

Nanostructured Solutions for Global Decarbonization: A Comprehensive Analysis of Metal-Organic Frameworks (MOFs) in Direct Air Capture (DAC)

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Abstract- When atmospheric carbon dioxide (CO₂) concentrations increase rapidly due to human activities, the need for scalable carbon removal technology has grown considerably. Direct Air Capture (DAC) is a successful negative emission approach which is able to remove CO₂ by extracting it from the air and cleaning molecules to its target level even in an atmosphere. However, due to very low atmospheric CO₂ concentrations (~400 ppm) and moisture, for DAC materials to be used, high selectivity and stability are essential attributes and low regeneration energy. To improve the application of DAC in dilute environments, metal-organic frameworks (MOFs) are a new class of very porous nanomaterials whose unique chemical and structural properties have made them promising for DAC applications. Functionalized MOFs (i.e., amine-grafted materials and hybrid ultra microporous structures) exhibit improved adsorption performance and are able to capture CO₂ even in humid conditions. Recent progress of MOF-based DAC technology in solid sorbents and membranes, and the relationship of molecular structure and CO₂ capture efficiency, also contribute to this. In this review, we also discuss the best computational and machine learning approaches for rapid screening and optimization of MOFs to ensure high-performance materials from a wide range of chemical compounds. Many challenges such as moisture sensitivity, high synthesis costs, structural instability, and energy-intensive regeneration are among the critical difficulties for MOF-based DAC systems to be achieved at large scale. To eliminate such limitations, innovative design solutions such as surface functionalization as well as scalable synthesis routes with high productivity of materials, and process integration should be adopted. Nanostructured MOFs represent a new path to global decarbonization with efficient and flexible DAC systems. Further interdisciplinary research in materials science, process engineering, and techno-economic analysis will translate laboratory results into carbon capture technologies that are compatible with net-zero climate targets at the scale of commercial production.

Keywords: Direct Air Capture (DAC), Carbon Dioxide (CO₂) Removal, Negative Emission Technologies, Metal-Organic Frameworks (MOFs), Functionalized MOFs.

I. INTRODUCTION

The increasing CO₂ concentration, driven most of all by humans through fossil fuels, deforestation, and industrial processes, is one of the greatest environmental problems of the 21st century. That carbon dioxide concentration is also above 420 ppm, significantly surpassing pre-industrial levels, and it contributes to global warming, ocean acidification,

and climate instability. We should be concerned in the wake of global efforts to reduce it in climate change which have not made enough progress but have not taken measures to keep it away from global temperatures according to the Intergovernmental Panel on Climate Change and the Paris Agreement. There is an urgent need to improve NETs which would efficiently remove CO₂ from air and be able to provide a reduction to greenhouse gas.

Among many NETs, direct air capture (DAC) has been shown to be a very promising and adaptable way. Instead of carbon capture technologies aimed at point sources (power plants) on earth, there is a natural option of pulling the gases in air and using local sources in a system directly (water vapor to create DAC) by plugging it into an appropriate area to effectively store, or access to, the carbon gas or carbon with the existing infrastructure and its energy source in the first place. DAC is an intensive work and involves high development of highly efficient and sustainable materials and methods of the process is very necessary as these in our current state are already quite demanding to get.

Nanostructured materials have revolutionized the science of gas capture/separation where we have gained unprecedented control over physicochemical conditions at this level. Metal-organic frameworks (MOFs) are of special interest and have a variety of structure and function. These materials usually contain metal ions or clusters linked by organic ligands in a crystalline coordination structure. As a result, they are highly porous and ordered three-dimensional networks. High surface areas (often exceeding $5000 \text{ m}^2 \text{ g}^{-1}$), tunable pore sizes, and many different chemical properties make MOFs of high relevance in gas adsorption applications like DAC.

The potential effectiveness of MOFs can be even higher due to their tunability. By rational design in chemistry and then by modification after synthesis, we can tailor their composition such that these MOFs are strongly and selectively towards CO_2 molecules. By combining amine functional groups with formation chemistry of CO_2 through chemisorption in MOFs, we build carbamate MOFs which are highly selective and not susceptible to low partial pressures - for example, the high formation of adsorption energy when using amines. Furthermore, the presence of open metal sites and polar functional groups facilitates physisorption as well as electrostatic interactions and quadrupole-dipole forces. Such as ultramicroporous MOFs (pore size $< 0.7 \text{ nm}$) have also enabled selective adsorption of CO_2 molecules, which would also allow for selective capture of CO_2 with different MOF structures.

