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Review Article

Recent Achievements in Microporous Metal-Organic Frameworks Based on Ionic Liquids/ Deep Eutectic Solvents for Efficient Removal of Toxic Gases and VOCs

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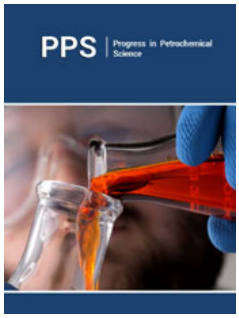

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Abstract

In recent years, micro-porous metal-organic frameworks, often known as MOFs, have received a great deal of interest owing to the remarkable gas storage and separation characteristics that they possess. This literature review investigates the progressive concepts surrounding the integration of Metal-Organic Frameworks (MOFs) with Ionic Liquids (ILs) and Deep Eutectic Solvents (DESs) for the purpose of the effective capture of Volatile Organic Compounds (VOCs). This study provides a full overview of the achievements made in this area by discussing the synergistic effects and increased performance gained via the combination of MOFs with ILs and DESs. These effects were accomplished through the combination of MOFs with ILs and DESs. In addition, the difficulties and potential benefits of the future that are related to this forward-thinking attitude are discussed.

Keywords: Micro-porous Metal-Organic frameworks (MOFs); Integration; Ionic liquids (ILs); Deep eutectic solvents (DESs); Volatile organic compounds (VOCs)

Introduction

Capturing CO_2 can be completed directly with the help of green plants absorbing the sunlight to convert CO_2 and water into diverse hydrocarbons and oxygen through photosynthesis. This fundamental action occurs over a series of connected reactions which are catalyzed by special metallo-enzymes [1]. The main matter of the researchers that made a gradual concern is human-related climate change. In the current issues, the presence of carbon dioxide gas has a great effect [2]. In order to meet the daily demands of humans, fossil fuels which were a consequence of millions of years photosynthetic activity are considered as a convenient supplier. With the beginning of the industrial revolution, the consumption of fossil fuels has increased dramatically. Consequently, the concentration of the CO_2 in the atmosphere increased consequently. During the recent years a huge amount of worries of the researchers and active environmental protection organizations made them concentrate on the universal energy deficiencies and environmental issues [3]. Human society tries to substitute renewable energies instead of fossil fuels and hereby reduce the CO_2 emissions. One of the most favorable solutions is using solar technology which solves both the mentioned problems in CO_2 emission via ascending the atmospheric CO_2 levels and producing cherished compositions as chemical ingredients or fuels for energy consumption [4].

Finding a more stable technique such as photo-redox CO_2 reduction catalysis can be an important challenge [5]. The capture, activation and successive reduction of the CO_2, in a successful CO_2 transformation, requests a super-fit catalyst, which has the ability to reduce through multi-electron transportation and prevention of thermodynamically huge energy consumption [6-7]. Hence, the creature and progress of techniques, in which carbon capture increases are in the socio-economic viewpoint. As an incredible exceptional gas storage source, Metal Organic Frameworks (MOFs) organize an important class in the mesoporous materials [8-10]. Because of the porous nature and tunability of the MOFs, they can store a large capacity of various gasses [11,12]. Also, it can be stated that moreover the potential of the CO_2 conversion in MOFs is extraordinary, they depict a large amount of chemical and thermal stability. This feature of high tunability in MOFs makes them viable in the field of gas treatment [13-15] but the issue of selective separation as an important consideration must be defined which counts as an unanswered subject for MOFs in separation procedure. To overcome the aforementioned problem, mix matrix membranes and functionalization by setting the selective adsorption can be a proper solution [16-18]. Ionic liquids can be applied to metal surfaces for miscellaneous applications [19,20]. Besides the separation power and recycling ability they have a high potential in CO_2 solubility [21] which leads to be a great candidate for gas handling procedures [22].

