

# Carbon Dioxide Capture Technology

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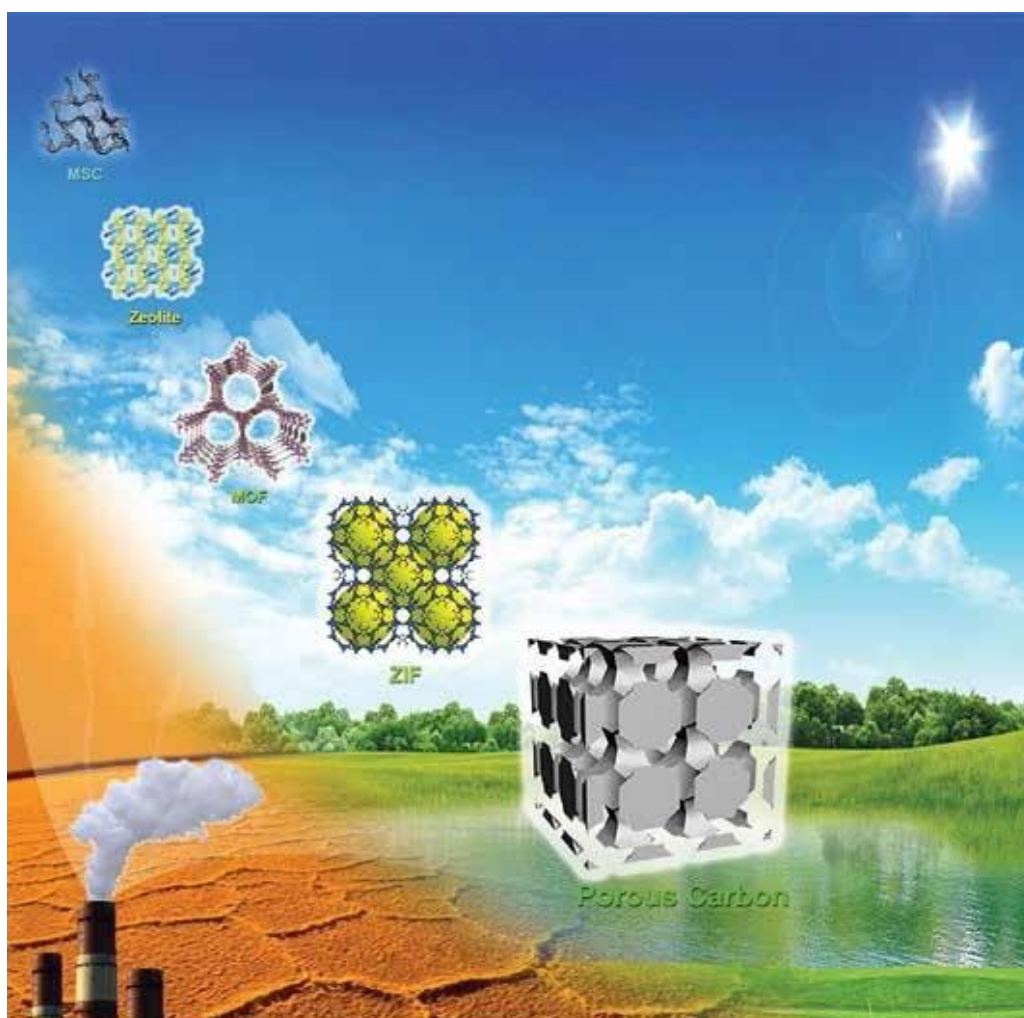
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Image Courtesy: Angew. Chem. Int. Ed. 2010, 49, 6058 – 6082

**ASSIGNMENT ON CARBON CAPTURE TECHNOLOGY  
SUBMITTED TO NATIONAL CENTRE FOR CATALYSIS RESEARCH  
IIT MADRAS**

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This short writing highlights the challenge for capture technologies, especially post combustion, precombustion capture and natural gas sweetening. The recent developments such as solvent absorption, chemical and physical absorption, and membranes, will be talked about.

## **1. Introduction**

The increasing level of carbon dioxide in the atmosphere is the single of the most environmental problem the land faces. The drastic rise has been ascribed to an increasing dependence on the burning of fossil fuels like coal, natural gas and gasoline, which accounts for nearly 86% of anthropogenic greenhouse gas emissions, the remaining from land usage change and chemical processing.

Schemes to reduce global atmospheric concentration of GHG's has received a great heap of attention in recent times, especially from governments and industries, and a routine of high profile collaborative programs has been installed, including IPCC (Interogovernal Panel on Climate Change) & GCCI (Global Climate Change Initiative). The capture and sequestration of carbon dioxide is the main strategy in these initiatives. CCS (Carbon Capture & Sequestration) include a group of technologies for the capture of carbon dioxide from various sources followed by compression, transport and permanent storage. CCS also focusing to improve the energy efficiency, shifting to less carbon intensive fuels (natural gasoline) and phasing in the role of alternative energy resources like solar energy, wind and biomass.