A very similar aspect of the MOF-based DAC systems is the possibility that the dynamic environments of these systems are realistic (hydrostatic), such as humidity (climate). Water vapor can compete with CO_2 for adsorption as well as degrade some MOFs as an efficient product of an adsorption mechanism. As such, recent developments have resulted in water-stable MOFs and hydrophobic frameworks, but also hybrid materials that maintain high CO_2 uptake in humid air. Functionalization techniques such as grafting amines or by adding hydrophobic linkers for hydrophobic MOFs are providing positive evidence that MOF-based DAC in humid environments can improve the MOF and its adsorption performance.

Even beyond material design, we have made significant progress in understanding the structure-property relationships leading to CO_2 capture in MOFs. New characterization methods and computational techniques (density functional theory and machine learning) are being widely adopted to predict the adsorption behavior and to screen large databases of MOFs. It would seem that the rapid identification of promising candidates can be useful for the development of efficient DAC materials as a whole. However, many hurdles still need to be overcome to move MOF DAC technologies from the laboratory to the field of industrial applications. One is the high cost of synthesizing MOF compounds, the scalability issues and the requirement of energy-efficient regeneration processes.

The long-term stability of cyclic adsorption-desorption and environmental contaminants must also be addressed. All material innovations of the design and development of a DAC model and techno-economic and engineering aspects of the design and operation should be taken into account in order to overcome them. On a general note, the use of nanostructured MOFs in DAC systems is the key step toward global decarbonization. They are very tunable, have high adsorption capacity, and can be used to capture CO_2 in an ambient environment, which makes them the material of choice for the next generation of carbon capture technologies. Thus further research on MOF technologies can help us to overcome the shortcomings of current MOF

development and build on MOF solutions to the next generation of carbon capture in order to create sustainable communities on Earth.

II. ADVANCED ARCHITECTURAL CLASSIFICATION OF MOFS IN DAC.

The strength and the utility of metal-organic frameworks (MOFs) in direct air capture (DAC) are characterized by their architecture and their function as well. Many recent MOFs are currently developing which have unique physicochemical properties, thus providing easier and affordable CO₂ capture when it comes to high atmospheric quality conditions. All MOF architectures are widely classified by pore structure, functionalization, hybridization, and application-based design strategy.

2.1 Pristine (Conventional) MOFs

Pristine MOFs are the first class of frameworks made with metal nodes and organic linkers that have added functionality but do not change material properties. These MOFs have very high surface area and their pores have complex structures, making them especially suited for physisorption-based capture of CO₂. However, in DAC processes, pristine MOFs tend not to perform well as very weak interactions with CO₂ that will remain under low partial pressures are present. This adsorption rate is dominated by van der Waals forces, which still do not allow efficient capture of CO₂ at low atmospheric levels. Pristine MOFs represent our starting point as they possess sufficient functionalization but need further architectural and chemical improvements as well.

2.2 Amine-Functionalized MOFs

Amine-functionalized MOFs are by far the most studied models for DAC because of their strong chemisorptive interactions with CO₂. The use of amine groups (e.g., -NH₂, -NH-, -NH₃⁺) increases CO₂ affinity strongly, and this is accomplished by providing carbamate or bicarbonate sites or by introducing carbon isotopes with the specific presence of the amines and carbonic acid (nanoparticle compounds).

Amine-functionalized MOFs are produced by various methods, such as in situ functionalization by amine-filled ligands, post-synthetic modification, and physically impregnating amines into the platform. These methods improve their performance significantly in that they achieve high selectivity for CO₂ even in very low concentrations and make adsorption more durable even in humid situations. In addition, the addition of amine groups adds high energy interaction between CO₂ molecules and the platform, leading to strong and efficient absorption of the molecules. This kind of MOF is then beneficial for the Direct Air Capture (DAC) of CO₂, as it compensates for weak physisorption with strong chemical binding that is needed to absorb CO₂ from a frame.

2.3 Both Ultramicroporous and Pore Engineered MOFs

Ultramicroporous MOFs having pore sizes less than 0.7 nm are very effective for CO₂ capture and utilize molecular sieving and confinement effects. In these structures, gas molecules can be selectively adsorbed based on their size and quadrupole interactions at a very small pore size and the CO₂ molecules are more selective compared to other atmospheric gases. The confinement of these small pores, which is very tight and strong to the pore walls, allows the CO₂ molecules to be adsorbed more effectively so that they are more absorbed in these pores even when they are very small. The competition from other gases such as N₂ and O₂ is also reduced and the CO₂ molecules are more selective. Pores are developed for selective adsorption using pore size, geometry, and topology.