Likewise, the human is looking for eco-friendly absorbent materials showing high reactivity and sufficient surface region [23]. Despite all efforts in probing green and environmentally friendly techniques in the construction process of MOFs, it still needs more attention [24]. Remarkably, The quick progress in creating, designing and innovating new applications in the field of material science has stimulated the researchers to substitute old fashioned organic solvents for making the functional ingredients to achieve the moralities of Green Chemistry. Deep Eutectic Solvents (DESs) as a fast growing green solvent, via the features of biodegradability, non-volatility, non-toxic and low cost, with the inherent characteristic of open metal sites in the organization of MOF could effectively support the pore size and also modify the pore surface and finally improve the adsorption selectivity of certain toxic gases which are detrimental to the environs [25-27]. So, these newcomer materials can also open up an undeniable consequence in catalytic and photo catalytic degradation of contaminants [28,29]. These green and eco-friendly solvents have been talented as great alternates for non-liquified solvents and room temperature ionic liquid solvents, as Abbott and his team had practical experiments on these moisture stable and unique quaternary ammonium salts that are functional in various applications the same as those previous MOFs [30-32]. In this research, the researcher will focus on the development and forecasts in the fabrication of novel and innovative MOFs using alternatives from green solvents (deep eutectic solvents), simulation of the studied systems with Molecular dynamic calculations and consider the data validation by Matlab software and finally apply the prepared MOFs for the adsorption and remediation of toxic gaseous (especially CO_2 capturing) pollutants in the environment.

Literature Review

In recent years, there has been a substantial uptick in the investigation of micro-porous Metal-Organic Frameworks (MOFs), which are seen as potentially useful materials for the removal of harmful gases and Volatile Organic Compounds (VOCs). The discovery of crystalline coordination polymers in the 1990s led to the conception of Metal-Organic Frameworks (MOFs). These polymers are formed of metal ions or clusters that are coupled by organic ligands [33]. Since that time, a significant amount of research effort has been focused on the design and synthesis of MOFs with structures and characteristics that have been specifically tuned for a variety of applications, including the storage and separation of gases. A more recent development in this area is the combination of Metal-Organic Frameworks (MOFs) with Ionic Liquids (ILs) and Deep Eutectic Solvents (DESs) with the purpose of trapping hazardous gases and Volatile Organic Compounds (VOCs). ILs, which are organic salts with low melting points and have garnered a lot of interest owing to their distinctive features such as low volatility, thermal stability and tunability [34], are salts with low melting points and are classified as organic. On the other hand, DESs are eutectic mixtures that are generated by mixing a hydrogen bond donor (such as a quaternary ammonium salt) with a hydrogen bond acceptor (such as a metal halide) [35]. This process results in the formation of DESs. DESs have earned a reputation for being solvents that are safe for the environment and possess exceptional solubility and selectivity qualities.

When it comes to the removal of harmful gases and volatile organic compounds, a number of studies have shown that mixing MOFs with ILs and DESs may result in synergistic effects and improved performance. The addition of ILs and DESs to MOFs permits enhanced adsorption capabilities, selectivity and stability [36,37]. In addition, the addition of these functionalities provides MOFs with new functions. The interactions that take place between MOF frameworks and ILs/DESs play an essential part in the adsorption and separation processes, which ultimately results in a more effective removal of hazardous gases and volatile organic compounds. In general, the historical development of progressive views on micro-porous MOFs based on ILs and DESs for collecting dangerous gases and VOCs has opened the way for creative methods in gas storage and separation. These visions are based on the idea that micro-porous MOFs might be used to store and separate gases with very small pores. The combination of Inorganic Liquids (ILs) and Gaseous Electron Sources (DESs) with Metal-Organic Frameworks (MOFs) is a viable method to addressing the issues associated with environmental degradation and the need for environmentally responsible gas treatment technologies.

Construction and Synthesis of New MOFs

In order to produce MOFs in the presence of toxicants, researchers have, over the course of many years, used a technique called high-temperature annealing [38,39] that included a lengthy period of time [40]. Surfactants in which precipitate through rapid procedures play a vital function as agents to guide [41], which is a role that is essential for controlling the size and morphological

tunability of MOFs-derived nano crystals. Nevertheless, throughout the course of the last several years, a variety of research concerning the independent competence of MOFs including DESs have been documented. The use of DESs as effective organic linkers will pave the way for the development of organisms composed of very permeable, tunable, and adaptable materials that thrive in favorable environmental circumstances [42-44].

Construction method

The synthesis method that is based on iono-thermal is the most advantageous and environmentally friendly technique to create these novel structures, consisting of zeolite, zeo-type-meso and macro porous material and metal-organic frameworks, which applied ionic liquid or DES as reaction media or structural matters [45,46]. These novel structures consist of zeolite, zeo-type-meso and macro porous material, and metal-organic frameworks. The approach that was just discussed may be used effectively throughout the manufacturing process of DES-MOF-based products. In this study, the researcher's objective is to develop an efficient method for creating a reaction medium out of ionic liquids based on imidazolium. At first look, it seems as if the created MOF would have some restrictions when it comes to the adsorption and catalysis of gases. The next stage is to evaluate these composites in light of recently developed and cutting-edge structures. When it comes to imidazolium based ILs, the components in DESs serve more as supporting ligands than they do as the reaction medium. This results in the development of new MOFs that have more open holes that may be used as efficient materials [47].