A critical point is the development of CCS scheme which is a multifaceted problem. It requires shared vision and worldwide collaborative efforts from governments, industries, policy makers and economist, as well as scientists, engineers and venture capitalist. From these views, it is clear why the carbon dioxide capture is regarded as one of the grand challenges for the 21<sup>st</sup> century.

The existing methods such as amine scrubbers and cryogenic coolers are energy intensive and are not cost effective for carbon emissions decrease. This is the primary cause for the dense deployment of fully integrated commercial CCS schemes.

Several parts of the CCS process chain including compression, transportation (by pre-existing pipelines for Instance), and storage of CO<sub>2</sub> are technologically mature and Available, and a growing number of fully integrated CCS Projects are reaching the pilot and demonstration phases prior to commercialization.

In accession to three large-scale demonstration projects which are presently underway in the Sleipner West (Norway), Weyburn (Canada), and In Salah (Algeria), several smaller projects have started out on the Dutch continental shelf (Netherlands), Snøhvit (Norway), La Barge (Wyoming, United States), Fenn Big Valley (Canada), Ketzin (Germany), and Schwarze Pumpe (Germany). All of the current projects demonstrate carbon storage or reuse in enhanced coal-bed methane recovery systems, although one project at Schwarze Pumpe in Germany,

captures CO<sub>2</sub> at a coal-based plant. A further 40 CCS projects have already been proposed worldwide between 2008 and 2020.

One explanation for the dense deployment of fully integrated commercial CCS schemes is the considerable cost of the capture phase, which represents approximately two thirds of the total cost of CCS.

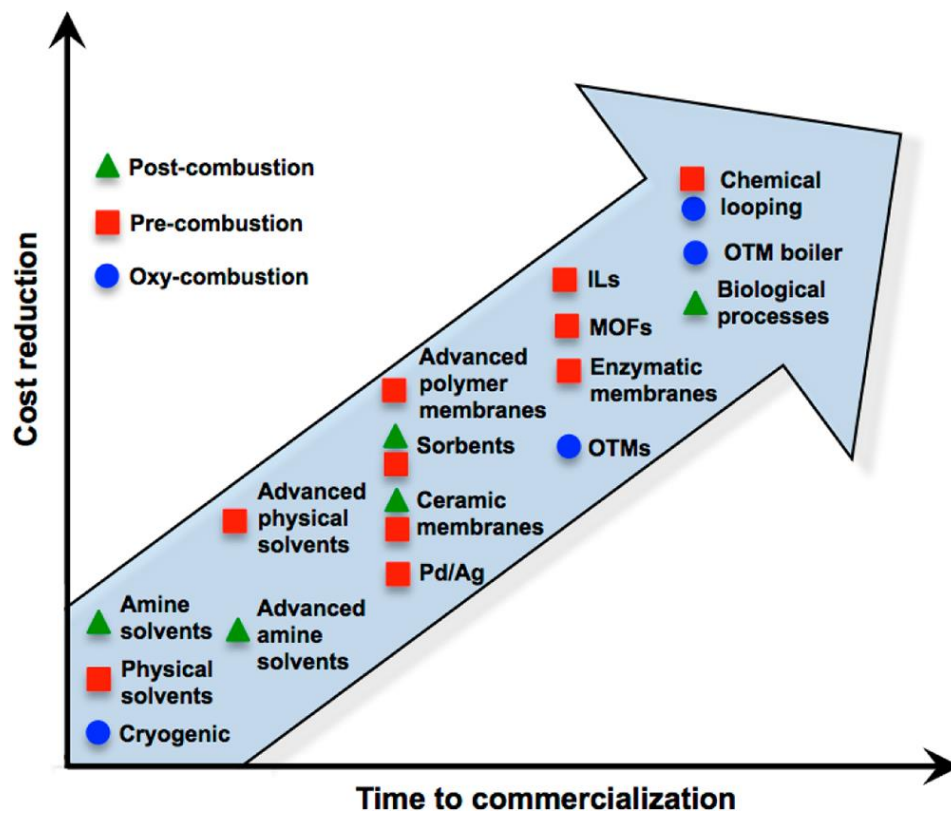
A recent comprehensive report on post combustion CO<sub>2</sub> capture technologies has seen that the regeneration energy, succeeded by the capital cost of capture specific equipment are the two variables contributing most significantly to the monetary value of CO<sub>2</sub> capture. One significant contributor to the regeneration energy is the maximum separation efficiency, which can be achieved by a given capture material. Enhancing this efficiency, will possess the greatest potential for turning down the overall monetary value of capture systems in near-term, with improvements in the capture phase for new materials representing one of the first challenges.

As recorded in Figure 1, there exists a serious demand for research on innovative new fabrics in order to shorten the time to commercialization.

It is apparent that a consideration of the process design and economics costs are needed to evaluate fully the potential of any given new material.

Here, we concentrate on the important challenge of CO<sub>2</sub> capture and highlight recent improvements in materials and emerging concepts. The stress is on

designing materials in which a molecular level of command can be achieved as a means of tailoring their performance in separating relevant gas mixtures.



