

METAL ORGANIC FRAMEWORK AS EMERGING MATERIALS FOR NEXT GENERATION SUPER CAPACITORS: A COMPREHENSIVE REVIEW

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Abstract

The growing global energy demand and environmental concerns have intensified the search for efficient, sustainable and high-performance energy storage devices. Super capacitors with their rapid charge-discharge rates, long cycle life and high-power density have emerged as promising candidates for next generation application and the performance of conventional electrode materials remain limited. Metal organic frameworks and emerging class of crystalline forest materials offer unit advantages including tuneable structures, high surface area and chemical versatility. These features make MOFs and their derivatives attractive for enhancing electrochemical performance, improving capacitance and enable stable cycling in supercapacitors. The recent advances highlight the use of pristine MOFs, MOF composites and MOF-derived nanostructures across different dimensionalities as electrodes, separators and catalysts, delivering improved conductivity, energy-power density and structural stability. Despite these advantages, challenges such as low intrinsic conductivity, restricted potential windows and limited long term stability are still facing in new materials. This review summarizes recent developments, synthesis strategies and structural performance relationships of MOFs for supercapacitors and outlines future research directions for scalable, conductive and robust MOF-based materials for next generation energy storage technologies.

INTRODUCTION

Energy is the fundamental driving force of human civilization, forming the backbone of industrial development, technological progress, and daily life.

Almost every aspect of modern society relies on

energy for construction, manufacturing, transportation, and operation of infrastructure [1, 2]. However, this reliance has come at a significant environmental cost. The production and

consumption of energy account for nearly two-thirds of global greenhouse gas emissions, and the predominant dependence on fossil fuels has resulted in resource depletion, severe air pollution, and climate change. The extensive use of coal, oil, and natural gas over the past century has contributed to long-lasting and potentially irreversible damage to the planet's climate systems [3, 4]. These environmental and resource challenges have prompted global efforts to transition towards sustainable and low-carbon energy solutions. In recent decades, global attention has shifted toward renewable energy sources such as solar, wind, hydropower, geothermal, and hydrogen energy [5-7]. These alternatives offer advantages including renewability, low emissions, and environmental friendliness, positioning them as crucial pillars for a sustainable future. However, while renewable energy generation has grown rapidly, it is inherently intermittent. Solar energy output depends on daylight and weather conditions, while wind energy fluctuates with atmospheric patterns. This intermittency creates challenges in maintaining a constant and reliable energy supply, as the timing of energy production rarely matches the timing of energy demand [8]. Consequently, the development of efficient and effective energy storage systems has become an essential component of the renewable energy landscape, enabling the capture of excess

energy during peak production and its release during periods of low generation [9, 10].

Energy storage technologies are vital for bridging the gap between fluctuating renewable energy production and the steady energy needs of households, industries, and infrastructure. A variety of energy storage systems are currently in use, including conventional capacitors, batteries, electrochemical capacitors (commonly known as supercapacitors), fuel cells, and hydrogen storage systems [11]. Among these, rechargeable metal-ion batteries, such as lithium-ion and lithium-air batteries, have achieved commercial success owing to their high energy densities and are widely used in portable electronics, electric vehicles, and stationary energy storage [12, 13]. Fuel cells, particularly solid oxide fuel cells, have attracted attention for their high efficiency and fuel flexibility. Nevertheless, all of these systems have limitations. Batteries, while capable of storing large amounts of energy, often suffer from slow charging and discharging rates, limited power output, and shorter cycle lives [14]. Fuel cells require complex infrastructure and specific fuel sources, making them less practical in some applications [15]. Supercapacitors have emerged as a complementary energy storage technology, offering a unique balance of high-power density, rapid charge-discharge capability, long cycle life, and environmental friendliness [16]. Batteries which

store energy through bulk electrochemical reactions, supercapacitors store energy either via electrostatic charge accumulation at the electrode–electrolyte interface or through fast surface redox reactions. This enables them to be charged and discharged within seconds, with minimal degradation over hundreds of thousands of cycles. Their design typically consists of two electrodes separated by an ion-permeable membrane and immersed in an electrolyte [17-21]. Hybrid supercapacitors combine these two mechanisms to optimize performance. Supercapacitors are increasingly recognized for their role in applications where rapid energy delivery or uptake is essential, such as regenerative braking in electric vehicles, emergency power supplies, grid frequency regulation, and portable consumer electronics. They offer advantages over batteries in terms of power density, with typical values ranging from 500 to 10,000 W/kg, and they can operate across a wide temperature range with minimal maintenance. However, their energy densities, typically between 1 and 10 Wh/kg, are considerably lower than those of batteries, which limits their ability to serve as the sole energy storage device in

applications requiring long-duration power delivery [22, 23]. Bridging this performance gap has become a major research focus, with strategies centered on increasing the specific capacitance and expanding the operating voltage window of the device. The performance of a supercapacitor is largely determined by the properties of its electrode material. High-performance electrodes should possess a large accessible surface area, excellent electrical conductivity, appropriate pore size distribution for ion transport, and good mechanical and electrochemical stability [24]. Conventional EDLC electrodes are typically made from activated carbon due to its high surface area and chemical stability, while pseudocapacitor electrodes often use transition metal oxides or conducting polymers to enhance capacitance through redox activity. In recent years, the development of advanced nanostructured materials has opened new pathways for improving supercapacitor performance. Among these, metal–organic frameworks (MOFs) have attracted growing interest due to their tuneable structure, large surface area, and chemical versatility [25].

