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Review

MOFs and Three-Dimensional Device Architectures for Sustainable Hybrid Supercapacitors

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Abstract

The demand for sustainable energy storage technologies has generated enormous research interest in hybrid supercapacitors, which combine the high power density of supercapacitors with the favorable energy density of battery-type systems. Metal–organic frameworks (MOFs) are promising electrode candidates in energy storage due to their tunable compositions, high degree of porosity, various active sites, and their versatile chemistry. However, these fascinating properties of MOFs do not necessarily translate into high performance in hybrid supercapacitor devices, as their performance largely depends on the configuration of the electrodes, the interfacial contact, and the mass-transport pathways. One appealing approach to address these challenges is the development of 3D electrode architectures, in which MOFs and derivatives are embedded in interconnected conductive frameworks and hierarchical pore networks with favorable electron transport, electrolyte infiltration, and ion diffusion. This review critically assesses the progress on MOF-based 3D architectures for sustainable hybrid supercapacitors, focusing on MOF composition, defect engineering, MOF-derived materials, conductive scaffolds, hierarchical porosity, and device architecture. We focused on green synthesis, bio-sourced ligands, bio-based carbon, low-energy processes, and scaling up fabrication. The sustainability–performance relationships and the difficulties associated with the implementation of the laboratory-scale architectures into actual devices are critically analyzed.

Keywords: sustainable MOFs; 3D architecture; supercapacitors; sustainable energy

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1. Introduction

The rapid advancements in portable electronics and distributed power technologies have driven the need for electrochemical energy storage devices that are capable of simultaneously delivering high energy density, high power density, fast charge and discharge rates, and extended operational life cycle [1]. Supercapacitors, which have a high-power capability with excellent cycling durability, are important as they bridge the performance gap between conventional dielectric capacitors and rechargeable batteries. However, their relatively low energy density is still a substantial drawback for applications that demand long-term energy output. Developing hybrid supercapacitors (HSCs) where capacitive and Faradaic charge-storage processes occur in a single device has thus become a promising approach to achieve high performance in this regard [2]. HSCs can incorporate complementary charge-storage mechanisms with a capacitor-type electrode, using a battery-type

or pseudocapacitive electrode to realize higher energy density with desirable power and cycling properties [3].

The rational design of electrode materials and their structural organization is closely related to the development of high-performance HSC [4]. The carbonaceous materials, transition-metal oxides, hydroxides, sulfides, phosphides, and conducting polymers, as well as their composites, have been widely studied due to their electrical conductivity, redox activity, or accessible surface area [5,6]. However, the performance of a device may not always be guaranteed when the optimization of a single material property is performed. The electrochemical response of an electrode can be significantly affected by the thickness of the electrode, the connectivity of the pores, the accessibility of the electrolyte to the pores, the electronic pathways, the interfacial resistance, the mechanical integrity of the electrode, and the utilization of the active materials. This has led to a greater focus on simultaneous engineering of the chemical compositions and the architecture of the electrodes [7].

Among this diverse range of materials, metal–organic frameworks (MOFs) have been the subject of intense research. MOFs are crystalline porous materials that are composed of metal ions or clusters that coordinate with an organic linker, offering an exceptionally adjustable composition, topology, pore size, surface chemistry, and functionality [8]. The highly porous nature and internal surfaces of these materials can host many sites for electrolyte interactions, and the addition of redox-active metal centers and/or organic linkers can enable sites for Faradaic charge storage. Notably, the use of MOFs as simple porous powders is being replaced by new emerging applications in energy storage. Their incorporation with materials of other dimensions such as carbon nanotubes, graphene, conductive polymers, metal oxides and three-dimensional conductive frameworks can lead to hierarchical architectures that combine and enhance conductivity, structural stability, and electrochemical accessibility. In recent years, the importance of combining MOFs with functional 0-, 1-, 2-, and 3-dimensional materials has been emphasized to overcome the limitations of conventional MOF electrodes [9].

In this regard, three-dimensional (3D) electrode architectures are highly attractive, as they add structural control at the device level to MOF energy storage [10]. In contrast with planar electrodes, 3D architectures can offer interconnected electronic networks, continuous electrolyte pathways, high accessible surface areas, and hierarchical pore structure. Currently, conductive scaffolds, such as nickel foam, carbon cloth, carbon felt, graphene aerogels, porous carbon frameworks, and the newly developed additively manufactured structures, can be used as both current collectors and structural supports. The active phase can be directly grown or deposited on these scaffolds, which can decrease inactive interfaces and enhance contact between the active phase and the current collector. However, the spatial arrangement of the active material can affect ion diffusion length, electrolyte penetration, electron transport, and utilization of electrochemically active sites [9].