**Figure 1.** Advanced concepts in CO<sub>2</sub> capture and their cost reduction benefits versus their remaining development needs/time to commercialization.  
Adopted from Chem.Rev.2014, 114,1413-1492

In this respect, special care is pointed towards the latest developments in CO<sub>2</sub> separations using porous solids or metal–organic frameworks.

## 2. Key Challenges for CO<sub>2</sub> capture

The central challenge for gas separations materials is that the differences in properties between the gases that have to be separated are relatively small.

Novel concepts for capture therefore require a molecular level of control that can take advantage of differences in the chemical reactivity of the gas molecules. A further challenge is that the selectivity of a separate procedure is influenced by a combination of adsorption and diffusion selectivity, which are partnered off with most materials. For instance, the first appearance of a functional group that specifically binds one species, and improves on the adsorption selectivity, will simultaneously decrease the diffusion of these particles. The need, therefore exists to design materials in which one can independently tune the diffusion and adsorption selectivity at the molecular level. The capture of CO<sub>2</sub> from ambient air has also been adduced as a strategy to reduce atmospheric CO<sub>2</sub> levels, particularly with respect to small and mobile emission sources. Distinctly, the low concentration of CO<sub>2</sub> in air (0.04 %) presents a significantly higher thermodynamic barrier to get compared with post combustion methods, while the expense of running great masses of air through an absorbing material presents a further challenge. Although current post combustion capture technology is suitable for the retrofitting of existing plants, the energy penalty for a coal-fired power station using traditional aqueous monoethanolamine (MEA) for capture has been estimated at 25 to 40 %.

### **3. Conventional Chemical Absorption**

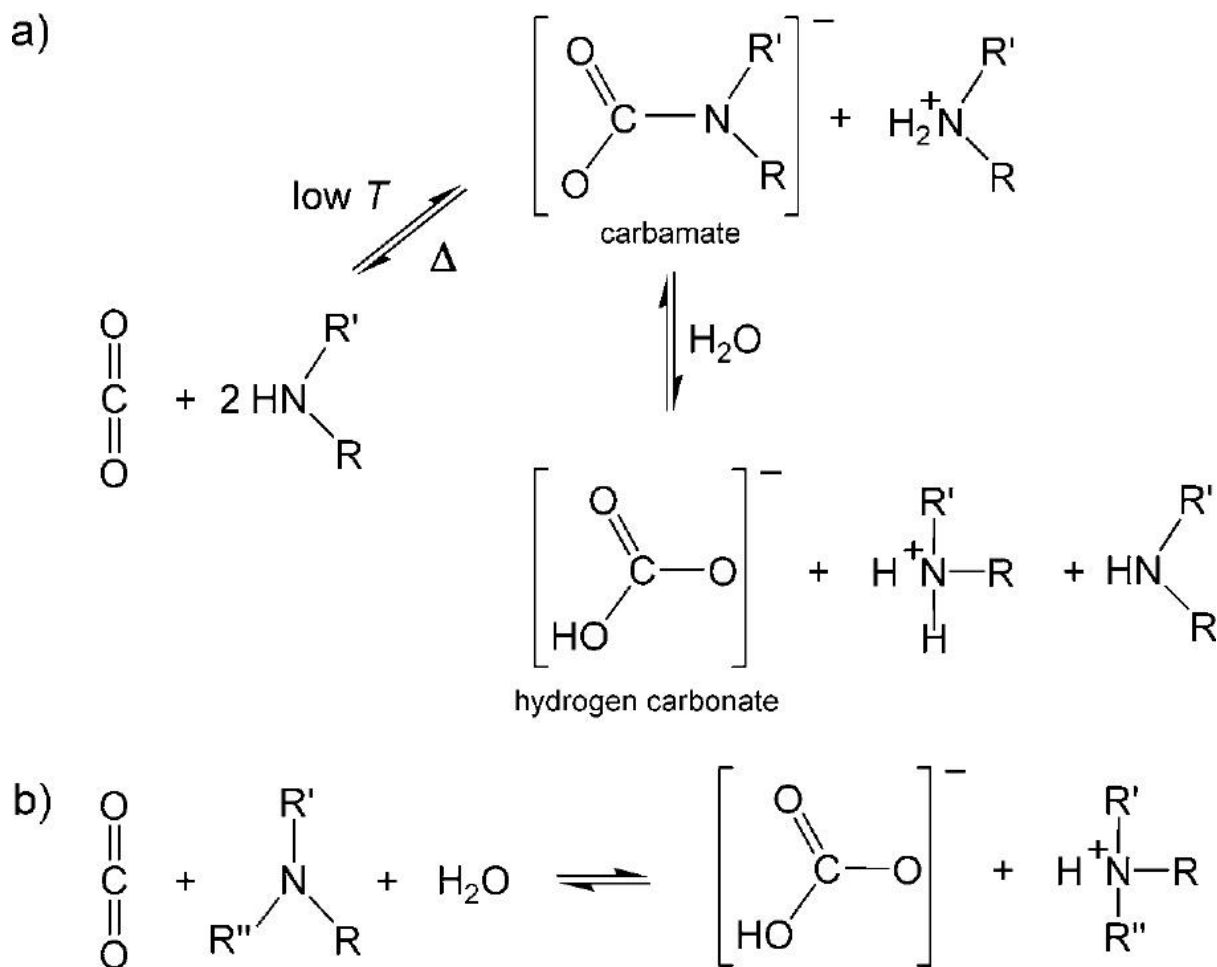
One technology suggested to get rid of CO<sub>2</sub> from the air, is the soda lime process. This calls for drawing air through a sodium carbonate solution and then calciner, to separate out CO<sub>2</sub>, which can be stacked away. But the scale that would be