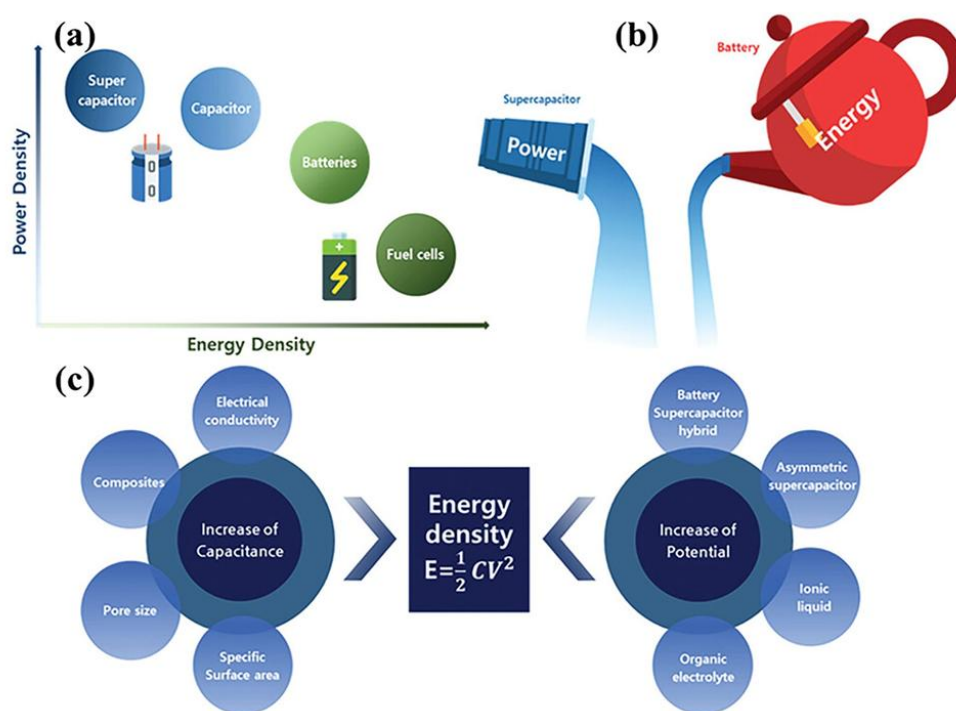


Fig 1: **a** Ragone plot depicting the range of power and energy densities for various traditional energy storage and conversion systems, **b** Schematic illustration showing the correlation between power density and energy density; **c** Diagram presenting various approaches to improve the energy density of supercapacitors. Reproduced from Ref. [2].

Metal Organic Frameworks (MOFs) are crystalline materials composed of metal ions or clusters coordinated with organic ligands to form one, two, or three-dimensional frameworks with well-defined porosity [26-28] as shown in Figure 2. First reported by Yaghi and co-workers in 1995, MOFs offer exceptionally high surface areas, adjustable pore sizes, and the ability to tailor their structure and functionality by selecting appropriate metal

nodes and organic linkers. This modularity makes MOFs attractive for a range of applications including gas storage, catalysis, drug delivery, and increasingly, energy storage [29, 30]. The application of supercapacitors, MOFs provide abundant active sites for ion adsorption and redox reactions, tuneable pore structures for efficient ion transport, and the possibility of incorporating electrochemically active metal centres. Despite these advantages, pristine MOFs face significant challenges when applied directly as supercapacitor electrodes. Their poor intrinsic electrical conductivity limits charge transport, while structural instability under repeated electrochemical cycling can lead to performance degradation [31]. Additionally, in some cases, only a fraction of the active sites is accessible

to the electrolyte due to restricted ion diffusion into the framework. One approach is to create MOF composites by integrating them with conductive materials such as graphene, carbon nanotubes, MXenes, or conductive polymers. This enhances the electrical pathways while preserving the high surface area of the MOF. Another strategy is to derive functional materials from MOFs through thermal or chemical conversion, producing porous carbons,

metal oxides, metal sulfides, or hybrid nanostructures that inherit the parent MOF's porosity but possess higher conductivity and stability [32, 33]. Morphology control and nanostructures such as forming nanosheets, nanorods, hollow spheres, or aerogels can further improve ion accessibility and shorten diffusion paths, enhancing electrochemical performance [34].