The significance of the Advancement of DES-Based Metal–Organic Frameworks

MOFs based on imidazolium ILs for the capture of carbon dioxide

Ionic liquids based on imidazolium have a remarkable potential to trigger and then transform CO_2 into fuels or other suitable compounds, as has been highlighted in a number of papers [48-50]. These liquids may be found in numerous forms. Although in control experiments, the construction of urea and its derivatives, for example, were near to zero in most cases once the imidazolium ILs were eliminated, and the presence of imidazolium ILs in separating CO_2 seems to be accompanied by a high efficiency, ILs are not always environmentally friendly, inexpensive, or simple to prepare.

DES-based metal organic frameworks as a more suitable separator

The preservation of a green and manageable environment should be the primary focus of all research efforts since it is of the utmost importance. As a result, the creation of MOFs based on DESs as a green medium has given an excellent environment for the adsorption and capture of harmful gases throughout the union. Some of the most significant developments pertaining to this matter have been brought to your attention in Table 1 [51-57].

Table 1: Development and application of MOFs based on DESs in toxic gases adsorption media.

Affiliations: ChCl: Choline chloride; PEI: Polyethyleneimine; BN: Boron Nitride; DBT: Dibenzothiophene; btc: 1,3,5 benzenetricarboxylate; EU: Ethyleneurea; PVIm: Polyvinylimidazole.

DES Configuration	Kinds of MOF	Application	Adsorption Capacity	Ref.
ChCl, Urea, PEI	Mesoporous silica	CO_2 capture	51mg/g	[51]
SnCl2, Urea	SnO_2 supported BN	Adsorptive desulfurization	17.93mg/g	[52]
ChCl, EU	$[In_3(btc)_4]_n^{3n-}$	Gas adsorption (N_2, H_2, and CO_2)	70.60cm^3/g	[53]
Zinc nitrate hexahydrate and 2-methylimidazole	ZIF-8	CO_2 adsorption	82.8cm^3/g	[54]
$Fe(acac)_3$, oleylamine, and oleic acid	Fe_3O_4@nSiO	Oxidative desulfurization	9.2mg/kg	[55]
PVIm with halid derivatives	PVIm-Rn-Co	NH_3 uptake	20.1mmol/g	[56]
Imidazole derivative and benzyl halide derivative	Cobaltous thiocyanate $^{2-}(Co(II)(SCN)_4$, TA	NH_3 uptake	13.2-20.1mmol/g	[57]

Application of MOFs

The principal greenhouse gas contributing to climate change, Carbon Dioxide (CO_2), may be captured and stored in Metal-Organic Frameworks (MOFs), which have shown significant promise as useful materials for this purpose. The enormous surface area, controllable pore size and chemical flexibility of MOFs make them promising candidates for CO_2 collection. Incredibly high levels of adsorption capacity, selectivity and regeneration have all been established by MOFs in several research looking at their usage for CO_2 collection. MOFs have found usage as adsorbents in post-combustion capture techniques for CO_2. In order to efficiently separate and absorb CO_2 from exhaust gas streams at power plants and industrial sites, MOFs can absorb CO_2 selectively. MOFs like MOF-74 (CPO-27) and HKUST-1 (Cu-BTC) have been used successfully for CO_2 collection [58,59]. These MOFs have significant CO_2 adsorption capabilities and can be recycled for several uses, making them competitive candidates for industrial-scale CO_2 collection. MOFs may also be used as adsorbents, in particular for gas separation and purification, in pre- combustion capture procedures. By manipulating their structures and functions, MOFs may selectively remove CO_2 from gas mixtures, paving the way for the generation of ultra-pure hydrogen gas.