needed to lessen the concentration of CO<sub>2</sub> in the atmosphere by simply a few parts per million is almost inconvincible because of the depleted degree of CO<sub>2</sub> and the vast intensity of atmosphere that would need to be treated.

Most of the world's existing fossil fuel power plant operates by firing the fuel in the melodic line and then practicing the vitality produced by that combustion to drive turbines. To capture CO<sub>2</sub> from these power plants, it is necessary to split up the CO<sub>2</sub> from exhaust gases after the combustion process, through a process called post combustion capture or PCC.

“Wet-scrubbing” CO<sub>2</sub> capture technologies have been used industrially for over 50 years and are based predominantly on the industrially important primary alkanolamine MEA. The procedure requires the transition of an aqueous amine solution (typically 25–30. %) down the crest of an absorption tower, while a gaseous stream of flue gas containing CO<sub>2</sub> is put in at the tail end. A blower is needed to pump the gas through the absorber. At a temperature of around 40<sup>o</sup>C, the reaction of CO<sub>2</sub> with the amine occurs through a zwitterion mechanism to form carbomates, a response that has been extensively analyzed.





SCHEME 1: General reaction schemes for the chemical absorption of CO<sub>2</sub> by a) primary or secondary and b) tertiary amine-containing solvents  
Adapted from Angew. Chem. Int. Ed.2010, 49, 6058 – 6082.

The liquid amine CO<sub>2</sub>-rich solvent passes from the absorber column to a stripping tower where the mix is heated with steam to release the CO<sub>2</sub>. The regeneration of the chemical solvent is carried out at elevated temperatures (100–140°C) and pressures not much higher than atmospheric pressure. The high heat of formation associated with carbamate production leads to a considerable energy penalty for regeneration of the solvent. Following positive feedback, the amine solution is cycled back to the absorption tower for additional CO<sub>2</sub> absorption.

The principal advantages of the post combustion method are that it is commercially mature and can be easily retrofitted into existing power plants, they have a number of drawbacks. These include the considerable energy requirements for solvent regeneration and the necessary use of inhibitors to control corrosion and oxidative degradation due to residual oxygen in the flue stream. The sensitivity of the solvents to chemical degradation from other by-products in the flue gas streams, such as SO<sub>x</sub> and NO<sub>x</sub>, also lead to reduced efficiencies and increased costs for electricity production. Improved strategies for postcombustion capture include the use of liquids with lower heats of adsorption, increasing the concentration of the adsorbent molecules and improving the mass transfer and reaction kinetics. The use of other amine-based molecules with lower regeneration temperatures have thus been considered for chemical absorption. Secondary amines such as diethanolamine (DEA) possess a lower heat of reaction compared with primary amines: the lower stability of the carbamate formed upon CO<sub>2</sub> absorption gives rise to a more economical regeneration step for the secondary amines.

The CO<sub>2</sub> loading capacity for primary and secondary amine lies in the range 0.5-1 mol of CO<sub>2</sub> per mol of amine, since a fraction of the carbamate species is hydrolyzed to form hydrogen carbonates. The reaction of CO<sub>2</sub> with tertiary amines such as N-methyldiethanolamine (MDEA) occurs with a higher loading capacity of 1 mol of CO<sub>2</sub> per mol of amine, albeit with a relatively lower reactivity towards CO<sub>2</sub> compared with the primary amines. The carbamation reaction

cannot proceed for tertiary amines. MDEA is commonly utilized for natural gas treatment and exhibits lower solvent degradation rates in addition to a low energy penalty for regeneration of the solvent in the stripper. Addition of a small amount of primary and secondary amine enhances the CO<sub>2</sub> absorption rates for tertiary amines. The steric hindrance and basicity of the amine are the major factors controlling the efficiency of CO<sub>2</sub> capture reactions. Sterically hindered amines such as 2-amino-2-methyl-1-propanol (AMP) which contain bulkier substituents have been identified as the most promising absorption solvents due to the lower stability of their carbamates.

(carbamate stability constant: AMP, 0.1 < DEA, 2.0 < MEA, 12.5 at 303 K).

Inorganic solvents such as aqueous potassium and sodium carbonate as well as aqueous ammonia solutions have also been considered for chemical absorption. The chilled ammonia process for CO<sub>2</sub> capture involves the reversible formation of ammonium hydrogen carbonate, with the forward reaction to capture CO<sub>2</sub> as solid NH<sub>4</sub>HCO<sub>3</sub> occurring at temperatures below 20°C.