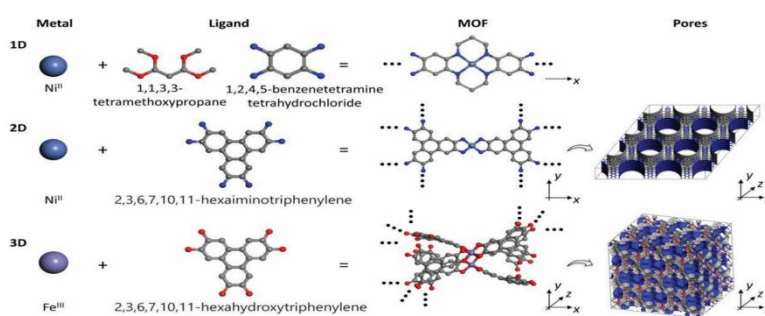


Fig 2: Design strategies for metal–organic frameworks (MOFs) are determined by the spatial arrangement of their inorganic and organic components. MOF has been successfully synthesized [26, 27, 35], with the pore structure highlighted in blue.

The rapid pace of research in MOF-based supercapacitors reflects the growing recognition of their potential to address the limitations of conventional materials. However, several challenges remain before these materials can be deployed in large-scale, practical applications. Issues such as cost-effective synthesis, scalability, long-term stability, and performance in real-world conditions must be addressed. Understanding the relationships between structure, composition, and electrochemical behaviour will be key to overcoming these obstacles

[36-38]. This review aims to provide a comprehensive overview of the recent progress in the development and application of MOFs and their derivatives for next-generation supercapacitors. It will discuss the synthesis strategies used to tailor MOF properties, the influence of structural features and dimensionality on electrochemical performance, and the role of composite formation and derivation in enhancing conductivity and stability.

Advanced Techniques for MOF Nanostructure Synthesis

Metal Organic Frameworks (MOFs) are a class of porous crystalline materials that were first introduced in 1995 by Omar Yaghi. Their innovation marked a significant advancement in materials science due to the strong coordination between metal ions and organic ligands. These materials quickly gained attention for their large surface areas, high porosity, and structural versatility [39]. Initially referred to as coordination polymers, early MOFs included isolated polyhedral or small clusters similar to traditional coordination chemistry. Over time, it became evident that more complex, extended inorganic structures could be incorporated, giving rise to MOFs multi-dimensional frameworks with ordered and tuneable properties. The development of MOFs can be traced back to 1989 when Hoskins and Robson created three-dimensional frameworks using metal clusters arranged in octahedral or tetrahedral geometries linked with organic components. A notable early example is the diamond-like structure $[N(CH_3)_4][CuZn(CN)_4]$, analyzed using single-crystal X-ray diffraction [40]. In 1997, Kitagawa and colleagues synthesized a 3D MOF capable of gas adsorption at room temperature. Later, in 1999, the publication of HKUST-1 and MOF-5 significantly advanced the field, with both frameworks becoming widely studied for their outstanding structural and functional properties. From 2002 onward, researchers such as

Férey expanded the MOF family by introducing new variants like the flexible MIL-47 and the more rigid MIL-88 and MIL-53 [41]. Among the most well-known MOFs is Cu-BTC, also known as HKUST-1, which has been commercialized by BASF under the name BASOLITE® C300. The growing interest in MOFs is reflected in the increasing number of patents filed by major companies such as Toyota, Ford, Strem Chemicals, and MOF Technologies. According to the World Intellectual Property Organization (WIPO), there has been a steady rise in MOF-related patents, demonstrating their commercial potential [42].

MOFs possess several key features that make them attractive for a wide range of applications. Their exceptionally high surface area, tuneable pore sizes, and chemical adaptability make them suitable for use in gas storage, separation, catalysis, drug delivery, and environmental cleanup. MOFs have shown particular promise in adsorbing gases such as hydrogen, methane, and carbon dioxide, making them valuable for energy storage and carbon capture technologies. In catalysis, their diverse functional groups enable high activity and selectivity, while their biocompatibility and structural tunability support their use in controlled drug delivery. MOFs are also being explored for environmental applications, such as the removal of pollutants from air and water. MOFs are composed of metal ions

coordinated to organic linkers, forming one-, two-, or three-dimensional frameworks [43]. These structures feature voids and channels that resemble a sponge, and their porosity remains intact even after guest molecules are removed this property is referred to as permanent porosity. The ability to tailor these frameworks through careful selection of metal clusters and ligands has enabled researchers to design MOFs with specific functionalities. The synthesis of MOFs can be achieved through various methods, including co-precipitation, solvothermal, mechanochemical, microwave-assisted, electrochemical, and sonochemical processes. Each method influences the material's crystallization rate, particle size, surface area, and morphology. Recent research has focused on using MOFs in

supercapacitors (SCs) through two main approaches: first, using MOFs as templates to produce porous metal oxides or carbon materials with large surface areas (typically 0.6–2 nm in size); second, employing MOFs directly as electrode materials [44]. Although the latter approach is more complex due to structural flexibility, this flexibility can be stabilized to create functional electrode materials. The electrochemical performance of MOFs can be fine-tuned to support various reactions, such as the oxygen reduction reaction (ORR), oxygen evolution reaction (OER), hydrogen evolution reaction (HER), and carbon dioxide reduction reaction (CO₂RR). These properties open possibilities for MOFs to serve in supercapacitors, batteries, and fuel cells [45].