2.4 Hybrid and Composite MOFs

Hybrid MOFs are formed by linking the standard MOF structure with other types of materials (polymers, carbon-based materials, nanoparticles, metal oxides) in order to overcome some of the known drawbacks of pure MOFs such as stability, processability, or scalability. The structural composition of such composites makes MOFs more powerful and suitable for concrete applications. More common hybrid systems are MOF-polymer mixed matrix membranes (MMMs), MOF-carbon composites, and MOF-metal oxide hybrids. They

provide stronger and more robust performance at the ground of operation. They are also resistant to humidity and are therefore useful in direct air capture (DAC) applications. So hybridization can be a very promising tool for bridging the gap between the high performance of MOFs on our laboratory scale and actual use in industrial DAC.

2.5 MOF-Based Membrane Architectures

MOF-based membranes are an emerging class of architectural materials that have remarkable advantages in gas separation, particularly in direct air capture (DAC) systems. Unlike adsorbent-based membranes based on cyclic adsorption and desorption processes, these membranes can be continuously separated and are therefore energy-efficient and operationally stable. With different membrane types depending on how we are building, MOF-based membranes can be subdivided: pure MOF membranes, mixed matrix membranes, and thin film composite membranes for structural and functional reasons. Gas separation through membrane systems is not only easier and energy efficient with no regeneration process but can continue in a continuous cycle with a long storage capacity.

Additionally, the membranes of amine-functionalized MOF can perform better in terms of separation, in that their adsorption and dispersion mechanisms are comparable with the ones used in the atmosphere in MOF membranes. The presence of amine is one of the main contributors to CO₂ affinity even at low temperature: the membrane is also porous so a selective transport process can take place which provides the highest capture efficiency. Overall, a framework for the creation of MOF-based DAC technologies can be made feasible.

2.6 Isorecticular and Topology-Controlled MOFs

Imorphic MOFs (IRMOFs) are designed by continuously arranging the organic linkers without changing the underlying framework structure. This allows the pore size, shape and the chemical environment to be selected but does not influence the physical structure of the material at all. Hence, the adsorption process of MOFs can be tuned to be selective and efficient in adsorption which makes

them very well suited for applications such as direct air capture (DAC). The advantage of IRMOFs is they allow rational pore structure for pore design, while their adhesion ability, such as selectivity, capacity, and their interaction strength with CO₂ molecules are fine-tuned. Therefore, the desired MOFs can be optimized for DAC conditions in the low CO₂ concentration and with other gases.

Furthermore, this same concept has now become the reality in topology-controlled MOFs where the framework was designed, carefully, to provide particular connectivity channels of metal to linkers, and as a result the gas diffusion pathways as well as adsorption energy distribution have been tailored to improve the efficiency of CO₂ capture. In a holistic approach, this and another isometrical and topological design strategies could be effectively used to develop new MOFs with optimal performance and versatility for DAC.

2.7 Zeolitic Imidazolate Frameworks (ZIFs)

Zeolitic imidazolate frameworks (ZIFs) are a different class of MOFs. They have zeolite-like structures and chemical stability. They are formed from metal ions such as Zn²⁺ and Co²⁺ with imidazolate linkers in highly thermally stable and strong structures, so are suitable for long-term environments. In the field of Direct Air Capture (DAC), ZIFs have many interesting facts about the field to know. They are stable in humid environments as well as good gas separation results and are durable and resistant to damage from overuse. They should be reliable for such long-term and lasting activities. They may not have highly functionalized MOFs as much in terms of CO₂ adsorption capabilities for the whole process, but their durability and toughness of operation as well as simple and fast transport make them particularly practical and versatile in the implementation of DAC.

2.8 Nanoengineered and Hierarchical MOFs

More recently, nanoengineered MOFs with hierarchical porosity (including the addition of micro-, meso-, and macropores) were introduced within our generation. This method can dramatically enhance mass transfer as it provides an efficient approach for gas diffusion, and hence the accessibility of active adsorption sites can be

improved. The structure thus successfully alleviates diffusion limitations in traditional MOFs, and it has proven an effective adsorption-desorption process, achieving better performance in the DAC.

III. KINETIC ANALYSIS AND COMPETITIVE ADSORPTION

The performance of MOFs in DAC is influenced primarily by adsorption and competitive molecule attraction in the air environment. In contrast to laboratory conditions, local atmospheric air should be dense in nitrogen (N₂), oxygen (O₂), and water vapor (H₂O). Humidity is traditionally considered the main inhibitor of CO₂ capture efficiency in the room environment, but water molecules have a higher attraction to MOF sites than the other gases to be adhered to (see Fig.). MOF adsorption can be hindered by an intense level of pressure to the MOF, and water can lead to the formation of pores or wall formation, which in some cases of organic compounds and framework hydrolysis.