Despite the strongly absorbing nature of the solutions, the large energy demands of the regeneration step present a comparable problem with that taken in post combustion CO<sub>2</sub>.

#### **4. Emerging Methods for CO<sub>2</sub> capture**

Three strategies for incorporating capture into power generation scenarios are of primary focus today: post-, pre- and oxy-combustion capture.

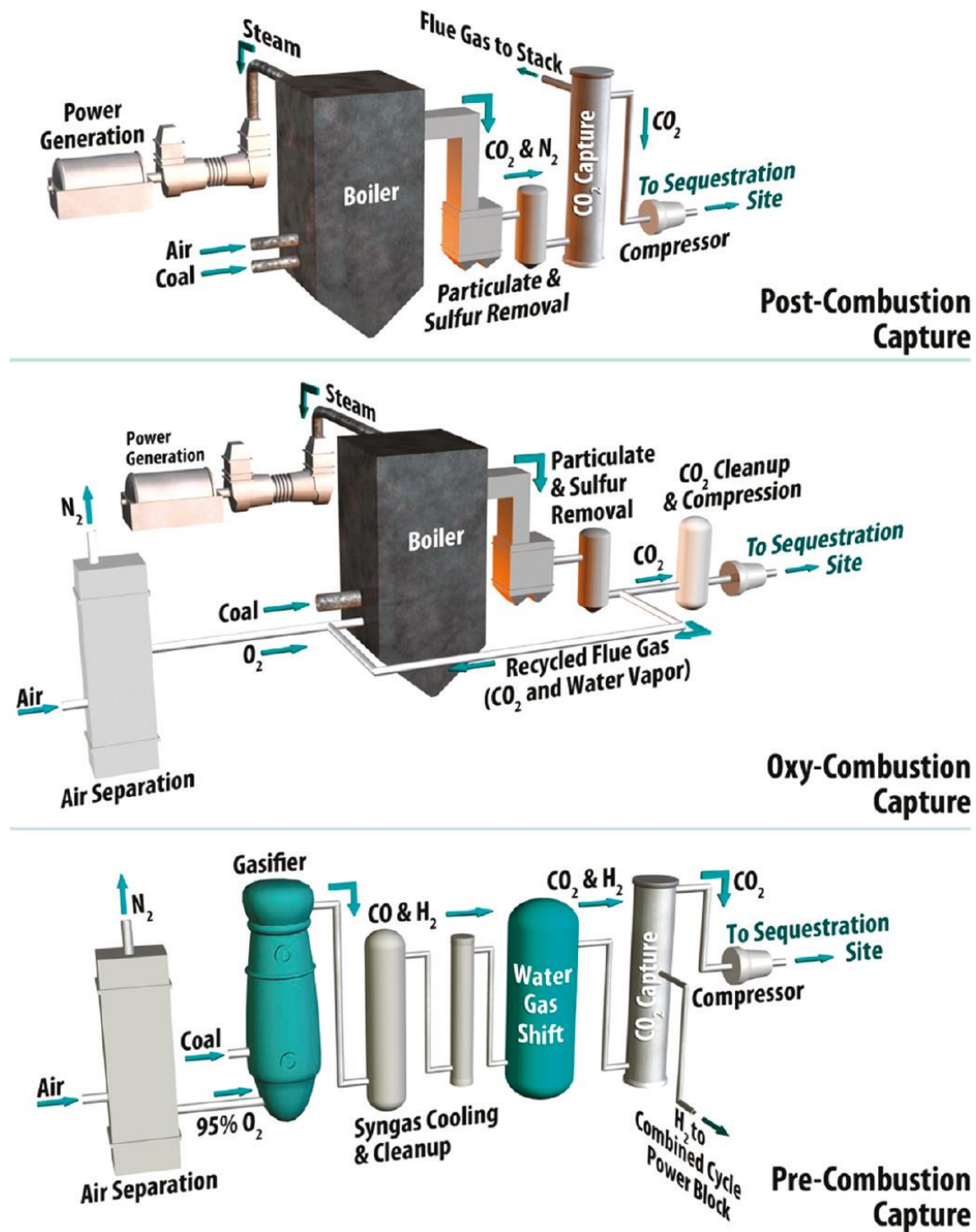


Figure 2: Post-, oxy-, and precombustion concepts and separation system integration into power plants.

Adopted from Environ. SCI. Technol.2011, 45, 8597–8604

#### 4.1. Physical Absorbents

Monoethanol amine is a commonly used physical adsorbent, which selectively binds CO<sub>2</sub> at high partial pressure and low temperatures, in accordance with

Henry law. Selexol and Rectisol are industrially used physical adsorbent for 40 years for natural gas sweetening and the treatment of synthesis gas. Selexol is a mix of dimethyl ethers of polyethylene glycol and rectisol, a methanol chilled to  $-40^{\circ}\text{C}$ . The advantage of this case is the lower heat consumption in the solvent regeneration step and the stripping process can be driven by heat or pressure reduction, called flash distillation. Physical solvents are suitable for  $\text{CO}_2$  capture from high pressure streams, hence mainly used in the post combustion method. Ionic liquids are another class of physical solvents which are also known to be selective towards  $\text{CO}_2$  adsorption. These contain a mix of organic cations and inorganic anions and are viscous liquids at room temperature. These are nonflammable, environmentally benign and can exhibit thermal stability and low vapour pressure.