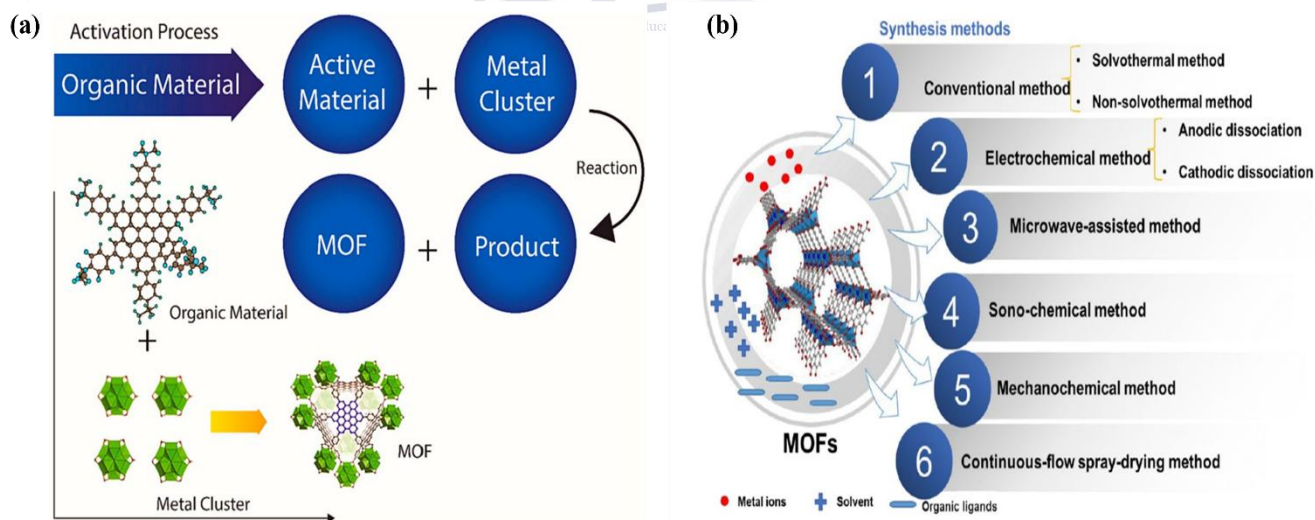


Fig 3: *a.* An overview of a typical MOF synthesis method [43]. *b.* Schematic representation of the MOF synthesis route [45].

Despite their potential, MOFs face limitations such as low electrical conductivity and structural

instability. These issues can hinder their performance in electronic applications. To address

this, researchers have developed MOF-derived materials by modifying their structure to enhance electrochemical behaviour and stability. The intrinsic framework of MOFs consisting of metal nodes and organic linkers allows for extensive customization. Producing crystalline and porous MOF materials typically requires mild conditions, and a variety of synthetic routes have emerged over the last two decades to achieve optimal architectures and performance characteristics. These efforts continue to drive innovation in energy storage, environmental remediation, and advanced material design, confirming MOFs as a vital class of materials in modern science and technology [46, 47].

Conventional Route for MOF Synthesis: The most widely adopted method for synthesizing metal-organic frameworks (MOFs) is the solvothermal or hydrothermal approach, where a metal salt and an organic linker are dissolved in a solvent and heated under pressure in a sealed vessel such as an autoclave. This technique promotes the formation of crystalline MOFs by operating at temperatures above the boiling point of the solvent under autogenous pressure. The type of setup (solvothermal vs. non-solvothermal) depends largely on the temperature and pressure conditions used during synthesis. In solvothermal synthesis (SVT), reactions typically occur in closed vessels at elevated temperatures, whereas non-SVT reactions proceed at

or below the solvent's boiling point, often simplifying reaction conditions. Critical parameters in MOF synthesis include reaction temperature, solvent choice, pressure, concentration, and reaction time. Temperature notably affects crystallinity and morphology [48]. For instance, higher temperatures can promote more compact structures but may risk framework collapse if extended for too long. Water as a solvent denotes the hydrothermal variant, while common aprotic solvents such as DMF, DEF, and DMSO are used in other solvothermal setups. Protic solvents like methanol and ethanol are also employed. Solvent selection influences the solubility of reactants, deprotonation rates, and coordination interactions, thereby impacting nucleation and crystal growth.

The use of modulators (chemical additives) in solvothermal synthesis plays a vital role in controlling crystallite size and reducing structural defects. Modulators compete with ligands for metal binding sites, altering the nucleation rate and sometimes even substituting ligands to affect crystal structure. Adjusting modulator concentration can tailor MOF morphology, especially when combined with solvent polarity and acidity variations. This strategy has enabled successful formation of well-known MOFs like ZIF-8 and MOF-5 at relatively low temperatures. To address the low capacitive performance of traditional MOFs in supercapacitors,

researchers have introduced new ligands like 3,5-diaminobenzoic acid (DABA) to create electroactive bimetallic MOFs (DABA-MOFs). These MOFs, synthesized using a one-step solvothermal method with Ni and Co ions, demonstrated improved morphology and performance when solvents of varying deprotonation abilities (methanol, ethanol,

and DMF) were used as shown in Figure 4. Solvent polarity affected the transformation from smooth rod-like to rough hydrangea-like structures, significantly enhancing electrochemical performance [49-51].