Since the adsorption capacity of MOFs is low in the experimental settings, in the present environment, efficient DAC can only be created in an efficient energy structure to give a fast rate of adsorption and desorption. The rate of diffusion of CO₂ to the porous structure of MOFs depends on their size, connectivity, and shape. So the pore channels are narrow in most MOFs such that the adoption of large CO₂ atoms will take place slowly, and time takes place when compared to its diffusion as a protein. With the development of MOFs by nanoengineered and hierarchical design based on multidimensional porosity, it is possible to use multi-scale porosity to provide a large scale in such a situation. The mass of CO₂ molecules moving at high concentration levels is carried quickly at a high rate in these MOFs, and that does not take place in a low concentration state, so the adsorption of CO₂ in the active MOF is faster as the adsorption and desorption is slower than in more concentrated and dense MOFs. Low concentration structures have higher adiabatic and non-active concentration materials.

In order to quantitatively measure the selectivity of MOFs in multi-component gas systems, the Ideal

Adsorbed Solution Theory (IAST) is widely used to predict this and others' adsorption response based on single-component isotherms and provides a measure of how selectively a material adsorbs one gas over another. A single molecule's selectivity is given by:

$$SCO_2/N_2 = (yCO_2/yN_2)/(xCO_2/xN_2)$$

Where xCO_2 and xN_2 represent the mole fractions of CO₂ and N₂ in the adsorbed phase, and yCO_2 and yN_2 are their mole fractions in the gas phase. A higher selectivity value means a better MOF will obtain for CO₂ rather than other gases.

Recent work has focused on the design of MOFs with water less attractive and more competitive adsorption. Hydrophobic MOFs in the form of alkyl chains or fluorinated linkers prevent water adsorption and maintain active sites for CO₂ collection. To date, these MOFs have found CO₂/N₂ selectivity up to 200 and can also be used quite efficiently and economically in realistic conditions. Additionally, amine-functionalized MOFs will also have strong chemisorptive interaction with CO₂ and, in the presence of water, also enhance the selectivity. In general, knowing the interaction between adsorption kinetics and competitive adsorption is important for controlling MOF performance in DAC systems. Designers of advanced materials together with theoretical models like IAST offer an excellent training ground for next-generation sorbents that work efficiently under environmental stress and require appropriate techniques to prepare them with various operating scenarios.

IV. INDUSTRIAL SCALING: THE "PELLETIZATION" PROBLEM

For DAC-based MOF systems on such a big scale, the transfer from laboratory-scale powders with all of the laboratory powdered materials to industrial applications remains a very big puzzle still being solved. MOFs in that state are usually prepared in thin powder form, which is much too fine to be used for large-scale applications because of high pressure drop, poor mechanical stability, and difficulty in handling within reactors (such as air-contacting

units). The solution for converting them into larger-scale forms such as pellets, granules, and membranes is an approach to concrete applications of that kind.

The most common method is pelletization in which MOF powders are merged with binders such as PVDF or alumina to create stable structures of materials, but such a method has some disadvantages as the binder material is prone to block the pores of MOFs leading to a smaller surface area with a reduced adsorption performance (at most 10-20% in the case of MOF). This balance is one of the main challenges facing the production of MOF-based technologies when they relate to its mechanical strength and adsorption performance.

Recently, structural issues have also been tackled with various options. One of the methods could be to use 3D printing to produce MOF-based monoliths with well-modelled shapes. The monolithic structures are more mass transfer oriented, reduce pressure drop, and provide better airflow for large-scale DACs - often air-contacting ones like fans or reactors. They can also be integrated without a binder, and the MOF itself remains intact in porosity and performance to be still used instead. Another important element for industrial use is the CO₂ adsorption regeneration of adsorbed MOFs.

Thermal swing adsorption (TSA) is one of the most widely used regeneration processes in which heat is applied to release stored CO₂ and restore adsorption capacity. However, this process is highly energy-costly (sometimes referred to as "parasitic energy") and can adversely impact DAC efficiency and economics. Thus, MOFs with low heats of adsorption require less energy to be recycled and can store CO₂. Overall, pelletization needs to keep structural integrity, adsorption efficiency, and energy consumption in mind in the optimal formulation. Technology innovations with binder-free structuring, 3D-printed monoliths, as well as the potential for the development of low-energy regeneration materials are needed to reach the ultimate development of MOF-based DACs.