The mechanism of  $\text{CO}_2$  capture is physisorption with the heat of adsorption around  $-11\text{KJ Mol}^{-1}$ . Hence solvent regeneration is energy economic. For this reason, ionic liquids usually employed for precombustion applications. Some ionic liquids adsorb  $\text{CO}_2$  in a chemisorption mechanism. In addition to these, ionic liquids serve as  $\text{SO}_2$  polishing agent. It simultaneously removes both  $\text{SO}_2$  and  $\text{CO}_2$  since the solubility of  $\text{SO}_2$  is 8-25 times greater than that of  $\text{CO}_2$  at the same partial pressure. Hence an additional step is required to remove  $\text{SO}_2$  from  $\text{CO}_2$ .

The viscosity of ionic liquids minimizes the solvent loss from gas streams, limit mass transfer, hence they suffer from low rates of adsorption. To surmount these shortcomings, task specific ionic liquids have been developed.

Introduction of amine groups into TSIL increases the rate of CO<sub>2</sub> adsorption, at pressures relevant to flue streams (ca. 1bar). Polyaminized ionic liquids also show high selectivity towards CO<sub>2</sub>/N<sub>2</sub> and enhanced CO<sub>2</sub> solubility than the monomeric ionic liquid itself.

## **4.2. Adsorption Materials**

A variety of physical adsorbents have been considered for CO<sub>2</sub> capture which includes microporous and mesoporous materials, metal oxides, and hydrotalcite compounds.

Metal oxides such as CaO and MgO are promising materials for carbon dioxide capture because of their ability to retain high adsorption capacities above 300<sup>0</sup>C. The reaction of CO<sub>2</sub> with calcium oxide produce CaCO<sub>3</sub> at 600-650<sup>0</sup>C. The reverse calcination reaction regenerates the oxide at 800-850<sup>0</sup>C. Adsorbent degradation has been observed after several cycles. Lithium based perovskites have attracted attention for their high carbon dioxide adsorption capacities.

Hydrosilicates and derivatives are also suitable for carbon dioxide capture. Usually these compounds are used in precombustion at temperature above 400<sup>0</sup>C. These compounds belong to a family of layered double hydroxide (LDH'S) with the general formula

$M^{2+}_{1-x} M^{3+}_x (OH)_2 A^{m-}_{x/m} \cdot yH_2O$  where the  $M^{2+}$  is typically  $Mg^{2+}$ ,  $M^{3+}$  is  $Al^{3+}$  and the anion A may be chloride, nitrite or carbonate ions. Recent studies indicates that Ga substituted hydrotalcites adsorb more carbon dioxide than naturally occurring hydrotalcite Mg-Al- $CO_3$ .

“Dry carbonate process” is currently in the stages. In this, flue gas mixes with a solid dry powered carbonate sorbent ( $Na_2CO_3$  or  $K_2CO_3$ ) in a fluidized bed to form corresponding hydrogen carbonate salt. The regenerative decarboxylation reaction can be achieved at a relatively low temperature of  $120^{\circ}C$ .

#### **4.2.1. Zeolites**

Zeolites are the most widely reported physical adsorbate for  $CO_2$  capture. The main functions that significantly influence the carbon dioxide capture performance involved basicity, pore size distribution, electric field strength stemming from exchangeable cation of zeolites, water vapor in gaseous streams (hence they need a high regeneration temperature) and Si-Al Ratio.

Normally used zeolites are X and Y Faujasite system. Some synthetic zeolite such as 5A and 15X are also used as physical adsorbents.

We can improve the adsorption capacity of zeolite by incorporating alkyl amine on to their internal surfaces, which in turn improves the gas sorption properties for low pressure capture applications. Hence we can use this method for capturing carbon dioxide from the atmosphere.

Common zeolites used for carbon dioxide capture are given below

- Polyethylamine impregnated MCM-41 (24 fold enhancement in the CO<sub>2</sub> adsorption)
- Amines immobilized in support such as poly (methyl methacrylate) - suffer from lack of stability over repeated cycles
- Alkyl amine covalently tethered to the surface of mesoporous support. For example, Aziridine polymerization at silica, which exhibit reversible carbon dioxide binding and multi-cycle stability under simulated flue gas conditions using a temperature swing adsorption (TSA) approach.

Type of amine tethered or surface density of an amine group influences the adsorption capacity. SBA-15 grafted with monoamino, diamino, and tetraamino ethoxysilanes will give an adsorption of 0.52, 0.87, and 1.10 mmol CO<sub>2</sub>/g adsorbent, respectively.