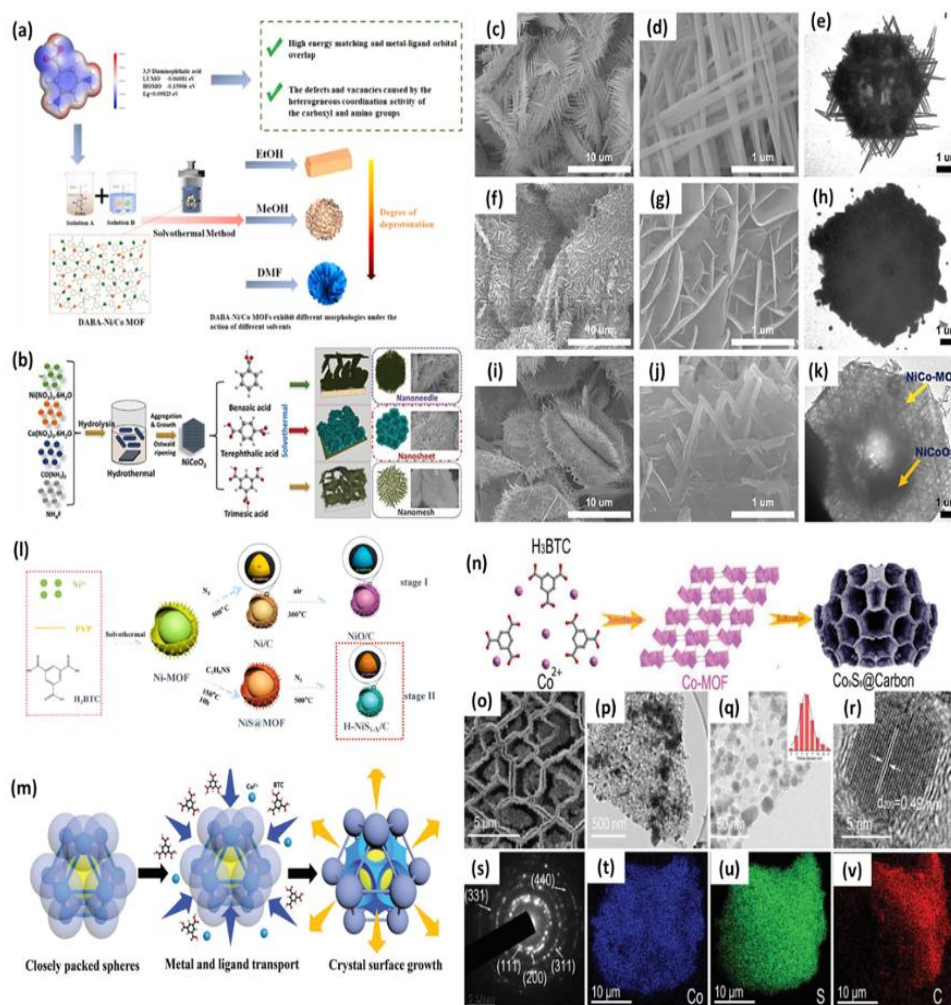


Fig 4: *a* A schematic diagram illustrating the preparation process of E/M/D DABANi/Co MOF. *b* Illustration of the synthesis pathway for NiCo-MOF@NiCoO₂@Ni.FESEM images of: (*c*,

d) benzoic acid-60 nanoneedles, (*f*, *g*) H2BDC-80 nanosheets, and (*i*, *j*) H3BTC-100 nanomesh electrodes. TEM images of: (*e*) benzoic acid-80, (*h*) H2BDC-80, and (*k*) H3BTC-100 electrodes.

Schematic depicting the synthesis of Ni-based hollow nanocomposites, starting from a Ni-MOF precursor, followed by NiO/C formation in stage I and H-NiS₁-X/C formation in stage II. **m** Illustration showing how the template influences crystal morphology: a single yellow sphere surrounded by 12 purple spheres in a close-packed face-centered cubic (fcc) arrangement, highlighting crystal growth restriction zones (blue), along with directional transport of metal and ligand and oriented crystal surface development. **n** Schematic of the synthesis approach for Co₉S₈@C-500. **o** FESEM, (**p**, **q**) TEM (with inset showing particle size distribution), (**r**) HRTEM, (**s**) SAED patterns, and (**t-vi**) elemental mapping images of Co₉S₈@C-500.

The choice of organic linker is a crucial factor in determining the structural and functional properties of MOFs. For example, using benzoic acid, terephthalic acid, and trimesic acid as ligands led to the formation of distinct morphologies nanoneedles, nanosheets, and nanomeshes, respectively. Among these, the H₂BDC-based nanosheet structure showed superior electrochemical performance,

indicating that morphology directly influences capacitor behavior. Ni-based MOFs were synthesized using trimesic acid and PVP via solvothermal methods to form hollow MOF precursors. These were further thermally treated to form Ni/C and NiO/C composites while retaining their hollow spherical shapes. An additional step involving sulfurization yielded NiS-based carbon-coated structures with sulfur vacancies, ideal for energy storage due to their high surface area and electrochemical stability. Advanced strategies have been developed to create ordered macro-microporous single-crystalline MOFs using hard templates and monodentate ligands. For example, HKUST-1 was synthesized using polystyrene (PS) spheres arranged in an fcc structure to direct crystal growth along specific paths. Sodium formate was found to facilitate nucleation and single crystallinity, while its excess led to polycrystalline structures [52, 53]. This method enables precise control over morphology and crystal quality, crucial for energy applications.

