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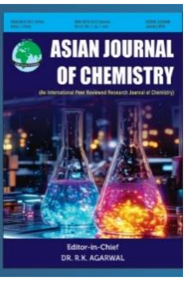

REVIEW

Recent Developments in Zeolites Based CO_2 Separation Technologies: A Review

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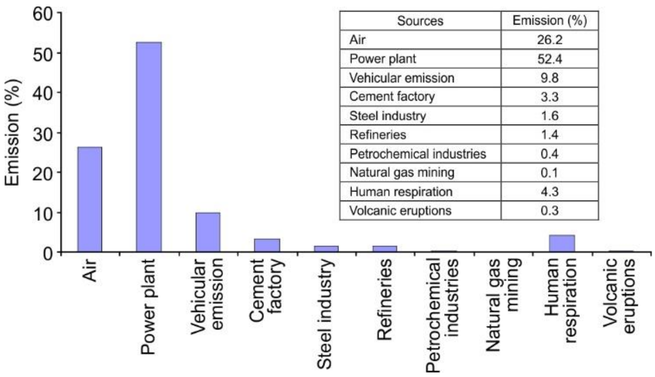
Increase in the concentration of carbon dioxide (CO_2) in the atmosphere due to anthropogenic activities has become an enormous problem in recent years. As CO_2 is one of the most potent greenhouse gases, increase in its concentration will lead to increase in the average temperature of the atmosphere, which is referred to as global warming. Global warming leads to unpredictable change in weather and climate, sea-level rise, decline in arctic ice caps, only to name a few. To get rid of this problem, carbon capture and sequestration has become one promising technological breakthrough. Porous materials have shown considerable advantages in the adsorption of CO_2. Among many porous materials like metal organic frameworks (MOFs), porous carbon-based materials, zeolites, *etc*., zeolites have shown certain advantages like tuning pore sizes and properties by ion exchange, easy regeneration, greater selectivity, *etc*. This review article explores the application of zeolites in the field of CO_2 separation, with a particular emphasis on recent advancements and emerging trends in their development and performance.

Keywords: CO_2 adsorption, Zeolites, Aluminosilicates, Porous materials.

INTRODUCTION

The world depends on carbon-based fuels for energy including fossil fuels like coal, petroleum and natural gas, as well as bio-based fuels. Combustion of these fuels lead to production of energy (heat) and gases like water vapour (H_2O), carbon dioxide (CO_2) and other products like ashes. With the advancement of science and technology, the energy demand increased sharply and as a result, the amount of CO_2 in the atmosphere have risen steadily and very fast. Not only from fossil fuels, the emission of CO_2 in the atmosphere due to different anthropogenic activities like vehicular emission, deforestation, burning of fossil fuels, power plants, *etc*. has almost doubled than 1970 and now its amount is more than 32 gigaton in atmosphere annually [1-5].

Presently, more than 98% of global energy requirement is supplied by burning of fossil fuels [6]. Although natural carbon cycle should steadily maintain the concentration of CO_2 in the earth's atmosphere, the combined effect of anthropogenic and natural emission of CO_2 has led to an increased amount of CO_2 in the atmosphere. This produces an average increase in the global temperature by 0.8-1.2 ºC with respect to pre-industrialization era and the intergovernmental panel on climate change (IPCC) predicts an increase by 1.5 ºC by 2030 to 2052 [7,8]. As a result, the sea water level may rise by 3.8 m [9,10] and average global temperature may increase by 3.7 ºC [11,12] by 2100 [13]. Besides, the increase in CO_2 in the atmosphere is expected to cause different air-borne diseases, increasing the risk of health hazards [14]. The loss in global economy due to climate change may be 5-20% of the global domestic production [15]. Fig. 1 depicts the relative amount of CO_2 emitted from different sources.



Sources	Emission (%)
Air	26.2
Power plant	52.4
Vehicular emission	9.8
Cement factory	3.3
Steel industry	1.6
Refineries	1.4
Petrochemical industries	0.4
Natural gas mining	0.1
Human respiration	4.3
Volcanic eruptions	0.3

Fig. 1. Different CO_2 emission sources in the atmosphere [16,17]

It is well-known that CO_2 is one of the most potent greenhouse gases and so, it's undoubtedly the need of the hour to limit the emission of CO_2 and other greenhouse gases so that the global temperature rise remains under control. To achieve this target, it is extremely necessary to limit anthropogenic activities releasing greenhouse gases and/or crucially removing the greenhouse gases from atmosphere. Therefore, to decrease the CO_2 emissions from different sources, many research projects are being undertaken at present time [18]. In this review, different aspects of CO_2 adsorption and separation by zeolites will be discussed keeping focus on the developments in this area in the recent times.

Aspects of separation of CO_2: For effective separation of CO_2 from flue gas or air, several factors are to be considered. These factors. Both physical and chemical methods are there for separation. The salient features of the adsorbent are also to be considered. The actual mechanism of adsorption is also discussed in forthcoming sections.