The importance of 3D architecture is more pronounced when comparing material-level performance to device-level performance. Specific capacitance values that happen to be extremely high under three-electrode conditions are not guaranteed to translate to high-performance practical devices [11]. Adding mass loading to the electrodes can increase electrode separation, internal resistance, inaccessible active sites, and non-uniform electrolyte distribution. Likewise, it may be possible to enhance the apparent electrochemical performance of the materials by adding conductive additives, polymer binders, metallic substrates, or more complex nanostructuring processes but at the expense of increasing the inactive mass, processing complexity, cost, or environmental impact. Thus, all the intrinsic material properties, the architecture-dependent transport phenomena, realistic mass loading, and the full-cell characteristics must be considered together for meaningful progress in MOF-based HSCs [12].

Sustainability is another important consideration when it comes to 3D architecture involving MOF-based hybrid supercapacitors [13]. Advanced research in energy storage technologies has led to the need to assess both the high electrochemical performance of a material and its synthesis, processing, energy storage integration, application, and disposal or recycling. The availability and selection of the metal centers, organic linkers, solvents, modulators, reaction conditions, activation methods, energy usage, yield, and post-synthetic processing are factors that affect MOF sustainability. This means that approaches involving earth-abundant metals, aqueous or low-toxicity solvents, bio-derived ligands, biomass-derived carbon, low-energy synthesis, and scalable processing of MOF-based electrodes are important avenues to pursue for more sustainable approaches. In recent years, green synthesis, flexible substrates, and 3D printing have been identified as pertinent fields for the development of the functional and sustainable properties of MOF-based supercapacitor systems [14].

In this regard, it is important to know what can be done to engineer MOF chemistry and three-dimensional electrode architecture simultaneously to gain high electrochemical performance and retain sustainability. To address this, the conventional comparison based on specific capacitance (or energy density) is not enough, and the relationships between MOF composition, structure, 3D architecture, charge-storage mechanism, ion/electron transport, device configuration, and environment should be established.

Several reviews have examined the development of MOF-based materials for supercapacitor applications, covering pristine MOFs, MOF-derived carbons and metal compounds, composite electrodes, synthesis strategies, charge-storage mechanisms, and approaches for improving electrochemical performance [15–19]. For instance, Wei et al. [20] reviewed the progress and future directions of MOF-based and MOF-derived electrode materials, while other studies have focused on MOF/carbon composites and the dimensionality–functionality relationships of MOF-based composite electrodes [21–23]. These contributions have substantially advanced understanding of the relationship between MOF composition, porous structure, and electrochemical performance. Nevertheless, an important complementary perspective is to examine how these material-level advances translate into the design, integration, and sustainable operation of three-dimensional hybrid-supercapacitor devices. Material properties cannot be considered independently of electrode architecture, electrolyte accessibility, interfacial contact, electron and ion transport pathways, and fabrication requirements. Furthermore, high electrochemical performance alone does not establish the environmental sustainability or practical viability of a device.

Therefore, this review critically evaluates MOFs and 3D device architectures for sustainable hybrid supercapacitors. We do not simply catalogue the MOF compositions but attempt to highlight how the design at the molecular level can be translated into hierarchical structures of the electrodes and finally into a working device. Publications between 2015 and 2026 were identified through keyword searches in scientific databases, focusing on MOF characteristics, electrode structure, electrochemical performance, and sustainable fabrication. Special focus is placed on pristine and multimetallic MOFs, defect engineering, MOF-derived materials, MOF/conductive-material composites, 3D conductive scaffolds, hierarchical porosity, freestanding electrodes, and new 3D-printing approaches. Finally, the sustainable–performance trade-offs, realistic electrode loading of electrodes, evaluation of the full cell, scalability and new possibilities for transport-informed and life-cycle-oriented electrode design are discussed. With this comprehensive overview, the review will set the stage as an essential link between MOF chemistry and the development of sustainable and viable hybrid supercapacitor devices.