#### **4.2.2. Carbonaceous Adsorbents**

Porous carbon are playing an important role in CO<sub>2</sub> sorption and separation for a long time due to their porosities, excellent stabilities, wide availabilities. Numerous studies are going on adsorption studies of different types of carbon forms like activated carbon, molecular sieve carbon, heteroatom doped porous carbons and nanocarbons. Out of which, heteroatom-decoration continues to dominate in this area. Another marked trend is the vast range of nanostructured carbons (graphite and CNT) as CO<sub>2</sub> sorbents which are being created. The carbonaceous materials are inexpensive compared to other solid adsorbents like zeolite 13X.



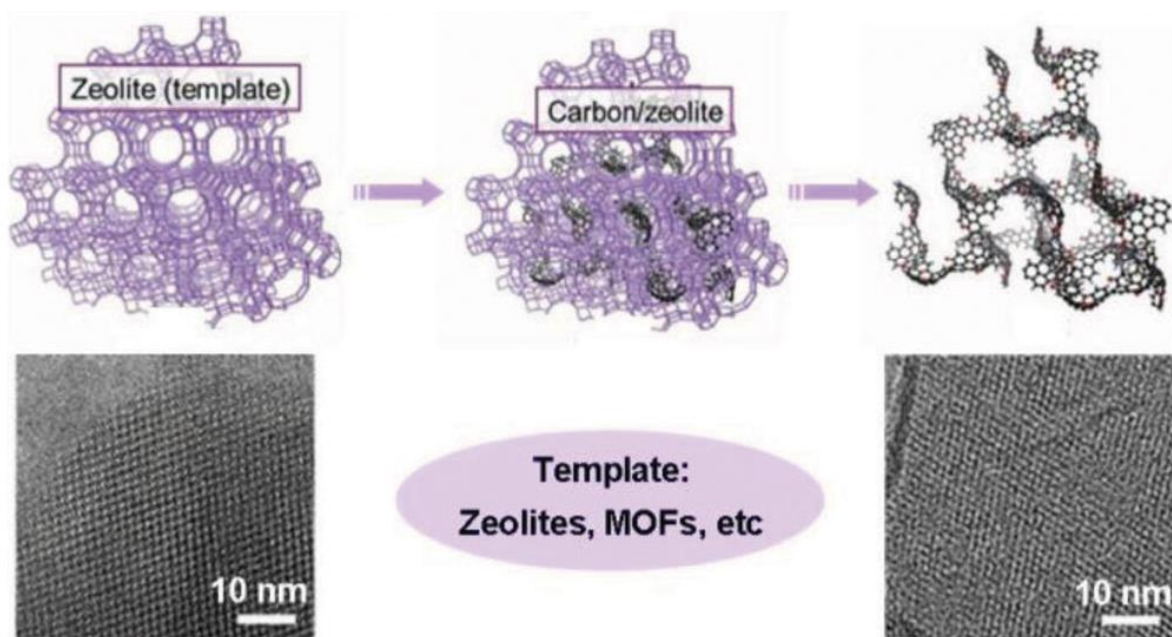


Figure: The synthesis of molecular sieve carbon through nanocasting pathway: the sacrificial template including crystalline zeolites and MOFs.

Adopted from Annu. Rep. Prog. Chem., Sect. A: Inorg. Chem., 2013,109, 484-503

### 4.2.3. Metal Organic Frameworks (MOFs)

Are microporous crystalline solids which composed of organic bridging to metal base nodes to make three dimensional extended network with a uniform pore diameter typically in the range 3 to 20Å°.The nodes generally consist of one or metal ions to which the organic bridging ligands coordinate through a specific functional group like carboxylate and pyridyl.

Unique structural properties of MOFs include robust, high thermal and chemical stabilities, unprecedented internal surface areas, high void volumes and low densities (from 0.21 to 1.00 g cm). It takes in an ability to modulate systematically the pore dimensions and surface chemistry within metal–organic fabrics. The high surface area-to-weight ratio of MOFs is such that they have enhanced

capacities for CO<sub>2</sub> capture at moderate pressures. Compared with activated carbon and zeolites, it delivers a high active surface area per unit weight (1500-4500 m<sup>2</sup>g<sup>-1</sup>).

The framework [Zn<sub>4</sub>O(btb)<sub>2</sub>] with a surface area (SA) of 4500 m<sup>2</sup>g<sup>-1</sup> exhibits the highest capacity for CO<sub>2</sub>, taking up 33.5 mmol g<sup>-1</sup> at 32 bar. By comparison, the benchmark material zeolite 13X adsorbs 7.4 mmolg<sup>-1</sup> at 32 bar.

The isotherms of terephthalate based materials exhibited an interesting S-shaped profile in which the presence of inflection points indicated electrostatic interactions between the CO<sub>2</sub> molecules themselves, which dominate as the pressure increases.