Methods of CO_2 separation: Four methods can be used to separate CO_2, namely adsorption, absorption, cryogenic methodologies and membrane technology [19-23]. The specific process to be followed depends on several factors, including the source of CO_2, the scale of the capture, the required purity of the captured CO_2 and its intended end-use [24]. Presently, the most followed procedure are adsorption and absorption [25]. Absorption utilizes selective solvents or chemicals which react with CO_2 and arrest it from flue gases or other sources. The chemicals are usually aqueous amine solutions, which utilizes the reaction between amine and CO_2 as their capture mechanism. The amine is usually monoethanolamine [26], diethanolamine [27] and methyl diethanolamine has been established since 1930 for CO_2 absorption [28,29]. The CO_2 rich solvent is then regenerated and the CO_2 is released for storage or utilization. The major drawback of this process is the regeneration of the absorbent due to high heat capacity of water. In search of a more sustainable, safe and renewable energy processes, two processes, which are gaining maximum importance are carbon capture and sequestration (CCS) and carbon capture and utilization (CCU) technologies [30-33]. The major focus of CCS is the capture of CO_2 from sources like power plants and injection in the ground for storage for a long duration. Alternatively, CCU aims at generating a sustainable employment of CO_2 in different functions like a solvent or in the synthesis of chemicals or fuels [34-37].

Carbon capture and storage (CCS) has been designated as the most encouraging CO_2 decreasing options among different strategies like using of non-carbon fuels or increasing the efficiency of the power plants, by IPCC. According to them, this process can lead to about 19% decline in global CO_2 emission within 2050 [38]. However, the CCS technology is only a temporary solution for greenhouse effect and global warming until new green and sustainable energy source is found to replace fossil fuels [38]. Among the three main steps *e.g.* capturing of emitted CO_2 from sources without allowing them to diffuse into atmosphere, transportation of the captured CO_2 after compressing them and lastly storage of the captured CO_2 underground, the first step is the most-costly step (*ca.* 80% of the total cost) and has been shown to be the most difficult one [38-41]. Nowadays, carbon capture, storage, and utilization (CCSU) is considered one of the most promising approaches, as it enables the conversion of captured CO_2 into commercially valuable products [16,42,43].

In the industrial scale CCS or CCU techniques, the most applied current procedures are adsorption in a liquid phase, adsorption in solids and membrane separation techniques. Absorption in amine solution has been employed in the CO_2 separation for many years, but the regeneration of the solution is high energy requiring process and also amine corrodes the equipment. Membrane technology has some advantages like low energy of the process, no liquid waste generation, *etc.* but there is always a competition between selectivity and permeability of the membrane used [44-49].

Solid adsorbents for CO_2 separation: For successful implementation of carbon mitigation processes, it is of pivotal importance to find an ideal sorbent for adsorption of CO_2 or any other gas. Solid adsorbents offer considerable advantages in this process as there is no liquid waste generation and regeneration of the adsorbent is much easier, as the gas is physisorbed in solid. In a process called pressure swing adsorption (PSA), the solid adsorbent can be regenerated in a facile way by lowering the pressure. Or in another more advanced technique, vacuum pressure swing adsorption (VPSA), desorption of the adsorbed gas is performed at a much-reduced pressure (< 1 bar). In temperature swing adsorption (TSA), the regeneration is carried out by increasing the temperature of the bed. These processes are less complicated than the regeneration of the liquid adsorbent, because a separate column is needed for them to strip off the adsorbed gas. The adsorbent should possess some very important desirable qualities. In terms of CO_2 adsorption, they should have high capacity for adsorption of the targeted gas (in mmol/g), along with high working capacity for CO_2. Working capacity refers to difference in adsorption capacity of the adsorbent at operating and regeneration temperature and pressure. High adsorption capacity is the key to high working capacity. Selectivity is another extremely important quality, selectivity towards CO_2 over other gases will affect the purity of the adsorbed gas, if it is to be used in other purposes (like CCSU). Another key factor is fast adsorption/desorption kinetics. There should be easy rejuvenation of the adsorbent. The higher the enthalpy of adsorption of CO_2, greater is the energy required to desorb the adsorbed gas, thereby increasing the cost of regeneration. The adsorbent must have high mechanical stability under the operating conditions of adsorption and desorption. There should also be high chemical stability and they should be able to tolerate the impurities. For CO_2 sorption, there are impurities like CH_4, H_2, N_2, water and also many sulphides. Sometimes, flue gas is pre-treated to remove the impurities to lower the competition for adsorption. The production cost of the adsorbent should also be minimum, to make this process economically viable [30].

Solid adsorbents such as silica [50,51], zeolites [52-54], metal organic frameworks (MOF) [55-58], activated carbon [59-62], graphene [63,64], metal oxides [65,66] and organic polymers [67-70] are majorly used adsorbents of CO_2. All the adsorbents have their own advantages and disadvantages. Among different promising solid adsorbents, for example, carbon-based materials have been found to be interesting due to their low cost, high thermal stability, wide availability and their hydrophobicity. But, their affinity towards CO_2 may be inherently low, particularly at low pressure. But functionalizing the material with basic nitrogen functionalities have been proven to enhance the adsorption of CO_2 in these materials, but at the expense of losing the hydrophobicity [71,72]. Likewise, MOFs have high CO_2 adsorption capacities with values as high as 33.5 mmol/g at 35 bar pressure [73], but at lower pressure ranges, MOFs may become less effective (< 5 bar). In these situations, zeolites exhibit better adsorption [73-75]. This is due to the stronger interactions between CO_2 and zeolites compared to those with MOFs, which allows zeolites to adsorb CO_2 more effectively at lower pressure ranges. However, modern research showed some MOFs like SIFSIX-3-Cu and SIFSIX-3-Ni show high CO_2 adsorption capacity even at very low pressures, making these MOFs promising candidates for direct air capture [76,77]. The main disadvantages of MOFs include high synthesis costs and limited hydrothermal stability, which in turn make their regeneration more expensive [78]. At lower pressures, zeolites demonstrate average adsorption of CO_2, but the adsorption can be greatly enhanced by post synthetic moderations. Their low synthetic costs and robustness of structures are other notable positive attributes for this type of separation. But, like other adsorbents, zeolites also suffer from certain disadvantages, like presence of water in the gas mixtures make the separation extremely difficult, because water and CO_2 possess competitive adsorption in zeolite adsorption sites [47,79-82].