2. Fundamentals of Hybrid Supercapacitors

Electrochemical energy storage devices that integrate the advantages of electric double-layer capacitors (EDLCs) and rechargeable batteries or pseudocapacitors are referred to as hybrid supercapacitors (HSCs) [24]. The two electrodes in a typical HSC provide two different types of charge storage that enable the device to deliver a balance of energy density, power density, rate capability, and cycling stability. The basic function of the capacitor-type electrode is to store charge by reversible accumulation of ions at the electrode/electrolyte interface, while the battery-type electrode is a Faradaic electrode that is characterized by the transfer of electrons and ions. These mechanisms complement each other and enable HSCs to overcome some of the inherent limitations of purely capacitive or purely battery-type systems [25].

Because these devices can realize the improved kinetics of the EDLC materials while improving the energy storage capacity of the pseudocapacitive or battery electrode materials, the hybrid electrochemical capacitors with different energy storage mechanisms for each electrode (e.g., EDLC/pseudocapacitive and EDLC/battery electrode) are very effective in improving the electrochemical performance. An overview of hybrid supercapacitors is shown in Figure 1.

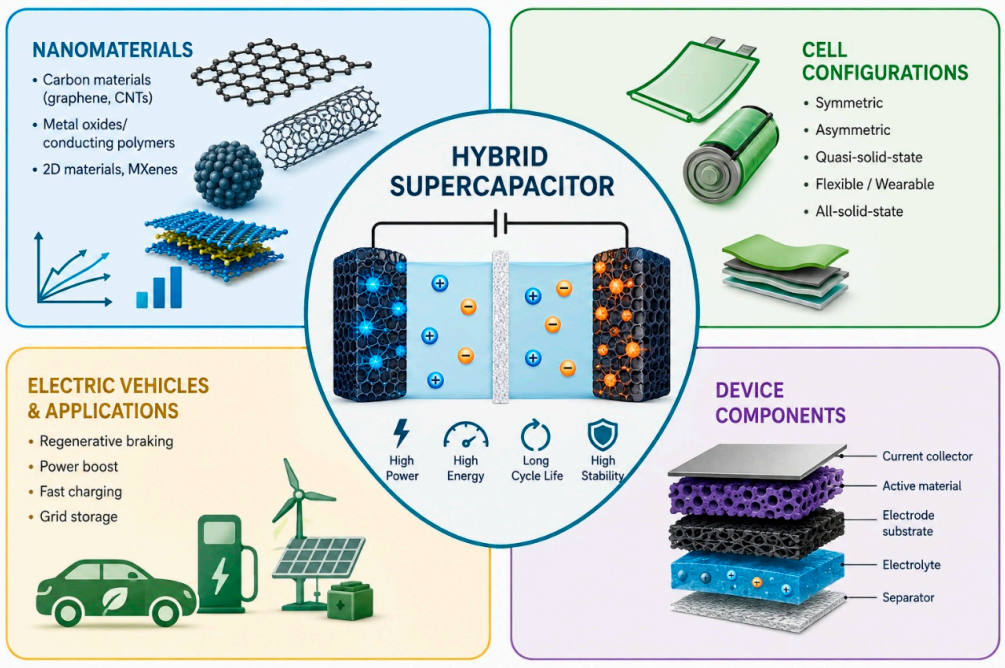


Figure 1. Overview of hybrid supercapacitors.

2.1. Charge-Storage Mechanisms

Charge storage in supercapacitors is commonly classified in three ways: electric double-layer capacitance (EDLC), pseudocapacitance, and battery-type Faradaic storage [26], as shown in Figure 2. The major source of EDLC is the accumulation of electrostatic charge at an electronically conducting electrode/electrolyte interface. Due to the lack of any significant chemical change in electrode material, EDLC typically can provide fast charge/discharge rates and high cycling stability. Common EDLC materials include porous carbon, graphene-based materials, carbon nanotubes, and activated carbon. They are affected by surface area, pore size distribution, size of electrolyte ions, pore connectivity, and electronic conductivity [26].