Table 1: Overview of MOF-Based Material Syntheses Using Different Methods

Synthesis Route	MOF Material	Metal Precursors	Ligands	Solvent(s)	Conditions
Solvothermal (SVT)	HKUST-1	Cu(NO ₃) ₂ ·3H ₂ O	H ₃ BTC	EtOH, H ₂ O	50 °C, 3 h
	ZIF-8	ZnCl ₂	Hmim	NaHCO ₂	130 °C, 4 h
	MOF-5	Zn(NO ₃) ₂ ·6H ₂ O	H ₂ BDC	DMF	120 °C, 16 h
	Cu-FMOF-4B	Cu(NO ₃) ₂ ·3H ₂ O	3-Methylpyridine	DMF, DEF	85 °C, 96 h

	UiO-66	ZrCl ₄	H ₂ BDC	DMF	120 °C, 24 h
	MIL-101	Cr(NO ₃) ₃ ·9H ₂ O	H ₂ BDC	HF, H ₂ O	210 °C, 60 min
	Mg-MOF-74	Mg(NO ₃) ₂ ·6H ₂ O	H ₄ DOBDC	DMF, EtOH, H ₂ O	125 °C, 24 h
	FeMIL-101	FeCl ₃	BDC	DMF	150 °C, 15 min
Microwave-Assisted (MWA)	IRMOF-1	Zn(NO ₃) ₂ ·6H ₂ O	BDC	DMF	150 °C, 24 h
	HKUST-1	Cu(NO ₃) ₂ ·6H ₂ O	H ₃ BTC	Ethanol	120 °C, 10 min
	MOF-5	Zn(NO ₃) ₂ ·4H ₂ O	H ₂ BDC	NMP	100 °C, 15 min
	ZIF-8	Zn(NO ₃) ₂ ·6H ₂ O	MIIm	DMF	120 °C, 1 min
Electrochemical (ELC)	Cu-MOF	Cu plate	BTC	MeOH	1.3 A, 12 V, 150 min
	HKUST-1	Cu rod	BTC	TBAH, H ₂ O	1.3 A, 19 V, 5 min
	Zn-MOF	Zinc plate	1,3-H ₂ BDC, Hbzim	DMF, EtOH	60 mA, 60 V, 2 h
	UiO-66-NH ₂	Zirconium (metal)	TABA	DMF, Acetic acid	2 A, 4 V, 1 h
Sonochemical (SCL)	HKUST-1	Cu ₂ (OAc) ₆ ·4(H ₂ O) ₂	H ₃ BTC	DMF, EtOH	40 kHz, 30 min
	MIL-88A	FeCl ₃ ·6H ₂ O	Fumaric acid	MeOH, EtOH, DMF	20 kHz, 2 h
	Mg-MOF-74	Mg(NO ₃) ₂ ·6H ₂ O	2,5-H ₄ DHTP	DMF, H ₂ O, EtOH	500 W, 20 kHz, 1 h

Microwave assisted Route for MOF Synthesis:

Microwave-assisted (MWA) synthesis has emerged as an effective technique for the rapid crystallization of metal-organic frameworks (MOFs), providing a faster and more uniform alternative to traditional heating methods. This process benefits from the formation of localized superheating zones and rapid

heat transfer, which promote consistent nucleation and result in uniform particle sizes. MWA relies on electromagnetic radiation interacting with mobile charge carriers either ions or polar molecules in liquids or solids. Typically conducted at temperatures above 100 °C and within an hour, this method allows for efficient MOF production.

Adjusting parameters such as temperature, solvent, reactant quantities, irradiation time, and intensity can further optimize the synthesis conditions. Compared to conventional electric heating, MWA achieves faster crystallization, smaller and more uniform crystal sizes, and higher product purity. Numerous MOFs based on metals like iron, chromium, titanium, cerium, and zinc have been synthesized successfully through MWA [54, 55]. For instance, CrMIL-100 was synthesized with a 44% yield at 220 °C within 4 hours. CrMIL-101 nanocrystals, around 50 nm in size, were produced by increasing the pH of the reaction mixture, avoiding the use of hazardous hydrofluoric acid. Using DMF as a solvent, FeMIL-101 nanoparticles with diameters of 200 nm were synthesized. In another case, the size of highly crystalline IRMOF-1 crystals was reduced to 1 μm by lowering the concentration of H_2BDC in the starting solution. HKUST-1/MW@ $\text{H}_3\text{PW}_{12}\text{O}_{40}$ was produced via an enhanced microwave approach, resulting in materials with excellent surface area, pore volume, and water stability due to polyoxometalate incorporation. Similarly, Ni-CPO-27 nanocrystals (22–28 nm) were created, with their pore structure tuneable by adjusting irradiation time and temperature [56].