Also, the binding of CO_2 in zeolites have been stronger, thereby rendering the regeneration process much costly [83]. Moreover, there is scope for upgrading the adsorption capacity and physico-chemical properties of zeolites (like pore dimension, organization, *etc.*) by different modifications. These advantages make zeolites a leading adsorbent of CO_2. High surface area, appropriate pore size and thermal and chemical stability of zeolites, especially at low temperatures, make them most sought-after adsorbent material of CO_2 [84-87].

Zeolites as effective adsorbents: Zeolites are silicate materials composed of tetrahedral SiO_4^{4-} units interconnected by oxygen bridges, forming a robust three-dimensional framework. These interconnections produce 3D crystalline microporous structures (*i.e.* with pores with $d < 2$ nm). There exist many different types of zeolite structures. Many zeolites have been widely used as catalysts support, in water purification and other very important industrial and technological applications [88,89]. These are mainly differentiated by the size and shape of the framework channels and cages. It has been found that in zeolite structures, a fraction of the Si atoms can be substituted by other elements, most commonly with aluminium. So, basically, zeolites are aluminosilicates [90-93]. The presence of Al^{3+} instead of Si^{4+} renders the structure negatively charged [94]. These negative charges are compensated by extra-framework cations like H^+, Na^+, Ca^{2+}, La^{3+}, *etc.* Cations in zeolites play a critical role in CO_2 capture since they can attract CO_2 into zeolite [95,96]. Ion-exchange procedure is the most frequently technique used to modify these counterions. Different zeolites demonstrate different efficiency in the separation of CO_2 from gas mixtures [97,98].

Zeolites may be divided into two classes *viz.* natural and synthetic [99]. The physical and chemical characteristics like crystal structure and chemical composition determines the efficacy of zeolites in CO_2 separation [100]. Various natural zeolites such as clinoptilolite, heulandite, stilbite and chabazite are used in CO_2 capture [101-103]. To enhance their CO_2 adsorption performance, researchers have investigated modifications and functionalization such as acid treatment, ion-exchange and metal loading [104,105].

Natural zeolites are cost-effective compared to synthetic zeolites, but their properties may be less consistent. On the other hand, Synthetic zeolites have high-purity and they have miscellaneous range of applications. Synthetic and natural zeolites usually undergo modifications to enhance their efficacy in CO_2 adsorption. The most prominent modification technique has been ion-exchange of zeolites [106,107].

Depending on the size and nature of channels and cages, zeolites are distinguished in different categories. Presently, 248 types of fully ordered zeolite frameworks are known [108]. To allow CO_2 to diffuse in to the pores, the aperture must be at least 3.3 Å. Accordingly, the ring size should be at least 8 membered (8MR) ring. The size of the pores can be tuned by incorporating alkali or alkaline-earth metals, which affect the adsorption mechanism. Apart from H^+, increasing the atomic mass of the counterion leads to decrease of the surface area. The topology and size of the pore of a zeolite are important parameters, which can affect the diffusion of adsorbates and determines its performance as adsorbent.

In this context, the microporous zeolites are further divided into two groups. The medium- and large-pore zeolites which have pore sizes in the range of 0.45-0.60 nm and 0.60-0.80 nm, respectively. The second group is small-pore zeolites, which have pores in the range of 0.30-0.45 nm. Small pore zeolites have their pore sizes comparable to the adsorbent gases like CO_2, CH_4, N_2, *etc*. As a result, medium- and large-pore zeolites primarily rely on electrostatic interactions for adsorption, while small-pore zeolites also depend on diffusion limitations and molecular size exclusion to enhance selectivity [109]. Table-1 shows the kinetic diameters and polarizability of important relevant gases related to flue gas or biogas separation.

TABLE-1
KINETIC DIAMETERS OF THE
GASES CO_2, N_2 AND CH_4 [110,111]

Gas	Kinetic diameter (Å)	Polarizability [(× 10^{-25}) cm^3] [112]
CO_2	3.30	29.11
N_2	3.64	17.40
CH_4	3.80	25.93

For example, zeolite X and Y both have the FAU (faujasite) structure, meaning their crystalline nature are same. They differ in the Si/Al ratio, which is 1 for X and more than 1.5 in Y. thus zeolite Y has less proportion of Al atoms and as expected, will have lesser number of extra-framework counterions per unit mass of zeolite (Fig. 2).