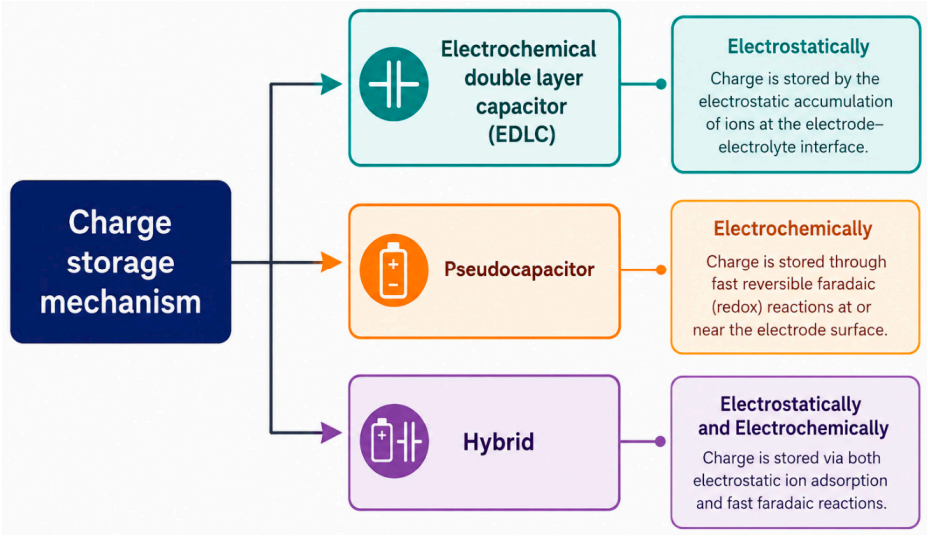


Figure 2. Classification of charge-storage mechanisms into EDLCs, pseudocapacitors, and hybrid supercapacitors.

Pseudocapacitive storage is based on fast and reversible Faradaic processes near or at the electrode surface. The processes may involve surface redox reactions, ion intercalation, or electrosorption with charge transfer. Pseudocapacitive behavior can be found in transition-metal oxides, hydroxides, sulfides, phosphides, conducting polymers, and redox-active MOFs. While ideal EDLCs can be stored only by electrostatic surface adsorption, pseudocapacitive materials can store a much higher charge-storage capacity. The kinetics and the reversibility of the structure of Faradaic reactions are, however, increasingly important, especially at high current densities [27].

Another type of Faradaic storage is battery-type electrodes in which the charge storage is linked to redox reactions that often involve significant ion insertion, conversion, or phase transformation. They have a high specific capacity but may show slower kinetics and more structural changes than the conventional capacitive electrode materials. The difference is used by HSCs, which involve a high-power capacitor-type electrode and a high-capacity battery-type electrode [28].

2.2. Motivation for Hybrid Supercapacitors

The primary driver for HSC development is the well-known energy density vs. power density trade-off. The energy stored in a capacitor is written as follows:

$$E = \frac{1}{2} CV^2$$

where is E the stored energy, C is capacitance, and V is the operating voltage. This correlation points out two key pathways to improving energy density: higher effective capacitance and a wider operating voltage window. The latter is especially significant, as energy density is proportional to the square of the voltage [29].

For a practical asymmetric or hybrid device, the operating voltage will be limited by the electrochemical stability of the electrode/electrolyte combination. Therefore, it is critical to balance the electrodes. The amount of charge that can be stored on the positive and negative electrodes should be suitably balanced [30]:

$$q_+ = q_-$$

and, in simplified form, as follows:

$$m_+ C_+ \Delta V_+ = m_- C_- \Delta V_-$$

where m, C, and V represent electrode mass, specific capacitance or capacity, and potential window, respectively. Appropriate mass balancing prevents overcharging of either electrode or allows the available cell voltage to be utilized effectively [31].

The practical energy and power characteristics of an HSC are therefore determined not only by the intrinsic properties of the electrode materials but also by electrode mass ratio, electrolyte composition, cell configuration, internal resistance, active-material loading, and voltage stability. This distinction is particularly important for MOF-based systems, where impressive results obtained from small active-material masses may not necessarily translate to equivalent improvements at the full-device level.

2.3. Electrode Architecture and Charge Transport

The operation of an HSC is controlled by the coupled flow of electrons in the electrode and ions in the pore network filled with the electrolyte. As the thickness of the electrodes and loading of the active material are increased, these processes become increasingly complex. As represented in Figure 3, a porous electrode should have continuous low-resistance pathways to the current collector with rapid access for electrolyte ions to electrochemically active sites across the electrode [32]. Significant resistance along this route may result in low utilization of the active site and rate capability. So, electrode architecture is not just a physical configuration of active material; it is a key factor in electrochemical kinetics.