Transition metal selenides (TMSs) are gaining attention for applications in sodium-ion

batteries (SIBs) and supercapacitors (SCs) due to their high redox activity and theoretical capacity enabled by multielectron reactions. Selenium's semi-metallic properties, superior conductivity compared to sulfur, and lower electronegativity make it particularly favorable [36]. Its large ionic radius leads to weaker metal–selenium bonds, which lowers the energy barrier for sodium-ion movement and enhances reaction kinetics. Furthermore, TMS-based electrodes avoid the formation of polyselenide intermediates, improving cycle stability. However, their practical use is challenged by volume expansion and particle aggregation, which degrade performance over time. To address these issues, this study proposes a microwave-assisted method to synthesize core–shell $\text{NiSe}_2@\text{NC}$ (nitrogen-doped carbon) nanocomposites using Ni-MOFs as precursors as shown in Figure 5. The MWA process offers several advantages, including speed, environmental friendliness, and scalability. The resulting core–shell structure not only limits ion diffusion distances but also enhances charge transport, making these materials highly promising for next-generation energy storage systems [57].

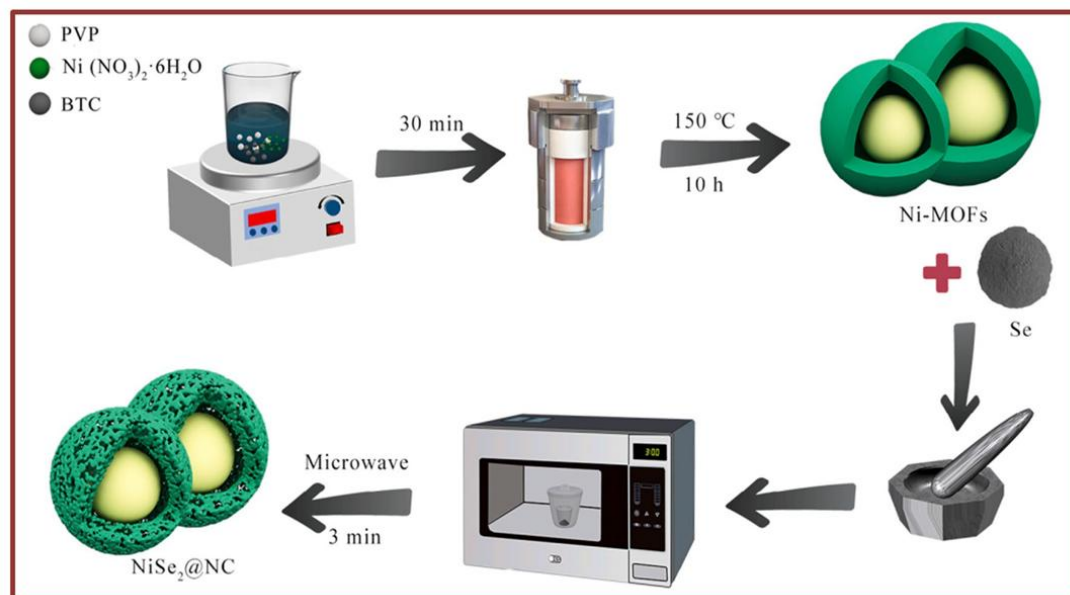


Fig 5: Microwave-assisted synthesis process for NiSe₂@NC materials.

Electrochemical Route for MOF Synthesis:

Electrochemical synthesis presents a cleaner and more controlled alternative to traditional metal salt-based methods for the fabrication of metal-organic frameworks (MOFs). Instead of relying on metal salts, which often introduce unwanted anions and impurities, this method sources metal ions directly from anodization. This in situ generation of ions near the substrate surface reduces contamination and promotes the growth of purer MOF crystals. As a result, electrochemical synthesis is gaining prominence in materials research due to its mild reaction conditions, rapid synthesis times, and minimal need for post-processing. In MOF synthesis, electrochemical methods enable direct and tuneable deposition of MOF thin films onto a variety of substrates without requiring any surface pre-treatment. This not only simplifies the synthesis process but also allows real-

time adjustment of the resulting structure and properties. Moreover, since no metal salts are used, there is no need for counter-anion removal, which streamlines the overall procedure [28, 58]. Well-known MOFs such as MIL-100(Al), HKUST-1, and NH₂MIL-53(Al) have been successfully synthesized using electrochemical cells via anodic dissolution.

Several studies highlight the versatility and scalability of this approach. For instance, Cao et al. developed a cost-effective method to synthesize Ni-MOFs directly on nickel foam, resulting in rod-like structures that function efficiently as electrodes without further processing [59]. Mueller et al. extended this technique to various Zn- and Cu-based MOFs using multiple combinations of metals and linkers, enabling the large-scale production of high-surface-area, porous materials. In comparison studies, materials synthesized through electrochemical methods

demonstrated better performance than those produced by conventional solvothermal techniques, particularly in terms of stability and sensing capability, as seen in the case of UiO-66-NH_2 for Fe^{3+} ion detection. Electrochemical synthesis also plays a crucial role in enhancing the performance of MOF-derived materials in energy applications [60]. For example, Co–Mn layered double hydroxide (LDH) structures have been electrodeposited onto nickel foam using controlled potentials, resulting in uniform, conductive layers suitable for energy storage. Reaction conditions such as voltage, time, and ion concentration directly influence the growth and thickness of these layers, optimizing their electrochemical properties [61].