ZSM-5 is a MFI type zeolite framework with medium pore size, having interconnected channels (Fig. 2). In this case, the Na^+ sites are the places where the interaction with CO_2 takes place. Only the Na^+ sites which are present in 10 membered ring channels, because CO_2 can access only these sites. The

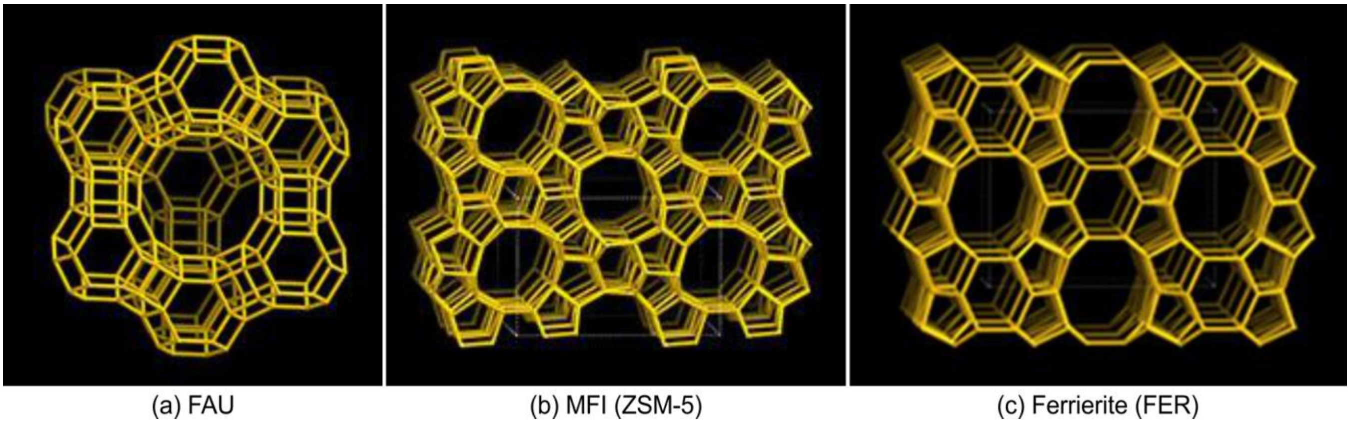

(a) FAU (b) MFI (ZSM-5) (c) Ferrierite (FER)

Fig. 2. Frameworks of some medium and large pore zeolites from IZA website [108]

kinetic diameter of CO_2 (3.3 Å) being higher than the 5MR pore sizes (~ 1.5 Å), are usually inaccessible by CO_2 [113].

Ferrierite [FER] is another medium-pore zeolite having straight 10 membered ring channels connected through cages with 8 membered rings (Fig. 2). In Na-Ferrierite, the Si/Al ratio has been found to govern the adsorption mechanism [114]. For example, in Na-Ferrierite having Si/Al ratio of 8.7, DFT calculations indicate a two Na^+ cation having a bridge with the adsorbed CO_2. But, when the zeolite has very low Al content (Si/Al is 26.6), the Na^+ concentration decreases to such an extent that the bridge between two Na^+ ion and CO_2 becomes improbable and CO_2 may interact with one Na^+ ion only.

In small pore zeolites like zeolite A, chabazite, *etc.* (Fig. 3), the adsorption mechanism is almost same as that in medium or large pore zeolites with an extra parameter. As the size of the pore becomes comparable to the adsorbent size, the accessibility of the pore becomes the governing factor for the selectivity and adsorption capacity. So, the adsorption and separation of CO_2 depends heavily on the Si/Al ratio of the zeolites. Not only for CO_2, has the Si/Al ratio determined the adsorption capacity of other polar molecules as well. For low Si/Al ratio, where the Al content of the zeolite is high, there are obviously more number of extra-framework counter cations are present. These extra-framework cations are majorly the site for adsorption. So, amount of adsorption active sites per unit mass of zeolite increase as Si/Al ratio decreases [115-117].

Again, the Lewis basic oxygen site which is adjacent to the Al sites of zeolite interacts with acidic CO_2 molecule. This number of basic sites also increases with decreasing Si/Al ratio. So, in total, the adsorption of CO_2 increases for a zeolite with decreasing Si/Al ratio or increasing Al content [118,119]. But, with increasing Al content, there are a greater number of extra cations, which results in the decrease of pore volume, increasing steric hindrance inside the pore, thereby affecting the adsorption capacity. In a recent study, Palomino *et al.* [120] have shown that zeolite LTA with Si/Al value 1 showed lower adsorption capacity than LTA zeolites having Si/Al values 2, 2.35 and 5. The decrease in adsorption was attributed to the decrease in pore volume with increase in Al content. This observation was also corroborated by computer simulation programme [121]. It is also pertinent to mention here that zeolites having higher Al content have more hydrophilicity and lower thermal stability and the Si-O-Al bond is more prone to hydrolysis than Si-O-Si bond. Therefore, aluminosilicates with a lower Si/Al ratio are more difficult to regenerate and exhibit reduced hydrothermal stability [79,118].

Major factors related to CO_2 separation with zeolites: Separation of CO_2 from other major components like N_2 or CH_4