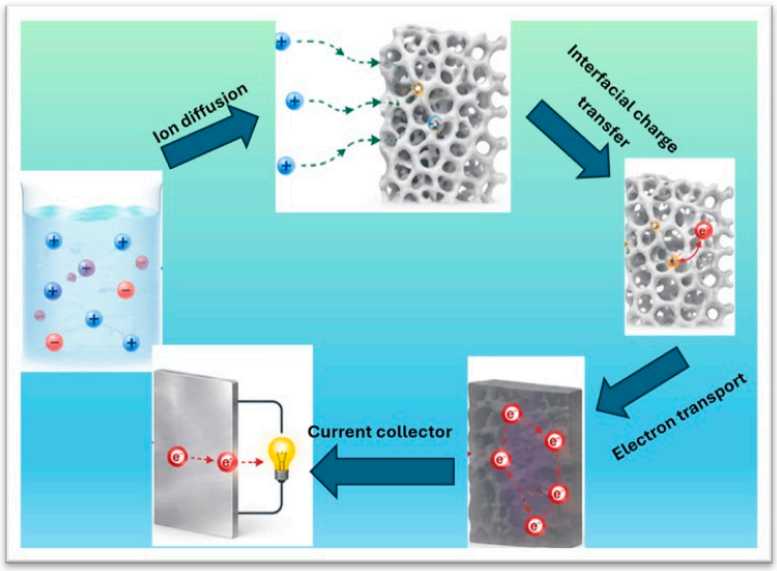


Figure 3. A simplified representation of the transport process.

2.4. From Material Properties to Device Performance

One of the fundamental issues being increasingly considered in HSC research is the difference between intrinsic material performance and practical device performance. The materials of electrodes are often compared by various parameters, including specific capacitance, specific capacity, rate capability, and cycling retention. These parameters can, however, be significantly influenced by testing configuration and some experimental conditions [33].

Three-electrode measurements are especially helpful for studying the intrinsic electrochemical characteristics of an electrode but are not completely representative of the limitations of a complete HSC. Therefore, two-electrode full-cell measurements are necessary to establish realistic energy density, power density, voltage stability, internal resistance, and cycling behavior [34].

This is especially important for 3D MOF architecture. The thickness or loading of an electrode can also be increased to store a greater amount of charge but may also result in

increased ion-diffusion distance and electronic resistance. Unfortunately, highly porous structures can offer excellent electrolyte accessibility but have a relatively low percentage of electrochemically active material. The optimum architecture thus needs to consider the utilization of the active materials, the efficiency of transport, mechanical integrity, and volumetric performance [35].

2.5. Relevance to MOF-Based Hybrid Supercapacitors

The composition and structure of MOFs can be manipulated on different length scales, which makes it an unusual opportunity to manipulate these parameters. The redox properties and coordination environments at a molecular level are defined by metal centers and organic linkers, respectively [36]. The size of the particles, defects, crystallinity, and pore structure are important at the nanoscale in the interaction with the electrolyte, as well as in charge transport. The MOF particles can be connected to conductive networks and hierarchical pores at the microscale. Finally, at the device level, these components can be combined in 3D architectures to control the flow of electrolytes, electron conduction, and mechanical stability [27].

Wang et al. [37] developed two electrode materials with different energy-storage mechanisms: the three-dimensional (3D) nanostructured MoS_2–ZIF composite and the porous carbon polyhedra in the form of a capacitor. The X-ray diffraction (XRD) analysis (Figure 4a) of the MoS_2–ZIF composite shows broader (002) reflections at 14.4° (2 theta), indicating few-layer MoS_2 nanosheets on ZIF-8-800 polyhedra. The Raman spectrum (Figure 4b) reveals Mo–S vibrations between 380 and 407 cm^{-1} and carbon peaks at 1355 and 1597 cm^{-1}, differing from pure MoS_2 and ZIF-8-800. Figure 4c illustrates the charging and discharging curves of the composites at a current density of 100 mA g^{-1}, showing two voltage plateaus at approximately 1.2 V and 0.6 V during the initial discharge, indicative of a Li^+ insertion reaction that produces Li_xMoS_2 and a subsequent reaction to form Li_2S and metallic Mo, while the composite achieves specific capacities of 978 and 802 mA h g^{-1} at current densities of 0.1 and 1 A g^{-1}, respectively (Figure 4d).

