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A Short Review of Advances in Metal-Organic Frameworks for Energy Storage

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Abstract- In the present scenario of depleting conventional energy sources, energy storage composites are of significant importance. Because of the ever-increasing demand for energy, there is a need to shift towards renewable sources of energy. Therefore, the proportion of renewable energy sources in the world must be increased. However, the electricity produced from renewable sources often faces limitations in meeting real-time demand due to the inherent intermittency, geographic dependency and variability in supply patterns. So, due to this delocalized electricity production and inconsistent sources, energy storage is the key part of renewable energy utilization. Among these energy storage devices, supercapacitors attract more attention due to their high power density, faster recharge and long cycle life. Thus, the pursuit of new storage materials with enhanced capacity, reduced mass, and superior stability remains a critical objective. Metal-organic frameworks (MOFs) are an emerging class of nanoporous materials distinguished by specific surface areas significantly greater than those of conventional adsorbents like zeolites and activated carbon. Their straightforward synthesis, high surface area, diverse functionalization options, and customizable pore structures offer distinct advantages over other porous materials. This work is focused on the electrochemical energy storage in MOFs and various methods to improve the efficiency of supercapacitor applications. This article presents engineering approaches for pristine MOFs, examines composite structures, and provides insights into the relationships between composition, structure, and activity in MOF-based materials with integrated advanced components and nanostructures.

Keywords- Metal-organic frameworks, Energy Storage, Supercapacitors, Sustainable Energy.

I. INTRODUCTION

A critical challenge confronting humanity, demanding immediate attention, is the sustainable securing of future energy resources as fossil fuel reserves diminish amid rising global energy demands [1-2]. This challenge is exacerbated by the recent growth of the worldwide market for mobile and wearable electronic technologies, as well as the environmental disturbances caused by climate change resulting from the use of non-renewable energy sources [3]. Renewable energy is regarded as a favorable strategy due to its sustainability and minimal environmental impact [4]. At present, most renewable energy sources are retained and transmitted as electrical energy [5]. Consequently, technologies that improve energy efficiency without environmental impact, such as energy storage devices, are seen as pivotal for the future. Lithium-ion batteries (LIBs) and supercapacitors are the leading contenders in this domain [6]. However, LIBs, while offering high energy density, are restricted by their low power density and poor cyclic stability [7]. Supercapacitors providing high power density and exceptional cycling stability are limited by their low energy density [8]. Consequently, neither of the two storage devices meets the criteria for high energy density, high power density, and good cycle life. Supercapacitors have garnered significant research interest and emerged as a popular topic due to their rapid charge-discharge rates, long cycle life, high power density, and environmentally friendly attributes [9-10]. Supercapacitors store energy through two distinct mechanisms. The first mechanism involves the physical

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adsorption of ions, as seen in electric double-layer capacitor (EDLC) materials, which include carbon-based substances such as graphene and carbon nanotubes (CNTs). The second mechanism relies on electron transfer reactions, typical of pseudocapacitive materials, including metal oxides, metal hydroxides, and metal sulfides. While EDLC-type materials exhibit higher stability and conductivity compared to pseudocapacitors, they are hindered by lower capacitance [11-12]. The primary components of a supercapacitor consist of the electrode, an electrolyte (which may be aqueous or organic), and a separator that inhibits short-circuiting between the two electrodes [13]. The significance of the electrodes is paramount, as effective electrode materials for supercapacitor applications should feature a high surface area, optimal pore characteristics, excellent conductivity, and affordability [14]. Thus, the strategic selection of suitable electrode materials and their rational design for supercapacitor electrodes is the most effective method for improving the storage performance of supercapacitors in comparison to LIBs. The applications of supercapacitors are continuously broadening, with recent implementations in electronic consumer devices, backup systems, and various power and energy management applications [15-16].

II. METAL ORGANIC FRAMEWORKS (MOFs)

MOFs or porous coordination polymers are a class of compounds which consist of a continuous regular array of metal ions coordinated with organic ligand molecules as shown in figure1 [17]. The organic ligands are of neutral, cationic or anionic nature and are referred to as ‘struts’ or ‘linkers’ [18]. Metal ions present in the MOFs form a node, which binds with the organic ligands together, forming a continuous, repeating well-connected cage-like structure. MOFs possess an extraordinarily vast internal surface area reaching up to 7800 m^2 g^{-1} have been to date synthesized [17]. MOFs can also be defined as a coordination network extending with organic ligands containing significant potential voids. In contrast with other porous materials, MOFs have unique structural advantages like - uniform pore structures, tunable porosity, rigid or flexible framework, uniformity in atomic-level structure, extensive varieties, chemical functionality, geometry, dimensions etc. These specialities of MOFs help researchers to control their porosity, functionality and applications.