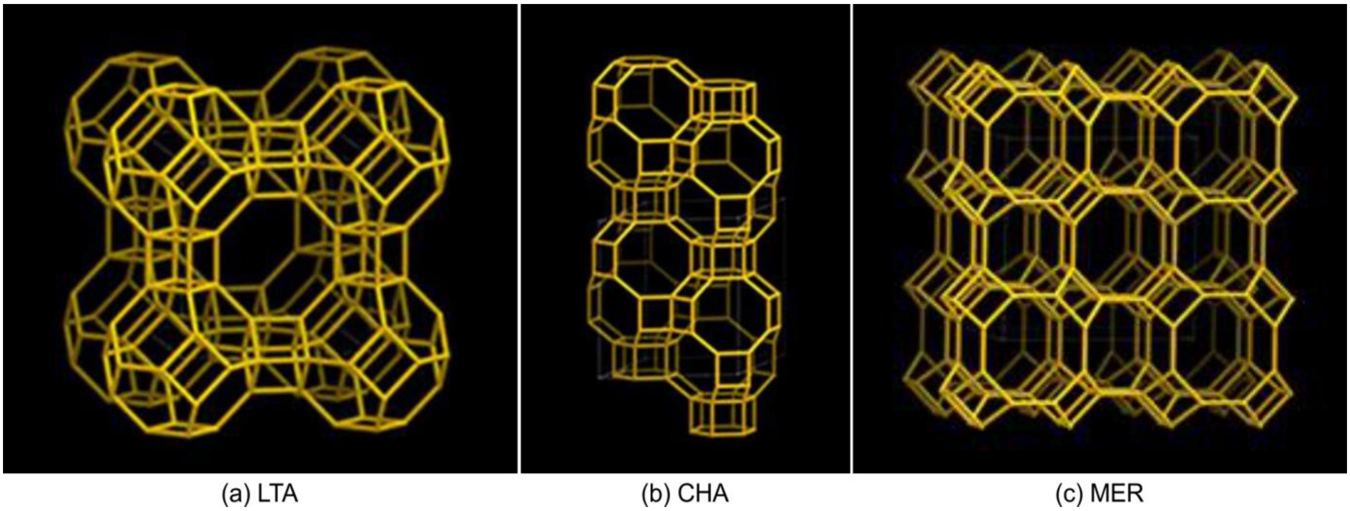

(a) LTA (b) CHA (c) MER

Fig. 3. Frameworks of some small pore zeolites from IZA website [108]

depends on equilibrium factors of preferential adsorption of CO_2 on zeolites [122-125]. Gas-zeolite interaction have been found be of three general types like van der Waals interaction, electrostatic interactions and weak chemical reactions. The van der Waals interaction depends primarily on polarizability of the gas molecule. Electrostatic interaction between the ionic framework of zeolites and the gas molecule is one of the most prominent factors for adsorption and weak chemical interaction refers to the formation of π-type interaction of the gas molecule and zeolite ionic sites like metal cations and also acid-base interaction among the acidic gases and basic sites like amine groups. CO_2 has greater polarizability, quadrupole moment and chemical reactivity among the gases like CH_4, N_2, *etc*. resulting in greater interaction with zeolites [126-128].

CO_2 being smaller than N_2 or CH_4, can be easily separated with zeolites having pore aperture size greater than CO_2 but smaller than N_2. Also, different diffusion rates of different gases lead to the kinetic separation of CO_2 from the other mentioned gases [129-133]. Depending on the characteristics of zeolites, there will be a 'gating effect' which will facilitate the admission of CO_2 in the zeolite structure [134-138]. For zeolites, the gate keeping groups *i.e.* the counterions will block or allow the entry of a gas molecule, thereby creating a trap door mechanism for incoming gases.

Mechanisms of CO_2 adsorption in zeolites: In zeolites, the extra-framework cations can modify the adsorption behaviour (usually Na^+, but other mono or multivalent ions are also used). The exchangeable cations are the acid sites and the oxygen atoms attached to aluminium in the framework are the basic sites [139]. As the electronegativity of the cation decreases, the basic site strength increases due to a greater negative charge localized on the oxygen atom [139-144]. So, for alkali metal exchanged zeolites, the basicity order is Cs > Rb > K > Na > Li. Now, CO_2 being acidic in nature, the zeolite with higher basicity will have a stronger interaction and hence, the adsorption and separation will be higher. But, again, higher the ionic radii, lower will be the polarizing power of the cation. So, ions with larger ionic radii will have lower interaction with CO_2 (Li >Na > K > Rb > Cs) [145].

Again, the pore volume and size are also affected by the ionic radius of the cation, thereby affecting the adsorption. So, in zeolites, there is a complicated balance between the basicity, polarizing power and pore size introduced by the extra framework cations in zeolites. This effect will vary from zeolite to zeolite. Among all these effects, which one will be the dominant one will depend on the zeolite type. For most of the medium and large pore zeolites like zeolite X, Y, ferrierite *etc*., the basicity has become the main governing factor and adsorption follows the order K > Na > Li exchanged zeolites upto 1 bar pressure [146-149].

For CO_2 loading, pore volume becomes an important factor. The order of size of the cations increases in the order Li < Na < K < Rb < Cs. It has been found for zeolite Na-X that when it is ion-exchanged with Cs^+, the available pore volume declines considerably (from 0.36 cm^3 g^{-1} to 0.19 cm^3 g^{-1}) [148]. As a consequence, for Cs exchanged zeolites, for lower partial pressure, the basicity of the exchanged ion governs the trend and the adsorption capacity follows the order Cs > K > Na > Li, but at higher CO_2 pressure the role of the lower pore volume becomes principal dominating factor and Cs-exchanged zeolite does not have the highest adsorption capacity [146, 148,149].

In some small pore zeolites, increasing the ionic radius of the exchanged cation has led to the decrease in the adsorption capacity of the zeolites, which may be due to either the change in the polarizing power of the cation or the change in the pore volume of the zeolite [150] for zeolite A, the order is Na > K > Cs exchanged zeolite [79,80]. So, exchanging with cation decreases the pore aperture and molecules like CO_2 has difficulty in entering the pores. Also, this prevents the diffusion of larger molecules N_2, or CH_4 more effectively, so the selectivity towards CO_2 sorption increases [143]. Multivalent ion exchanged zeolites showed lower adsorption than monovalent ion exchanged zeolites at 1 bar pressure [74,115,149]. This is because for multivalent ions, lower number of cations are required to balance the total charge, thereby leading to the lower number of basic adsorption sites [74].