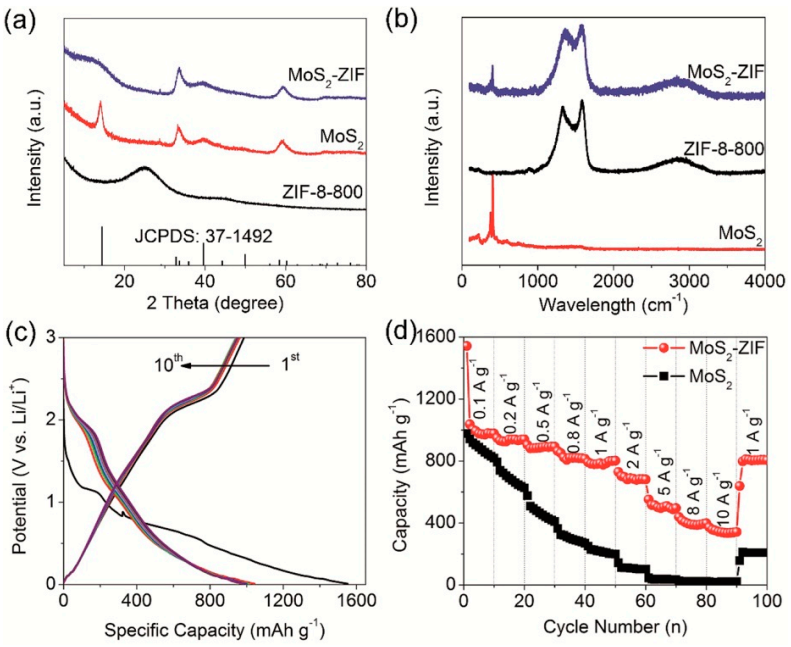


Figure 4. (**a**) XRD patterns and (**b**) Raman spectra of the MoS_2–ZIF composite; electrochemical performance of MoS_2–ZIF in a half cell: (**c**) the typical charging/discharging curves of the MoS_2–ZIF electrode within the potential window of 0.01–3 V (vs. Li/Li^+) at 0.1 A g^{-1}, (**d**) rate capability of pure MoS_2 and MoS_2–ZIF electrodes from 0.1 to 10 A g^{-1} [37].

3. Metal–Organic Frameworks for Hybrid Supercapacitors

Metal-organic frameworks (MOFs) are crystalline porous materials used in a wide range of applications, including electrochemical energy storage. They are built by coordination of metal ions or metal clusters and organic ligands to form well-defined architectures that have tunable pore environments, high surface areas, and a variety of chemical functions. MOFs have an unusually wide compositional and structural design space: both the inorganic nodes and the organic linkers can be systematically varied. This flexibility opens the door to controlling redox properties, porosity, surface chemistry, electronic structure, and interactions between the electrolyte to create rational hybrid supercapacitor electrodes [38].

3.1. Structural and Chemical Characteristics Relevant to Energy Storage

The metal centers, organic linkers, coordination environments, pore structures, and defect populations play a significant role in the electrochemical behavior of MOFs. Redox-active centers can be provided by metal nodes, and the organic linkers may provide charge storage or tune the electronic and chemical environment around the metal nodes. The Faradaic processes that may occur include reversible oxidation-state changes, ion adsorption, surface redox reactions, etc., in MOFs, depending on the composition and coordination chemistry [39].

One very relevant feature of MOFs is their porous structure, which is relevant to supercapacitors. Their interconnected pores can offer accessible pathways for the penetration of electrolyte and enhance the electrode–electrolyte interfacial area. But do not think that a high nominal surface area equates to high electrochemical surface area. The pore accessibility is determined by the electrolyte ion size, pore size, surface chemistry, pore connectivity, and stability of the pore walls under operating conditions. Thus, the design of MOFs for HSCs should be not only based on the maximization of surface area but also on the design of porosity that will be accessible to the electrode reaction process.

The chemical environment of the framework can also affect the adsorption of ions and charge transfer at the interfaces. The polarity and charge distribution of the pore walls can be modified by functional groups in the organic linker, coordinatively unsaturated metal sites, defects, and guest species. These features can increase the interaction between electrolyte and active sites and make the active sites more accessible. In aqueous electrolytes, proton or hydroxide-ion interactions may play a significant role in Faradaic storage processes [40].

3.2. Redox-Active MOFs

One of the most direct methods of improving the energy-storage capacity of MOFs is the incorporation of redox-active metal centers. Transition elements like Fe, Co, Ni, Mn, Cu and V can exhibit reversible changes in oxidation state in an appropriate coordination sphere. These processes can enable extra Faradaic charge storage in addition to the typical double-layer capacitance [41].

In addition, Fe/Mn-based frameworks may offer additional redox-active sites, and Ni/Co-based frameworks may have more than one accessible oxidation state. The added level of control comes from the ability to have two or more species of metal ions that can alter the electronic structure and coordination environment of the framework in mixed-metal MOFs [42]. This compositional engineering could facilitate synergistic redox reactions, create a larger number of electrochemically available sites, or change the ion-transport properties.