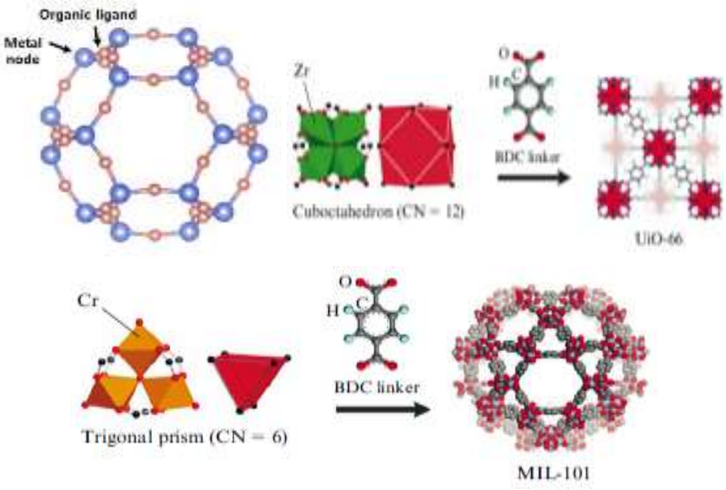

Figure 1: Schematic illustration of a MOF constructed of organic ligand and metal ion [18]

MOFs of various structures can be synthesized by choosing different metal ions and organic ligands in the structure. Researchers can develop materials which can absorb specific gases selectively into a well-organized hollow structure. Hence, MOFs open a great potential for their fruitful integration and exploration in various sensing applications in various fields. During some instances, the hollow structure (pores) remain stable even after the elimination of guest molecules (in most of the cases it will be solvent) and could be superseded by other compounds or molecules. Because of this feature, MOFs can be used to store gases like helium, hydrogen, carbon dioxide etc [10]. Also, MOFs offer applications in fields like in catalysis, gas separation, gas purification, water remediation, as superconducting materials and conducting solids [18].

A. STRUCTURE OF MOFs

Metal ions can be isolated in the MOF structure or agglomerate to form larger units, clusters referred to as secondary building units (SBUs). SBUSs and organic linkers are the two distinguishable components of MOFs [19]. Monovalent, divalent, trivalent or

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tetravalent ligands are the typical organic units. Because of this reason, MOFs are often referred to as hybrid organic-inorganic materials. The combination in which the metal ions are linked with ligands decides the structure and properties of MOF. For example, the coordination preference of metal ion decides the number of ligands to be bind and the orientation they possess, which ultimately influences the shape and size of the pores. The structural diversity can be represented in figure2, where it is evident that using different SBUs and the same linker, terephthalate (BDC), leads to different types of frameworks. For example, the combination of terephthalate with an octahedral SBU leads to the formation of MOF-5 with a cubic topology. The combination of SBU with trigonal prism shape and BDCs gives rise to MIL-101 with *mnt* topogy and cubooctahedron SBU leads to the formation of UiO-66 with *fcu* topology.

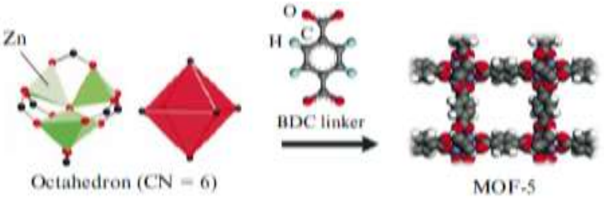

Figure 2: Structure of various MOFs with terephthalate dianion (BDC) Ligand [20].

MOF-5 is one of the initial MOFs published in 1999 by Yaghi and coworkers which consists of ZnO_4 moieties occupied at the cubic lattice sites and interconnected by the terephthalate ligands [19]. Also, materials having same structure but different pore sizes can be synthesized by increasing the carbon chain of organic ligands without changing the initial topology. The materials possessing these kinds of structural series are called "Iso Reticular Metal-Organic Frameworks" and are designated as IRMOF followed by a number. As it is already discussed, that the secondary building units can be combined through various ligands to form the framework. Choosing and substitution of ligands may lead to two situations:

- Structural symmetry is retained, and only the unit cell parameters are changed, when another ligand is used (see Fig. 3). For example, change in UiO-66 to UiO-68. In UiO-66, the ligand is a terephthalate dianion, in UiO-67, the ligand is biphenyl-4,4'-dicarboxylate dianion wherein UiO-68 terphenyl-4,4'-dicarboxylate dianion is used as the linker [21].