V. COMPARATIVE PERFORMANCE METRICS

The performance of different kinds of material used for the Direct Air Capture (DAC) systems can be compared in terms of breakthrough times, heat of adsorption, hydrolytic stability and scalability.

Zeolite 13X exhibits high breakthrough rate for dry production which indicates high CO₂ adsorption capacity even with its hydrophilic properties, which is negative for humid applications due to the competitive water adsorption. It shows adsorption heat (usually in the range of 35–40 kJ mol⁻¹) and is well-scalable, as a consequence of which such an industrial material should be widely used for commercial use despite its sensitive nature to moisture (Table 1).

PEI-functionalized silica (PEI-silica) exhibits good breakthrough performance but benefits from strong chemisorption interactions and has high heat of adsorption (60–70 kJ mol⁻¹). It is hydrolytically stable and is highly scalable in principle but energy cost for regeneration is higher (Table 1).

Among MOFs, CALF-20 is the most promising one with very successful breakthrough performance and a mild heat of adsorption of 40 kJ mol⁻¹. Its hydrolytic stability is very high and extremely suitable for humid DACs, indicating that commercial deployments of CALF-20 can be expected soon (Table 1).

UiO-66, another well-studied MOF with lower breakthrough performance, has great hydrolytic stability due to its robust zirconium-based framework. Its heat of adsorption in the range 25–30 kJ mol⁻¹ is favorable to low energy regeneration but highly scalable for industrial applications (Table 1).

And while traditional materials like zeolites have great scalability, emerging MOFs such as CALF-20 and UiO-66 offer a better balance between stability, energy efficiency and applications, being excellent products for future DAC (next-generation DAC technologies).

Table 1: Comparative performance evaluation of representative adsorbent materials for Direct Air Capture (DAC) based on breakthrough time, heat of adsorption, hydrolytic stability, and scalability.

Material Class	Breakthrough Time (min)	Heat of Adsorption (kJ/mol)	Hydrolytic Stability	Scalability
Zeolite 13X	High (Dry)	35–40	Poor (Hydrophilic)	Very High
PEI-Silica	Moderate	60–70	Good	High
CALF-20 (MOF)	Very High	~40	Excellent	Commercializing
UiO-66	Low	25–30	Excellent	High

VI. CONCLUSION

The recent development and successful generation of MOF framework such as CALF-20 has turned the corner in MOF carbon capture as MOFs can move from low-level fiddliness laboratory applications to industrial ones. There has been higher levels of hydrolysis stability, high efficiency and long-term stability from such materials available for commercial uses and this development was not achieved in MOFs, but as well as much more than it was expected by previous generations of MOFs owing to mechanical instability of MOFs and moisture sensitivity. It is indeed showing that MOFs are not just a laboratory curiosity any more. They now make for large applications on the global level based on the chemistry of atomic nuclei as CO₂ chains at molecular site clusters or at clusters of organic linkers. So if a structural instability of organic network components, for example (from an isotropic reaction to an oxygen hydrophobic structure for high CO₂ atom) can be established, the adsorption design can be engineered so precisely for the molecules forming the CO₂ clusters.

These traps capture CO₂ efficiently without any loss in the process even at relatively low atmospheric concentrations (parts per million levels) essential for DAC technology. In addition, through some techniques, such as the use of polar groups (e.g. amines), the interactions are functionalized to

achieve strong adsorption (high adsorption in a multi-component air system) while also providing excellent selectivity (good properties of the adsorption process). Modern building forms as well (ultramicroporous, hierarchical porosity and hybrid composites), integrated with the study of a more advanced architecture and the theory of adsorption and selectivity (Adramedi's method) have enabled a more comprehensible understanding of the functional properties of the materials to be more appropriate for MOF planning and optimization. Even these advances can never be completely overcome in terms of scale but are not without the use of technological, and even commercial and industrial methods to obtain the high performance, which the present (e.g. pelletization, energy-efficient regeneration and cost-effective synthesis) process.

However, current solutions such as binder-free structuring, 3D printed monoliths, and low-energy TSA are making significant progress in these areas. Making MOFs of higher heats of adsorption leads to less energy cost and better DAC systems to exist under this scenario in the future. Thus, the evolution of MOFs for long-lasting decarbonization such as CALF-20 demonstrates that rational materials design will go one step further in their design and will help improve the technology available to a lab in the future. Chemistry engineers engineering the coordination sphere make CO₂ and is taking from above to our laboratory the CO₂ that is being removed from our atmosphere. Through continuous cross-disciplinary works on MOFs and a system development process which combines materials and chemical engineering and environment, it can play an important and meaningful role to achieve global decarbonization or towards the sustainability goal of the climate at all.

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