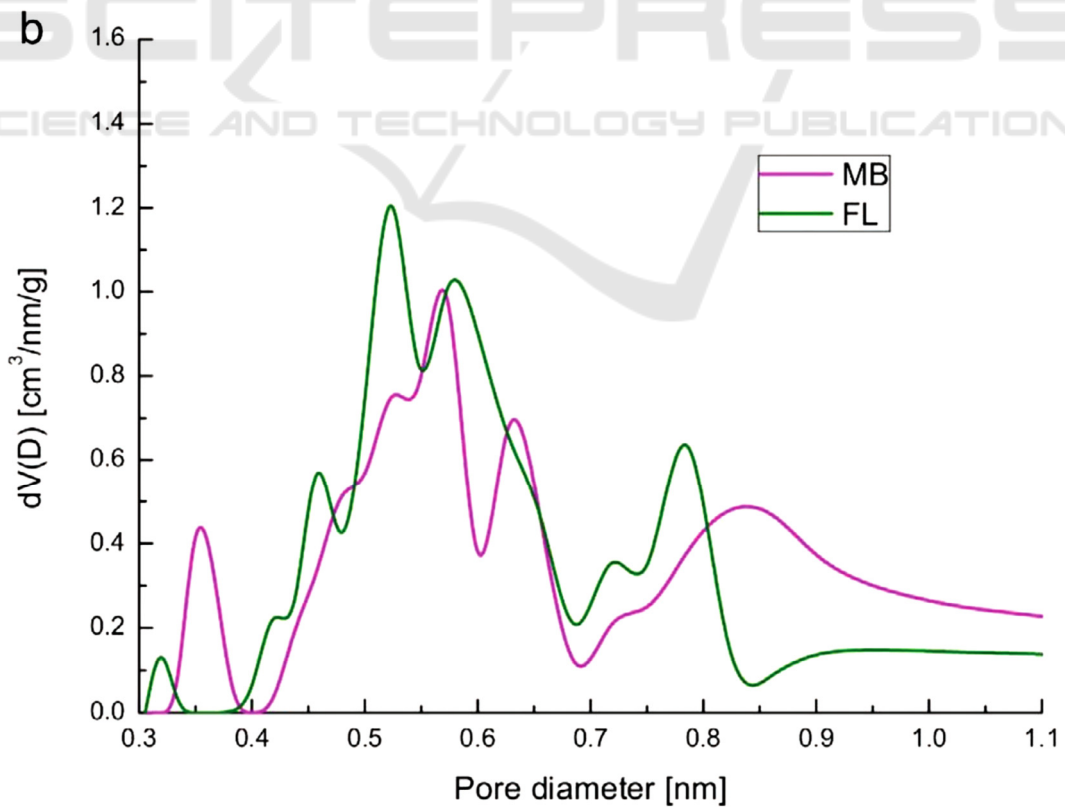
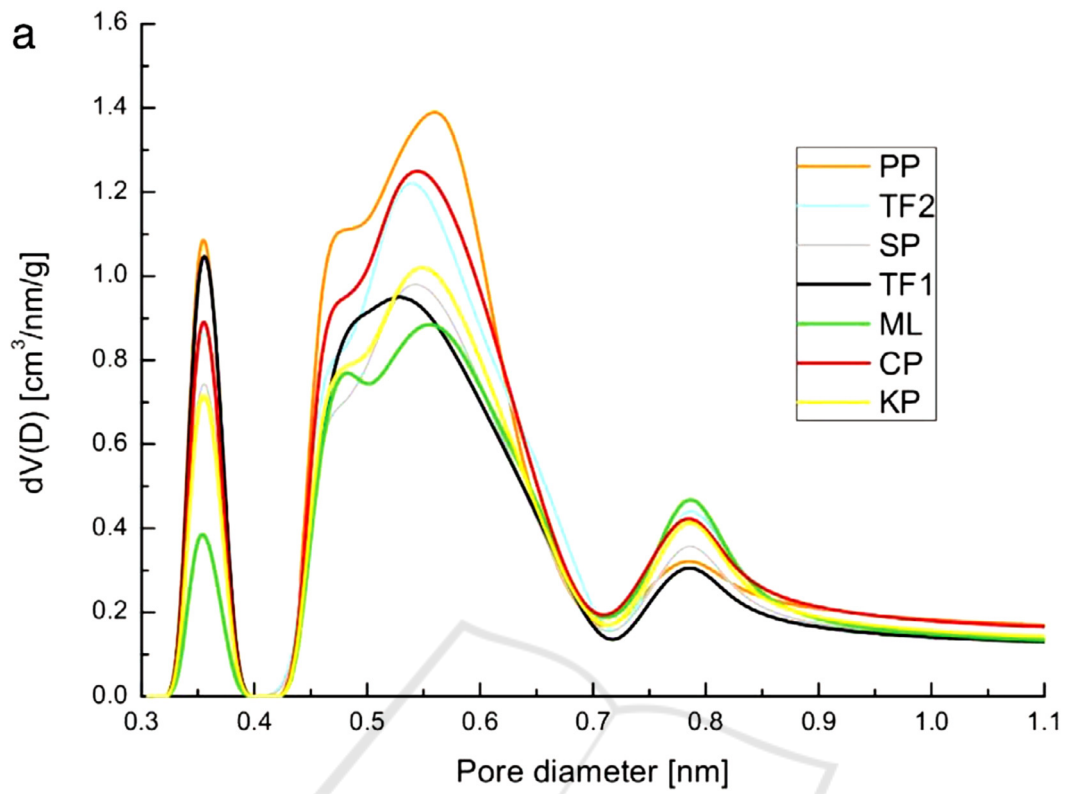