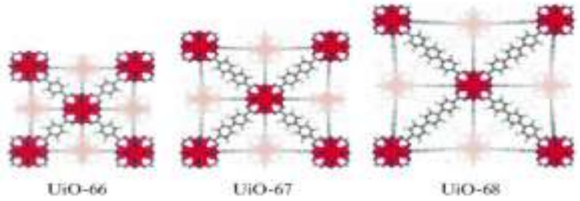

Figure 3: Framework symmetry retention by the elongation of linker chain [21]

- Change in geometry due to the change in the mutually arranged functional groups in the linker molecule [19]. Example is the following figure 4 , where yellow sphere is the pore.

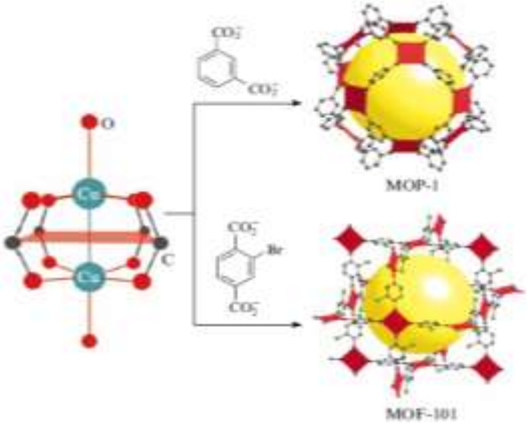

Figure 4: Example of change in geometry due to change in mutual arrangements of substituents in ligand [22]

As mentioned earlier, MOFs are usually distinguished by their pore size, difference in symmetry, structure and

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characteristics too. The size of pore is decided by the following factors:

- Length of the carbon chain of organic ligand.
- The number of benzene rings attached to the ligand [19]

Also, the addition of various functional groups and substituents to the organic ligand is the major reason for additional selectivity and the salient chemical properties of porous structure. The selection of structural components is important, and the linking nature is also very important. Studies related to the formation of frameworks from various Secondary Building Units (SBUs) have made it clear that the properties of synthesized MOFs can be predicted by some regularities found. Hence, by selecting appropriate starting components, MOFs properties can be v aried, which leads to the production of materials capable of absorbing molecules or integrating other large species selectively, e.g. fullerene (C_{60}) and myoglobin [19] .

B. How MOFs traits for electrochemical Energy Storage

This section explores synthetic methods for tailoring MOF properties to address specific needs in electrochemical applications (see Fig. 1) [23]. It emphasizes (i) structural and compositional attributes, (ii) pore structure and extensive surface area, (iii) electrical conductivity, and (iv) large-scale production and manufacturability.

The extensive specific surface area of MOFs and adjustable pore characteristics are the primary factors that enable their application as electrodes in electrochemical supercapacitors [28]. MOFs that incorporate both metals and carbon derivatives obtained through organic linkers, serve as potential candidates for the development of advanced electrode materials [29-32]. Recent research on MOFs for supercapacitor applications can be categorized into three main areas.

Manipulating the physicochemical properties of MOFs requires using them as precursors or molds in the fabrication of MOF-derived materials, including microporous carbon, inorganic metal compounds, metal hydroxides, metal sulfides, and a range of other derivatives. The synthesis method utilized for creating precursor electrodes is essential for fine-tuning the morphology, dimensions, and chemical composition of the resulting electrode materials in comparison to unmodified MOFs. Additionally, forming nanocomposites by integrating MOFs with conductive polymers (such as polyaniline (PANI) or polypyrrole (Ppy)) or carbon materials (including activated carbon (AC), graphene, and carbon nanotubes (CNTs)) is vital for enhancing the mechanical stability, electrical conductivity, and cycling performance of MOF-derived electrodes. Lastly, the fabrication of novel MOFs specifically designed for use as supercapacitor electrodes is also significant. MOFs stability and conductivity plays an important role for their uses as supercapacitor electrodes, even when newly synthesized. Its adaptable structures, large surface area and high porosity make it useful for adjustable storage of metal ions and provide noteworthy interfacial contact with electrolyte solution to enable ion transport [33-34].