Researchers have successfully integrated conductive polymers, like polyaniline (PANI), into MOFs via electrochemical polymerization to further boost conductivity. Hybrid materials like ZnCe-MOF and CuCe-MOF, coated with PANI, have demonstrated significant improvements in electrical performance, offering a promising approach for advanced electrode design. Despite the broad potential of MOFs, their application is often limited by inherently low electronic conductivity. To overcome this, bimetallic MOFs such as Ni-Co MOFs have been developed, offering enhanced charge storage and transfer capabilities due to synergistic interactions between metal centres. Electrochemical deposition onto conductive substrates like nickel foam ensures strong physical contact and facilitates self-assembly, leading to improved performance in supercapacitor

applications. Electrochemical synthesis provides a green, efficient, and highly controllable route for MOF fabrication. It enables the production of high-purity MOFs with tailored structures, improved electrical properties, and compatibility with scalable manufacturing [34, 62]. These advantages position electrochemical methods as a key strategy for advancing MOF-based technologies in sensing, catalysis, and energy storage.

Sonochemical Route for MOF Synthesis:

Sonochemical synthesis (SCL) uses high-energy ultrasound to enable rapid and uniform nucleation, producing smaller particles and faster crystallization compared to traditional solvothermal methods. Ultrasound frequencies range from low (20–100 kHz) to high (over 1 MHz), and the process relies on acoustic cavitation formation and collapse of bubbles creating extreme local temperatures and pressures that promote crystal formation. For example, using SCL, MOF-5 nanocrystals (5–25 μm) were made in 30 minutes with physical properties comparable to conventional samples [63]. HKUST-1 nanocrystals formed quickly (5 min) at 10–40 nm sizes, growing larger with longer reactions. Fe-fumarate (MIL-88A) nanoparticles (100 nm) were synthesized via ultrasound at 50 °C, with reaction conditions influencing shape and yield [57]. Similarly, $\text{Zn}_3(\text{BTC})_2$ nanorods and lanthanide-based MOFs ($\text{Ln}(\text{BTC})(\text{H}_2\text{O})$) were prepared faster and at lower temperatures using ultrasonication, yielding highly crystalline materials. ZIF-8, a widely studied MOF

known for its thermomechanical stability and porosity, was sonochemically synthesized and incorporated as a nanofiller in waterborne polyurethane (WPU) to create ZIF-8/WPU nanocomposites [64]. These composites exhibited enhanced thermal, mechanical, and barrier properties, demonstrating the effectiveness of sonochemical methods in producing advanced nanocomposites for industrial uses like coatings and packaging.

Conclusion and Future Perspectives

In conclusion, developing advanced materials for electrochemical energy conversion and storage, along with eco-friendly nanotechnologies, is critical. Metal-organic frameworks (MOFs), known for their high surface area, tuneable chemistry, and porous structures, have emerged as promising candidates for next-generation electrode materials. Their derivatives are also being widely explored for supercapacitor applications due to these distinctive properties. Numerous reviews discuss MOFs in supercapacitor technologies, few focus on their varying dimensional forms. This review highlights recent progress in designing MOF-based composites across different dimensions—ranging from 1D nano powders and 2D nanofilms to 3D aerogels and even 4D self-supporting electrodes.

Despite advancements, several key challenges hinder the commercial use of MOFs in energy storage. Firstly, most MOFs suffer from low electrical conductivity, limiting their direct use as electrodes. This impacts the electric double-layer capacitance and

overall performance. Hence, synthesizing MOFs with enhanced conductivity is vital. Secondly, many MOF-based electrodes display poor electrochemical capacitance and low energy density, making them unsuitable for high-energy applications. Designing nanocomposites with tailored surface functionalities could improve their performance significantly. Thirdly, rational structural design is essential to improve ion and electron transport. Creating 3D hierarchical self-supporting structures can enhance performance while eliminating the need for binders or conductive additives. Another area of focus is the development of flexible, transparent supercapacitors for smart wearable electronics. These require MOF-based electrodes that are lightweight, foldable, and durable. To support commercialization, standardized tests for bending, stretching, and long-term stability must be established. Additionally, the mechanisms behind energy storage and conversion in MOFs remain poorly understood due to complex reaction pathways. Theoretical modeling and simulation must play a greater role in guiding the design and synthesis of efficient electrode materials with predictive insight into their behaviour. Most MOF-based supercapacitor technologies remain at the laboratory stage, their potential for breakthroughs in energy storage is significant. Realizing this promise will require optimizing synthesis strategies, ensuring scalability, and addressing production costs. With continued research and innovation, MOFs and their derivatives

are poised to play a transformative role in next-generation energy storage systems.

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