But in small pore zeolites, like zeolite A or chabazite, the Ca-exchanged or Mg-exchanged zeolites have shown better adsorption than Na or K-exchanged zeolite, presumably due to the lower number of multivalent cations are required to balance the charge than monovalent ions, thereby increasing the available pore volume, increasing the adsorption capacity [74,75].

Presence of water in biogas and flue gas has been found to have great influence in separation of CO_2. For low Si/Al ratio zeolites, water vapour has significantly decreased the adsorption capacity for zeolites like zeolite A or X, because of competitive interaction of H_2O and CO_2 for same adsorption sites. As water is dipolar, the interaction of water and the adsorption sites of zeolites are quite strong. For zeolite X, the adsorption of water decreased consistently with increasing the loading amount (-78 kJ/mol at initial level to -50 kJ/mol, respectively). For CO_2, the enthalpy value is almost at steady level of around 40 kJ/mol [151,152]. So, it is obvious that adsorption of water vapour in zeolite X is stronger than that of CO_2. So, increase in the water vapour content in wet CO_2 streams lead to decrease in CO_2 separation and adsorption [153]. A faujasite zeolite with Si/Al ratio 31 has shown an adsorption of CO_2 almost double than faujasite zeolite with Si/Al ratio 2.1. The latter has demonstrated water adsorption 10 times higher than the first one. This is evidently due to increase in the amount of Al in the framework, which will lead to increase in the hydrophilicity of zeolite [116]. For zeolite 5A, same observation was reported, but in another study, it was shown that trace amount of water vapour leads to an increase in the CO_2 sorption, may be due to the electrostatic interaction with water molecules. But in general, as higher water vapour will gradually fill up the pores of the zeolites, smaller space will be available for the adsorption of CO_2 [154,155].

So, the complexity due to water may decrease with increasing Si/Al ratio. But, decreasing Al content will also lead to decrease in the number of active sites in zeolites. So, better option should be to dry the flue gas or gas mixtures before exposing to zeolite adsorbent within the PSA column. Another multi-layered PSA column has also been designed, in which two layers of adsorbents were used to adsorb water and CO_2 in the same bed [156]. The first layer in the adsorbent bed was activated alumina which will adsorb water and the second layer was zeolite X for CO_2 adsorption. This system leads to 77% CO_2 recovery from a humid (3-4% water) gas stream of synthetic flue gas with CO_2 purity of 67%. Still, the dry process has been the best in which a CO_2 purity of >95% and recovery of >70% could be achieved [157].

The most practical parameter to judge the performance of zeolites in carbon dioxide sorption and separation agents are the adsorption capacity and selectivity towards CO_2. There are many types of selectivity parameters used, but the most widely accepted parameter is the pure selectivity of CO_2 over pure N_2 or CH_4. This value is calculated by dividing the adsorption capacity at a chosen pressure and temperature (*e.g.* 1 bar, 25 ºC) of the pure component isotherm of CO_2 by adsorption capacity of pure component isotherm of N_2 or CH_4. This approach is commonly employed, as adsorption experiments with binary gas mixtures are significantly more complex than those involving pure components [47].

Comparison of CO_2 adsorption performances of some zeolites: The examples of some adsorption performances of some representative zeolites are described in Tables 2 and 3. These data demonstrates the CO_2 adsorption capacities of zeolites and metal exchanged zeolites over N_2 (Table-2) and that over CH_4 (Table-3). The largest adsorption capacity has been demonstrated by zeolite X and Y, both belonging to faujasite (FAU) family. Zeolite X having the highest adsorption capacity, are often employed as a standard for comparison of adsorption capacities of other types of adsorbents like MOFs and carbon-based adsorbents and also other types of zeolites. One of the highest adsorption capacities among zeolites has been shown by zeolite X (6.3 mmol/g), which can be further increased by ion-exchange (LiX has an adsorption capacity of 7 mmol/g). The selectivity is also high as adsorption of CH_4 and N_2 are both not significant. Zeolite Y having higher Si/Al ratio than zeolite X shows a lower adsorption capacity [116]. But, zeolites with higher Si/Al ratio tend to be less affected by water in gas mixture. Hence, Y is a good choice for adsorption. The selectivity towards CO_2 sorption for zeolites X and Y have similar values.

Zeolites X and Y having higher pore dimensions are not able to kinetically separate CO_2 from N_2 or other gases, whereas small pore zeolites like zeolite A, chabazite or merlinoite have shown greater promise. Zeolite A has ion exchange tuneable pore size. Partially exchanged Na^+ and K^+ exchanged zeolite A has been shown to adsorb CO_2 over larger gases like CH_4 or N_2 [79,150]. For the highest value reported for adsorption on zeolite A, the zeolite was exchanged with Na^+, K^+ and Cs^+ [160]. For merlinoite [MER] zeolite (Fig. 2c) the usual pore size is 4.2 Å, which is subject to change upon ion-exchange with K^+ or Cs^+. The selectivity towards CO_2 has been one of the highest for Cs-MER zeolite (250) and another advantage being this zeolite essentially does not adsorb CH_4 [136].