Park et al. [43] found that if highly nanoporous MOF-derived carbon was chemically activated using potassium hydroxide, the resulting carbon material had an extremely high surface area. This activated carbon was used in combination with a polyaniline (PANI nanoarchitecture) to introduce additional redox-active sites, which proved to be suitable for

EDLCs. The PANI nanorods not only effectively supported these sites but also increased the capacitance of the material. The capacitance of the carbon composite materials derived from PANI-ZIF-8 was as high as 635.3 F/g, which was much higher than that of ZIF-8-derived carbon (143.4 F/g) when tested at a scan rate of 5 mV/s. Further, the composite also showed strong cycling durability, indicating the possibility of its use as a high-performance hybrid electrode material for supercapacitors.

Gupta et al. [44] have developed a 3D nanostructure by integrating reduced graphene oxides (rGO) with dual-functionalized MOFs using an ultrasonication method, which increases the number of electroactive sites on the electrode, as illustrated in Figure 5.

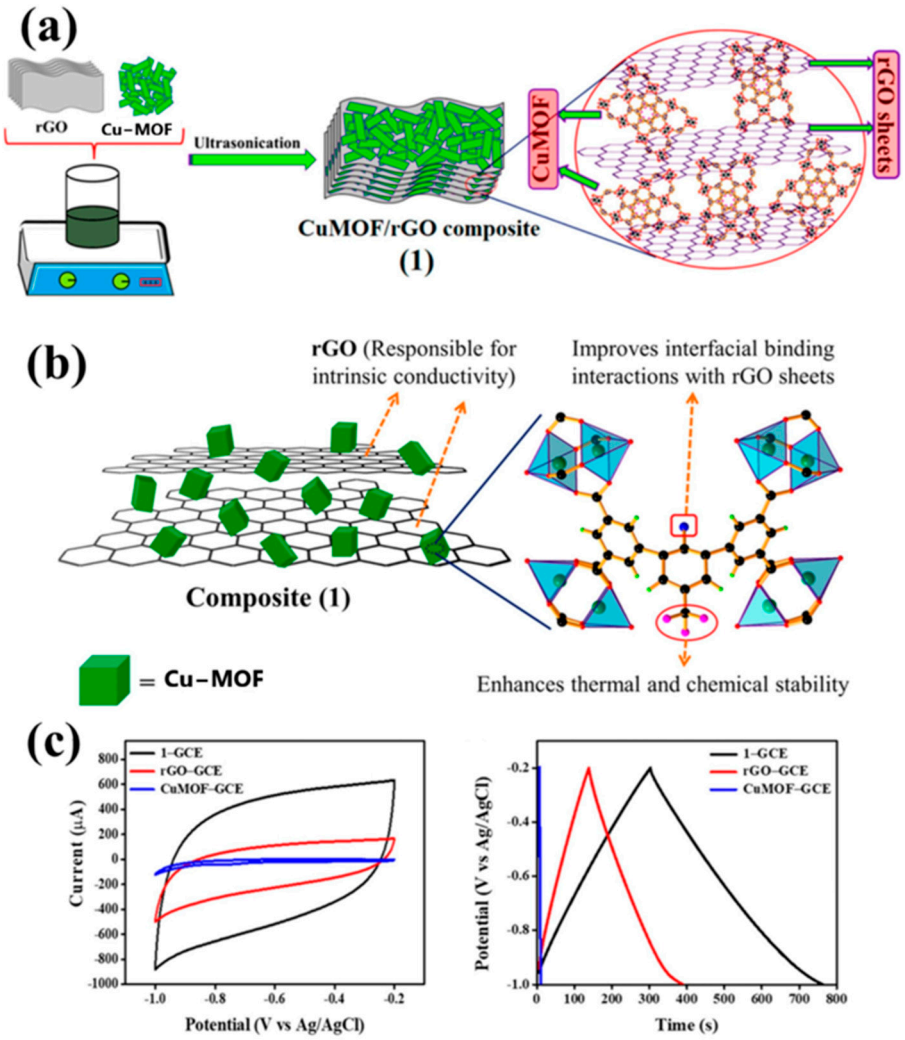


Figure 5. (**a**) Synthesis scheme of CuMOF/rGO composite (**b**) The potential method for fabricating composite between CuMOF and rGO. (**c**) Comparison of electrochemical performance of pristine and composite samples [44].