FUTURE DIRECTIONS AND RESEARCH OPPORTUNITIES

This analysis aims to explore the current state of energy storage using metal-organic frameworks (MOFs). This identifies promising research directions while highlighting areas that necessitate further exploration. This approach will facilitate the addressing of existing challenges and the unlocking of novel applications within this domain.

A. Emerging research opportunities in MOFs synthesis and customization.

Research efforts should prioritize the development of scalable and cost-effective techniques for synthesizing MOFs. Innovative approaches, such as post-synthetic modification and defect engineering, hold significant potential for optimizing MOF properties for specific energy storage applications. Furthermore, rational design strategies can enable the creation of customized MOFs with enhanced performance characteristics [35,36].

B. Improvement in stability.

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One of the primary challenges in MOFs-based energy storage is ensuring stability under demanding operational conditions. Research efforts should aim to develop MOFs with enhanced stability, mainly in the presence of humidity, high temperatures, and several chemical reactions. Additionally, exploring encapsulation techniques and protective coatings may further contribute to improving the resilience of these materials [37,38].

C. Electrode material for supercapacitors.

Optimizing MOF based materials is essential for the advancement of battery and supercapacitor technology. Researchers should look on creating composite structures or integrating MOFs with other conductive materials to improve electrical conductivity and charge discharge rates. This might entail producing MOF-based composite electrodes with customized composition and porosity [39,40].

D. In situ characterization. The electrochemical characteristics of MOF-based materials for energy storage can be better understood by using sophisticated in-situ characterization techniques. Researchers can employ in situ investigations to gain a deeper understanding of the structural changes, redox reactions, and ion dynamics that occur within MOFs during energy storage processes. The development of new materials can be guided by this information[41].

IV. CHALLENGES AND RESEARCH NEEDS

A. Cost efficiency and scalability

Regardless of MOFs remarakable qualities, bulky production is quite difficult. Therefore, development of cost effective and scalable production methods are required to make MOFs popularize for energy storage applications, In this respect, collaboration between material scientists and engineers is decisive [42].

B. Safety concerns.

MOFs may pose risks in certain energy storage applications, particularly under high-temperature or chemically reactive conditions. Further research is necessary to develop safe handling and storage protocols for MOF-based energy storage materials and devices [43].

C. Long-term stability.

For practical applications such as energy storage stability of MOFs is crucial. Effect of temperature fluctuations, revelation to impurities, impacts of cycling and strength testing should be comprehensively studied for performance of MOFs [44-45].

D. Integration and system-level research. Integrating MOF-based energy storage materials into practical devices and systems presents a significant challenge. Research should focus on the development of comprehensive energy storage systems, addressing design considerations, scalability, and compatibility with existing technologies.

V. CONCLUSIONS

In the context of sustainable energy storage, this study provides a comprehensive review of the rapidly evolving field of metal-organic frameworks (MOFs). MOFs are recognized as transformative materials for advanced energy storage systems, attributed to their distinctive properties, including unique adaptability, extensive porosity, and chemical configurations. The concept of hybrid systems that integrate MOFs with other materials, such as conductive polymers or nanoparticles of carbon derivatives, is emerging. These integrations have the potential to improve the functionality of MOFs and mitigate stability issues.

Moreover, the potential development of innovative MOF structures with specialized features-such as electron-transfer sites, ion-conductive channels, or reaction active centers-aimed at improving energy storage and conversion is a significant area for future research. The implementation of MOFs not only presents scope for innovative proposal to energy storage and conversion but also hold promise for addressing pressing environmental and energy sustainability challenges. However, it is essential to acknowledge the obstacles that must be overcome to transform these promising concepts into practical solutions.

Additionally, multidisciplinary collaboration among experts in materials science, chemistry, engineering, and policy are critical for integrating MOFs into functional energy storage systems. As we move forward, the potential of MOFs in sustainable energy storage remains both exciting and substantial. To fully leverage MOF materials, ongoing investment in the exploration of new materials, synthesis techniques, and applications is essential for researchers and industry stakeholders. With sustained effort, MOFs have the capability to make a significant impact on the global transition toward sustainable, efficient, and clean energy storage technologies.

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