TABLE-2
CO_2 ADSORPTION CAPACITY AND SELECTIVITY OVER N_2 OF SOME ZEOLITES[a]

Zeolite	Si/Al	nCO_2 (mmol/g)	nN_2 (mmol/g)	Pure selectivity[b] nCO_2/nN_2	Ref.
Ca-X	< 1.5	5.2	0.3	17	[75]
Na-Y meso	2	5.4	0.2	27	[157]
Na-A[c]	1	3.9	0.3	13	[79]
NaK-A	1	3.4	0.02	170	[79]
Ca-A[d]	1	5.1	0.6	9	[75]
Na-CHA	4	4.7	0.7	7	[117]
K-CHA	1.2	2.1	0.03	80	[135]
Cu-SSZ-13	6	3.8	0.3	13	[111]
Cs-BEA	7.4	1.2	0.06	24	[149]
SSZ-45	250	1.4	0.2	8	[158]
Na-ZSM 5	25	2.1	0.3	8	[159]

[a] Standard reaction condition is 1 bar and 25 ºC; [b] Pure selectivity is calculated nCO_2/nN_2; [c] at 0.85 bar pressure; [d] Si/Al ratio was not measured, but zeolite A always has a Si/Al ratio of 1 and for zeolite X the Si/Al ratio is ≤ 1.5 [30].

TABLE-3
CO_2 ADSORPTION CAPACITY AND SELECTIVITY OVER CH_4 OF SOME ZEOLITES[a]

Zeolite	Si/Al ratio	nCO_2 (mmol/g)	nCH_4 (mmol/g)	Pure selectivity[b] nCO_2/nCH_4	Ref.
Na-X	1	6.3	0.8	8	[74]
Li-X	1	7.0	0.5	13	[74]
Na-Y meso	2	5.4	0.4	14	[157]
NaK-A	1	3.8	0.03	127	[151]
NaCsK-A	1	3.5	0.01	350	[160]
Na-CHA-4	4	4.7	1.3	4	[116]
K-CHA	1.2	2.1	0.02	93	[135]
Na-RHO	3.9	4.9	0.1	49	[138]
Na-MER	3.8	3.8	0.3	13	[136]
Cs-MER	3.8	2.5	0.01	250	[136]
SSZ-45	250	1.4	0.26	5	[158]
Na-ZSM-5	25	2.1	0.8	3	[159]

[a] Standard reaction condition is 1 bar and 25 ºC; [b] Pure selectivity is calculated nCO_2/nCH_4.

However, that Cs-MER exhibits a slightly lower adsorption capacity (2.5 mmol/g) compared to NaCsK-A (3.5 mmol/g). Breakthrough experiments demonstrated that K-MER zeolite exhibits fast adsorption kinetics and yields high-purity CO_2 upon desorption, with high selectivity of 850.

For zeolite chabazite (CHA) (pore size 3.7 Å) is a small pore zeolite. Obviously, different cations will affect the pore dimensions and thereby will affect both selectivity and adsorption capacity. For K-CHA the selectivity is quite high, but the adsorption capacity (2.1 mmol/g) is lower than the other comparable zeolites [135]. Zeolite RHO (pore size 3.6 Å) has lower parameters than the above discussed zeolites, but Na-RHO and K-RHO have shown somewhat better parameters. Cs-RHO, however, showed little less adsorption capacity at lower pressures, but the saturation limit for Cs-RHO was quite high at 6 bar pressures while for Na or K-RHO, the values were nearly 1-2 bar [138]. Na-FER zeolites with two Si/Al ratio of 8.7 and 26.6 showed CO_2 adsorption capacity of 2.8 mmol/g and 2.3 mmol/g, respectively. Inspite of showing higher adsorption capacity, the lower Si/Al ratio zeolite has demonstrated higher enthalpy of adsorption (10-15 kJ/mol) probably due to different adsorption mechanisms [113]. Also, changing the temperature from 0 ºC to 60 ºC makes the adsorption capacity to decrease by approximately 0.7 mmol/g, signifying its applicability in higher temperature flue gas treatment [113].

Another remarkable zeolite is SSZ-45 with an extremely high Si/Al ratio of approx. 250. So, the CO_2 adsorption capacity at 1 bar pressure is relatively low, because it is a small pore zeolite. But here, upto even 35 bar pressure, the adsorption capacity keeps on increasing. Also, due to very high Si/Al ratio, the zeolite is less affected by water present in the gas stream [158].

Direct air capture (DAC) by zeolites: The discussions so far focuses mainly on the sources of large-scale production of CO_2, like industries, but CCS and CCU does not work with mobile sources of CO_2 [161]. So, DAC becomes an important approach for carbon neutrality, which can capture CO_2 from air directly at concentration nearly 400 ppm [162,163]. The decrease in the zeolite performance as adsorbent decreases in lower concentration of CO_2 due to poor gas zeolite interaction [164-166].

The CO_2 adsorption sites in the zeolites are extra framework cations and zeolite oxygen atoms. Studies by Stuckert *et al.* [167] studied the adsorption capacity of alkali metal cation (Li^+, Na^+ and K^+) exchanged low silica zeolite X (LSX) and reported that Li-LSX showed best adsorption capacity of CO_2 at 395 ppm [167]. The trend of CO_2 adsorption (Li-LSX > Na-LSX > K-LSX) associates with the charge-to-size ratio of alkali metal cations ($Li^+ > Na^+ > K^+$). This study suggests that metal cations are the predominant CO_2 adsorption sites [168]. Tao *et al.* [168] reported similar findings with cation-exchanged LTA zeolites, where Ca-LTA exhibited the highest CO_2 capacity at 400 ppm.