Figure 3: Different pore size distributions of the prepared AC (Serafin, 2017).

activation and/or chemical activation. Activation results in high porosity and large surface areas, with pores of different shapes and sizes. The AC uptake guest molecules by physisorption, as the interactions are caused by Van der Waal forces and induced dipoles. Textural properties, doped nitrogen contents and ultra-microporosity all affect CO₂ capture performance of AC. Pore characteristics depend on carbon precursors, preparation (activation) techniques and further modifications. Fig. 3 shows that different carbon precursors (biomass here) can be used to control different pore size distributions within AC.

The merits of using AC for carbon capture processes include high adsorption capacities, high thermal stability as well as high stability in moist conditions due to high hydrophobicity, well-developed production technologies leading to optimized porous structures and surface chemistry. Moreover, AC generally require mild conditions for regeneration because of low enthalpy of adsorption, which is caused by relatively low CO₂ affinity compared to other solid adsorbents, while also exhibiting good recovery of adsorption properties after regeneration, resulting in relatively high process efficiency. Most importantly, low costs and broad availability make AC the most frequently considered solid adsorbent in industry on a commercial scale.

Adsorption capacities and selectivity of AC can be enhanced by applying various functional groups into the porous structures. Nitrogen functionalities are the most common ones introduced into AC, achieved by heat treatment with gaseous ammonia at elevated temperatures. Experimental results show that N-doped AC demonstrate improved CO₂ affinity hence higher uptake at low pressures, but slightly lower adsorption capacity. After amination of the sample AC, adsorption capacity was significantly increased (Plaza, 2009). Greater CO₂ affinity improves selectivity towards CO₂ as well. Similar effects can also be achieved by doping sulfur atoms (oxidized-S). KOH activation is another technique to improve AC's CO₂ adsorption performance. Adsorption capacities of various ACs are significantly improved as KOH activation develops pore network, specifically micropores and mesopores. A series of chemical reactions take place, generally favored by high temperatures. After the reactions, the carbons are washed in order to remove metallic potassium and other K compounds, resulting in porous carbon lattices with high microporosity. Moreover, KOH-activated AC exhibit high recovery of CO₂ adsorption capacities after multiple adsorption/desorption cycles.