Mg-MOF-74 and UiO-66 are two Metal-Organic Frameworks (MOFs) that have shown promising CO_2 adsorption selectivity in hydrogen purification [60,61]. Because of their strong attraction to carbon dioxide, they may effectively filter out contaminants

and improve the cleanliness of the hydrogen gas that is produced. In addition, MOFs have shown promise for CO_2 collection from unexpected sources, including capture from ambient air. By selectively adsorbing CO_2 from air, MOFs open the door to atmospheric removal of this greenhouse gas. This strategy is gaining traction as a way to reduce carbon emissions and fight global warming. Amazing CO_2 collection capabilities from ambient air have been exhibited by MOFs as MOF-177 and MOF-74 [62,63]. In conclusion, MOFs have been shown to be very flexible materials for CO_2 collection. They may be optimized for post-combustion, pre-combustion, or ambient air collection because of their large surface area and selective adsorption characteristics. Researchers continue to make strides in the area of CO_2 capture and contribute to the development of sustainable solutions for mitigating climate change by creating and synthesizing MOFs with optimum structures and characteristics.

Computational Methodology

DFT calculations

Calculations based on the Density Functional Theory (DFT) are one computational approach that might be used for this purpose. In order to anticipate and understand the electronic structure and behavior of materials, DFT is a strong quantum mechanical approach [64]. Micro-porous Metal-Organic Frameworks (MOFs), Ionic Liquids (ILs), Deep Eutectic Solvents (DESs) and hazardous gases or Volatile Organic Compounds (VOCs) may all be studied by DFT simulations in this context. The adsorption processes, binding energies and stability of the captured species inside the MOFs functionalized with ILs or DESs may be better understood with the use of DFT simulations of the molecular structures and energies of these systems. The electrical characteristics and reactivity of MOFs and their composite materials may also be investigated through DFT computations. Band structure, density of states, and frontier orbital analysis may all provide insight into the charge transfer processes and catalytic activity of the materials, which are essential for the removal of hazardous gases and volatile organic compounds [64-67].

To further improve gas capture efficiency, DFT computations may be used in the rational design and optimization of novel MOFs, ILs and DESs. Computational modeling may help guide the creation of innovative materials with enhanced adsorption capabilities, selectivity, and stability by carefully examining the impacts of various functional groups, ligands and solvents. For all the structures containing ILs and DESs, the geometries of the designed structures can be optimized using Becke's three-parameter hybrid exchange functional and the Lee-Yang-Parr correlation functional (B3LYP) [65,66] with 6-311++G** basis set in Gaussian16 package [65]. CELLOPT protocol can be applied for optimizing the ILs and DESs single pairs connecting to the pore of ZIFs and to relax the composite structures. CELLOPT is used to expand the unit-cell parameters for electron diffraction data of small-molecule crystals. The CELLPOT method will be applied to assist in relaxing the structures through regulating the original lattice parameters. Dispersion correction has an incredible effect on the estimation of properties of materials [66] and consequently so effectively in finding the structure of different ZIFs [67]. As a whole, using Density Functional Theory (DFT) calculations to investigate the mechanisms behind the capture of toxic gases and volatile organic compounds in micro-porous metal-organic frameworks based on ionic liquids and deep eutectic solvents can speed up the discovery and design of efficient gas capture materials.

GCMC simulations

Grand Canonical Monte Carlo (GCMC) simulations are another computing approach that might be used for this study. When it comes to porous materials like ionic liquid and deep eutectic solvent-functionalized Metal-Organic Frameworks (MOFs), GCMC simulations are an invaluable tool for learning about adsorption and gas separation processes. It is possible to calculate adsorption isotherms, selectivity, and diffusion characteristics using GCMC simulations since they include the random insertion and removal of gas molecules into a porous medium. Adsorption behavior of hazardous gases and Volatile Organic Compounds (VOCs) within the micro-porous MOFs functionalized with ILs or DESs may be studied using GCMC simulations in the context of this issue. This helps provide light on how various solvents perform in terms of adsorption capacity, selectivity and gas capture. Adsorption enthalpy and entropy are two thermodynamic variables of the gas capture process that may be investigated using GCMC simulations. These characteristics may be used to assess the viability of the gas capture process in MOFs modified with ILs or DESs and to get insight into the driving forces involved. Gas diffusion inside porous materials is another area where GCMC simulations might provide light. The performance of MOFs in gas separation applications may be evaluated by measuring their diffusivity and permeability, both of which are derived from the motion of gas molecules. GCMC simulations may be further developed to investigate the impact of various factors on the adsorption and separation characteristics of the MOFs. These parameters include, but are not limited to, temperature, pressure, and the composition of ILs or DESs. Because of this, materials with better gas-capture capacities may be designed and optimized.