Alkali and alkaline-earth metal exchanged zeolites X and Y (with Si:Al = 1.2 and 1.6, respectively) demonstrated the importance of framework O-atoms. This result is proved by the consistent trend of mean charge of zeolite framework O and CO_2 adsorption heat [169]. But this result is contradictory to that of LSX zeolite previously discussed [106]. This contradiction suggests that the predominant CO_2 adsorption sites can vary in zeolites with different Si/Al ratio. Still regulating the charge-to-size ratio of metal cations remains the chief approach in producing metal cation-exchanged zeolites for DAC. For example, Ba^{2+} having smaller charge-to-size ratio (and smaller electronegativity) than Mg^{2+} and Ca^{2+}, can induce a greater charge on zeolite framework O, which in turn, can result in higher CO_2 capacity in zeolite X. The efficacy of metal cations as CO_2 adsorption sites is also affected by their locations. Oda *et al.* [170] demonstrated that the simultaneous presence of two Ca^{2+} ions at the 8- and 6-membered ring sites in LTA zeolites (with a Si/Al ratio of 1) plays a crucial role in CO_2 adsorption at 400 ppm. This arrangement strengthens CO_2 interaction by allowing one molecule to engage with both Ca^{2+} ions at the same time. Fu *et al.* [171] identified that the primary sites for CO_2 adsorption at 400 ppm are Na^+ cations situated in the side-pockets of eight-membered rings in MOR zeolites and Zn^{2+} cations positioned at the double six-membered rings in CHA zeolites.

The framework topology of zeolites is another important factor for DAC. Fu *et al.* [172] showed that a diminishing CO_2 adsorption capacity decreases (per adsorption site *i.e.* Na^+) with increasing confined space. MOR zeolites, having confined space approximately 6.3 Å, exhibited the highest CO_2 capacity per Na^+. This is due to stronger zeolite-CO_2 interactions originated from larger electric field in a small confined space. Xiang *et al.* [173] also reported that narrowing the pore of zeolite 13X by introducing Fe atom to replace Si, Al atoms resulted in a greater CO_2 adsorption capacity at 400 ppm. These findings suggest that zeolites with smaller cages are more favourable candidates for DAC.

Conclusions and future aspects

In summary, it may be understood that small pore zeolites like chabazite, RHO *etc*. have very high selectivity for CO_2 as well as good adsorption capacity. They are useful agents in biogas treatment. But, still there are scope for further investigating the role of small pore zeolites in CO_2 sorption [43]. Large pore zeolites usually have lower selectivity for CO_2, but zeolites like X or Y have shown highest adsorption capacity values. The performance of zeolites in this application usually decreases with increasing temperature, though it is a very crucial parameter for flue gas treatment. The temperature of flue gas are usually higher (*ca.* 80 ºC). There are zeolites like FER where there is relatively small decrease in the adsorption capacity with increasing temperature.

Regarding the impurities in the gas stream, apart from water, there may be other impurities as well. These impurities greatly affect the adsorption capacity of zeolites. The effect of these impurities in adsorption capacity and selectivity of the adsorbents are yet to be explored in great detail. Further, new types of zeolite-based materials can be synthesized and utilized in the adsorption of CO_2. Ion-exchange may be performed using different ions, which are so far not explored properly or altering the pore dimensions by new synthetic methodologies may also be an intriguing research in this field of materials chemistry. In spite of having high surface area comparable to MOFs, the pores of all small pore zeolites are not accessible to CO_2, so, synthetic strategies should be designed to synthesize mainly relatively larger pore zeolites. An interesting approach may be to coat a zeolite by hydrophobic layer of another zeolite- the so called core-shell zeolite systems may be explored. Zeolite incorporated polymeric membranes may also by applied for CO_2 sorption process [174]. A recent study demonstrated the effective use of zeolite SSZ-39 incorporated in a polyimide matrix producing a flexible membrane material for excellent separation of CO_2 from CH_4 [175].

Zeolites can find industrial application in (V)PSA/TSA processes [30,176-180]. Although powdered zeolites have been mostly employed for adsorption of CO_2, it is essential to shape these powders to give them some macroscopic figure like pellets or other shape [181,182]. Major processing techniques are extrusion, coating of scaffolds and honeycombs, colloidal processing and casting of porous powders, sacrificial templating, 3D printing [181,183-187]. For example, 3D printing technique was applied with gelatin and pectin as organic binders to synthesize zeolite X, 5A and ZSM-5 monoliths. These organic moieties were removed by calcination to produce honeycomb structured zeolite monoliths [188]. Resin beads were employed as hard template to synthesize binderless zeolite LTA [189]. To produce binderless zeolite X beads, metakolin was used as a temporary binder. Zeolite X was granulated with metakolin and the beads so obtained were subjected to hydrothermal conversion in alkaline medium to produce binderless zeolite X beads of approximate size 1.6 to 2.5 mm [190]. A chitosan assisted synthesis method was also applied to produce binderless zeolite X [191]. Silica sol and chitosan mixture was added into a sodium aluminate solution, producing microspheres.

Solid adsorbents present an important solution to the carbon capture because they are tremendously encouraging materials for efficient and selective sorption of CO_2 and other related gases. Although the adsorbent materials established to date are quite usable, they need further advancements. The improvement of such advanced materials will need substantial advances in materials design, synthesis and characterization and also detailed understanding of the structure properties relationships in materials.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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