3.3 Metal-organic Frameworks

Different from zeolite and activated carbon, MOFs is a novel porous material, mostly developed in the 21st century, exhibiting exciting properties that are suitable for a broad range of applications, including chemical sensing, drug delivery, gas separations, gas storage and CO₂ capture, owing to MOFs' tunable textural properties and surface chemistry. Highly flexible modifications are available to achieve optimized MOFs for specific applications, since there is a great variety of building blocks.

The adsorption performance of MOFs can be improved by various modification techniques. For example, surface modification for MOFs will polarize the surfaces and lead to larger initial isosteric heats, as adsorption of CO₂ tends to be chemisorption. Enhanced adsorption and higher affinity lead to higher selectivity towards CO₂, as well as greater capacity at low pressures. Moreover, N₂ adsorption will decrease at all pressures due to lower surface areas as the amine functionalities occupy empty spaces, while smaller pore sizes also result in decreased CO₂ uptake in relatively high CO₂ partial pressures, hence amine grafting is more suitable for MOFs used for post-combustion CO₂ capture processes. Amine incorporation also possesses other advantages. Generally, solid adsorbents such as zeolites are concerned for their hydrophilicity as well as other impurities competing for adsorption sites. However, various amine-grafted MOFs are not affected and even showed better performance in the presence of water vapor. For instance, moisture actually promotes CO₂ adsorption capacity of TEPA-grafted Mg₂(dobdc), due to complex mechanisms. The results show that CO₂ is preferably adsorbed to the grafted amine groups under humid conditions, resulting in greater adsorption capacity (McDonald, 2015). Furthermore, amine-appended materials exhibit better CO₂ capture performance with mixture gas. Amine groups can also be incorporated onto organic ligands by click reaction, enhancing the basic property within the porous structures. Generally, amine binding with metal clusters leads to greater improvement on capacity while amine binding with ligands is more stable. Amine-grafted MOFs should undergo breakthrough experiments and dynamic cycling in order to examine recovery over adsorption/desorption cycles as well as the relationship between amine loading and amine efficiency.