Measurements on the adsorption kinetics for CO<sub>2</sub> in [Zn<sub>4</sub>O(bdc)<sub>3</sub>] showed that the activation energy for diffusion is directly proportional to the ratio of the kinetic diameter of the diffusing molecule to the pore opening of the structure. Grounded on this rationale, frameworks with smaller pores poses a higher barrier to diffusion. A molecular sieving effect can be exploited for the separation of gas mixtures in cases where the pore sizes are sufficiently small, and are in the range of the kinetic diameters of the adsorbates. In accession to such kinetic size effects, favorable electrostatic interactions between CO<sub>2</sub> and the framework also provide enhanced capture, as in the case of [Al(OH)(ndc)] (2.3 mmolg<sup>-1</sup>, 10 wt % ,303 K, 5 bar). Gas uptake measurements revealed that while CO<sub>2</sub> (kinetic diameter 3.3 Å) is excluded from entering the 3.0 Å diameter pores, favorable interactions between the hydroxy groups of the framework protruding toward the 7.7 Å

diameter channels give rise to preferential CO<sub>2</sub> adsorption over N<sub>2</sub> (by a factor of ca. 10).

The majority of adsorption studies has reported selectivities based on single component adsorption isotherms, IAST (Ideal Adsorbed Solution Theory) serves as the bench mark for the simulation and computational analysis of binary mixture adsorption from the experimental isotherms.

Zeolite Imidazolate Frameworks (ZIFs) constitute a subclass of metal–organic fabrics that can adopt zeolite structure types based on the replacement of (1) tetrahedral Si<sup>4+</sup> and Al<sup>3+</sup> ions with tetrahedral transition metal ions such as Zn<sup>2+</sup> or Co<sup>2+</sup> and (2) bridging O<sup>2-</sup> ions with bridging imidazolate-based ligands. High-throughput synthetic methods have been used with great utility to produce ZIFs with a range of pore aperture metrics. Functionalization of the imidazolate and benzimidazolate linkers was also established to permit fine-tuning of the interactions between the pore walls and guest molecules, thereby varying the selectivity of adsorption. Significantly, in contrast to many metals–organic frameworks, ZIFs exhibit high thermal stabilities and chemical stability in aqueous and organic media, which are needed for practical separations processes. In especial, the stability of a framework toward long-term exposure to water vapor is a vital event in determining its suitability for CO<sub>2</sub> capture from flue gas.

#### 4.2.4. Membranes

Membranes, acting like a very fine sieve, can preferentially remove CO<sub>2</sub> from a stream of mixed oxide gases. They are normally made up of polymer or ceramics and can likewise be utilized in conjunction with liquids. Membranes are in many industries, including for natural gas separation, for the production ammonia and hydrogen and for the separation of nitrogen gas from the atmosphere. Yet at present they are not practiced on a big scale for separation of CO<sub>2</sub> from flue gases. There are two main type of membrane systems that can be used to capture CO<sub>2</sub>. One separate out the CO<sub>2</sub> from other gases (gas separation membranes); the other allows CO<sub>2</sub> to be adsorbed from a gas stream into a solvent via a porous membrane barrier (membrane gas absorption).

Gas separation membrane system has a key advantage over solvent systems: the equipment is smaller, more versatile and involves no solvent. This cuts material cost and reduces environmental concerns. The membrane acts as a semipermeable barrier through which CO<sub>2</sub> passes through more easily than do other gases. The CO<sub>2</sub> is driven across the membrane by a pressure difference. The energy required to maintain these pressure difference represents a large proportion of the operating cost of the technology at the present time.

Early membranes for CO<sub>2</sub> separation were made from cellulose acetate, a natural plastic made from wood or cotton. Generally glassy polymers are good at separating CO<sub>2</sub> from other gases, but overtime, they tend to become less effective, which is a challenge when using membranes with wet flue gases. Composite

membranes, with a combination of glassy and rubbery polymer segments, combines the structure of the glassy polymer (giving good selectivity for CO<sub>2</sub>) with the higher permeability of a rubbery polymer (allowing more gas to flow through). But like all polymeric membranes, they do not operate well at high temperatures; ceramic membranes, which are able to handle higher temperatures, represent an alternative approach.

Membranes can be used in conjunction with solvents, with the membrane separating the flue gas from the liquid solvent, thereby reducing flow problems that occur when the liquid and gases meet. The CO<sub>2</sub> passes through the membrane and is absorbed by the solvent. However this may lead to a smaller contact surface area between the liquid and the gas, limiting the absorption efficiency compared to other solvent absorption techniques. The CO<sub>2</sub> is then removed from the solvent by heating. Using membranes in conjunction with solvent can reduce the size of the equipment required to absorb the CO<sub>2</sub>.

Depending on the purity required, membrane separation at a coal powered station may need to be a multistage process, with different membranes needed to separate different gases. The separation process needs a pressure or concentration difference to be maintained between the two sides of membrane, which requires energy. More research and development is required to enable membranes to be used at higher temperatures, with lower energy requirements and to enable them to be upscaled to the necessarily massive scale that will be required. Nonetheless, membranes are a promising technology for CO<sub>2</sub> capture.

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**For this assignment, we took outline of the article [1]. Rather than copying the article we add some more details and figure to it and verbalize it in our own words.**