Adsorption, selectivity, diffusion and thermodynamic properties of toxic gases and volatile organic compounds within micro-porous metal-organic frameworks functionalized with ionic liquids and deep eutectic solvents can all be studied with GCMC simulations, making them a valuable computational methodology. To study single component adsorption isotherm of structures like CO_2, N_2 and CH_4 as well as binary mixtures such as CO_2/N_2 and CO_2/CH_4 mixtures and selectivity of these gases from pure ZIFs and composite materials GCMC simulations can be utilized. GCMC simulations may be used to investigate the single component adsorption isotherm of structures such as CO_2, N_2 and CH_4, as well as binary mixtures such as mixes of CO_2/N_2 and CO_2/CH_4 and the selectivity of these gases from pure ZIFs and composite materials.

Conclusion

In conclusion, this review of the literature has provided a wide-ranging survey of the cutting-edge ideas behind the capture of

Volatile Organic Compounds (VOCs) by combining micro-porous Metal-Organic Frameworks (MOFs) with Ionic Liquids (ILs) and Deep Eutectic Solvents (DESs). MOFs have come to the forefront in recent years owing to their unique ability to store and isolate gases. Synergistic effects and improved performance in VOC capture have been investigated by mixing MOFs with ILs and DESs. The study focuses on the progress made in this area, specifically how combining MOFs with ILs and DESs enhanced their adsorption capacity, selectivity and diffusion characteristics. There is hope that environmental and industrial problems may be solved by combining MOFs with ILs and DESs to better absorb volatile organic compounds. Nonetheless, we must recognize that we still face obstacles. Scalability and stability of the resultant MOF composites are discussed, along with other challenges such as adjusting the composition and characteristics of ILs and DESs. These difficulties might be seen as openings for new discoveries and innovations in the area of MOF-based VOC capture.

Future combination of MOFs with ILs and DESs shows promise for efficient collection of hazardous gases and volatile organic compounds. Exploring novel combinations of MOFs, ILs and DESs, as well as the effect of external elements like temperature, pressure, and composition, may contribute to future developments in this sector. Progressive initiatives in this area may aid in the creation of more efficient and sustainable solutions for VOC collection by tackling these problems and leveraging on the advantages of the integrated approach. This study sheds light on the current developments and potential applications of ionic liquid and deep eutectic solvent-based micro-porous metal-organic frameworks for the removal of hazardous gases and volatile organic chemicals. To propel innovation and growth in environmental and industrial applications, the combination of MOFs with ILs and DESs provides a viable road for the development of gas capture systems.

Future Prospective

There are interesting new directions that might be explored with the combination of Deep Eutectic Solvents (DESs) and Metal-Organic Frameworks (MOFs). Modern non-conventional solvents, such as DES solvents, have been shown to be useful as organic ligands in the building of Metal-Organic Frameworks (MOFs). Innovative and unique MOFs with broad applications in adsorption, catalysis, sensing, separation and gas capture/storage may be synthesized by combining the functional groups of DESs with the intrinsic sites of metal ions. More research is required to fully understand the potential of polymeric Deep Eutectic Solvents (PDESs) in the synthesis of Metal-Organic Frameworks (MOFs). Particle Depletion and Encapsulation Systems (PDESs) may be used to solve the problem of particle buildup in MOFs. They provide an opportunity to develop composite MOFs with improved stability and reactivity. Interestingly, PDESs may display diverse behaviors in bulk assemblies while having the same composition, which provides additional opportunities for modifying the characteristics of DES-based MOFs. Several more potential study avenues exist in addition to the aforementioned ones. First, MOFs built on DESs created by supramolecular self-assembly may undergo photo-degradation, opening the door to the creation of light-responsive MOFs with applications in environmental remediation.

Second, DES-based MOFs may be explored for their potential in photo-catalytic degradation of environmental toxicants, offering a potential solution to pollution problems. In order to fully comprehend and maximize MOF performance, it is necessary to look at how temperature and water affect their adsorption capabilities. Reactive hetero-atomic composites' potential to alter the conformer of DES-based MOFs is also worthy of study. Finally, molecular dynamic simulations may help us learn more about the processes involved in making MOFs and their potential uses in the future. It cannot be overstated how crucial it is to have multidisciplinary teams work together to create DES-based MOFs. To combat the negative impacts of hazardous gases and strive towards a cleaner, more sustainable earth, a multidisciplinary team comprising of chemists, chemical engineers, waste management companies and ecological specialists is required. The potential for innovative visions in the field of micro-porous metal-organic frameworks based on DESs for capturing toxic gases and volatile organic compounds, leading to advances in environmental sustainability and industrial applications, can be realized through the pursuit of these future research directions and the promotion of collaborative efforts.

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