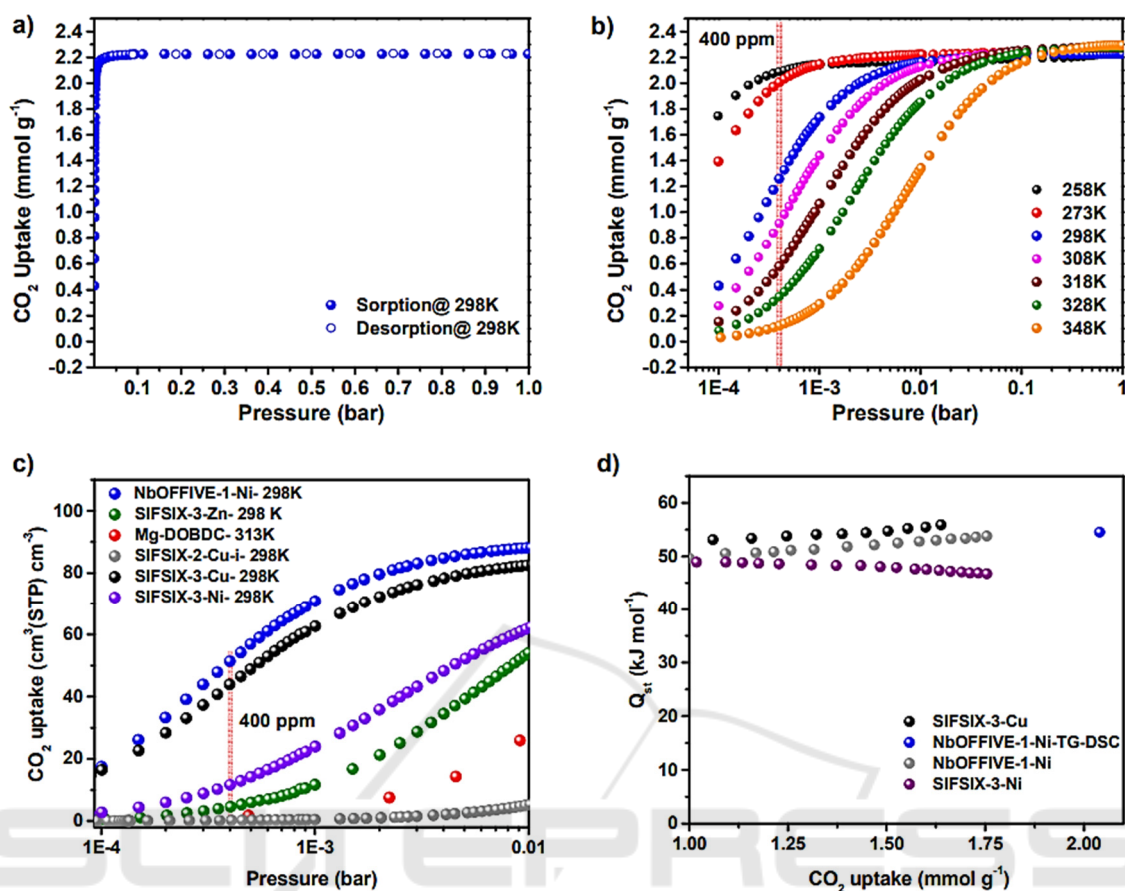


Figure 4: CO₂ adsorption isotherm, uptake and heat of adsorption for the prepared materials (Bhatt, 2016).

Adsorption performance of MOFs can also be promoted by activating open metal sites, which would lead to higher charge density and more favorable CO₂ uptake. Open metal sites are achieved either by desolvation, specifically removing solvent molecules in vacuum, or inserting metal cations into the porous structures. Due to the presence of unsaturated coordinate metal sites, CO₂ molecules transfer electrons to them, leading to stronger interactions. Pore size adjustment and distribution of adsorptive sites also play a key role in improving CO₂ adsorption. Homogenous distribution of adsorption sites is required in order to achieve identical interaction strengths, so that high selectivity towards CO₂ is maintained throughout wide pressure ranges. Favorable uptake across great pressure range leads to greater working capacity, which can lower capture costs. SIFSIX-3-Cu exhibits ultra-microporous structures and uniform distribution of adsorptive sites (Bhatt, 2016), with a 54 kJ/mol Q_{st} value leading to high CO₂ uptake at low pressures, as shown in Fig. 4. By using NbOF clusters, greater adsorption capacity (1.3 mmol/g) is achieved, because of the high charge

densities within the one-dimension microchannels accommodating CO₂ molecules, and the pore size is smaller than the kinetic diameter of CO₂ molecule, leading to slow gas diffusion and enhanced interactions.

Major drawbacks of MOFs include low mechanical stability and low density, which can be solved by monolith production and help from machine learning to make deterministic predictions for materials' performance. Monoliths are produced through the mild drying of wet gel produced from the gelation of primary particles, leading to robust, densely packed MOFs (Tian, 2018). Compared with MOF powders and pellets which contain glue or binders that take up great volume leading to poor volumetric performance and low mechanical stability, MOF monoliths exhibit great energy density as well as high stability, hence maximized performance. Machine learning constructs neural networks from database to automatically sort out MOFs fulfilling specific requirements efficiently and effectively, as the trade-off between density and pore

volume can be balanced computationally, and predictions can be further translated into applications.

4 CONCLUSION

Overall, this research discusses four carbon capture techniques and relevant materials, specifically aqueous amines for chemical absorption, zeolites, AC and MOFs, which are porous materials, for physical adsorption. The amine method is the mostly used one in industry nowadays, generally for post-combustion capture. However, more plants and factories are now considering to replace aqueous amines with porous materials. Intense researches have been conducted in recent decades, which greatly improve properties of several porous materials for carbon capture applications. More and more suitable porous materials have appeared, exhibiting optimized characteristics. If porous materials are to be used in industrial applications, engineering evaluations are required and the scaling up of production is also an issue. In the near future, mature porous materials will be able to replace present aqueous amines, which will greatly reduce regeneration costs and save energy.

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