Bimetallic and trimetallic MOFs are especially relevant to the development of HSCs; the presence of multiple metal centers can render properties that do not appear in a single-metal framework. Charge storage behavior can be affected by metal–metal interactions, electronic redistribution, defect formation, and differences in redox potential. For instance, Otun et al. [45] used a simple one-step solvothermal method to synthesize and optimize the bimetallic MIL-125(Ti,Mn) MOF to improve its electrochemical properties in hybrid supercapacitors. The flake-like sheets and spindle-like morphology of MIL-125(Ti,Mn) enhance their interaction with electrolyte ions, which enhances their performance. The optimized electrode comprised of MIL-125(Ti)-Mn-2 yielded a specific capacity of 137.2 $mAhg^{-1}$ at

1 A g^{-1} and over 76.3% capacity after 5000 cycles. The activated-carbon Mn-MIL-125(Ti)-2 asymmetric supercapacitor exhibited a specific energy of 22.74 Wh kg^{-1} when operated at a current density of 996 W kg–1 and retained 71.2% of its capacitance after 10,000 cycles at 10 Ag^{-1}.

3.3. Defect Engineering of MOFs

Another important strategy to modify the electrochemical properties of MOFs is defect engineering. Defects such as missing-linker, missing-cluster, vacancy, and substitutional defects can change the structure of pores, the coordination environment, the electronic structure, and chemical reactivity. Controlled defect generation can reveal hitherto-unaccessed metal sites, open pore space, provide extra adsorption sites, and allow electrolyte penetration [46].

In the scope of supercapacitor applications, defects thus may affect both charge-storage capacity and transport kinetics. Too many defects, however, can affect the crystallinity and stability of the framework. The correlation between defect density and electrochemical performance is therefore non-linear, and the optimum defect concentrations will vary with the framework composition, electrolyte, operating potential, and device architecture.

Nwaji et al. [47] studied 3D $NiCo_2S_4$ nanospikes embedded in a 1D Fe_3C microarchitecture by chemical surface transformation. In the resulting Fe_3C@$NiCo_2S_4$ nanospikes, high specific capacities of 1894.2 F g^{-1} and a rate capability of 59% were obtained, which have great application potential in high-performance hybrid supercapacitors. They exhibit outstanding cycling stability (92.5%) and Coulombic efficiency (98.7%). The improved charge storage in this case is due to the active components, increased active site availability, and efficient redox reactions. Nitrogen-doped carbon nanofibers are used to make hybrid supercapacitors with energy/62.98 Wh kg^{-1} and power/6834 W kg^{-1}, which shows that the hybrid supercapacitors are stable for 5000 cycles with 95.4% stability.

It is important to note that the defects need to be understood in the context of the electrode architecture. The electrochemical behavior of defect-rich MOFs integrated into conductive 3D networks can be very different from that of the same material when used as a traditional powder electrode. This relationship between intrinsic defects and extrinsic transport pathways, however, is an area that needs further study.

3.4. Conductivity and MOF–Conductive Material Integration

Many of the conventional MOFs suffer from low electronic conductivity despite their attractive porosity and tunability in terms of their chemical properties. This could affect electron transport in thick layers of MOFs and result in low utilization of the electrochemically active sites. Therefore, several strategies have been developed to enhance charge transport, such as the incorporation of carbon-based conductive materials, conducting polymers, metal nanoparticles, MXenes, and conductive MOFs [48].

The porous properties of the MOF can also be retained simultaneously with the electron-conduction properties of carbon materials, making MOF–carbon composites especially attractive. Various conductive components such as graphene, reduced graphene oxide, carbon nanotubes and carbon nanofibers, porous carbon and biomass-derived carbon have been studied. Additional pseudocapacitive storage can be gained using conducting polymers [49].

The best designs are not just a combination of MOFs and conductive additives. They would rather create close proximity contacts and uninterrupted transport channels. Improved accessibility to active sites and reduced interparticle resistance can be achieved by directing MOF growth on conductive substrates and/or by in situ formation of MOF/carbon interfaces.

Chen et al. [50] synthesized 2D conductive metal–organic frameworks (CMOFs) and conductive polypyrrole (PPy) based on hydrothermal methods. The hollow structure of PPy inhibits the aggregation of CMOF and increases the conductivity of the electrode; thus, the performance of the core–shell PPy@NiCo-CAT electrode is 572.2 F g^{-1}. As seen in Figure 6, this could be employed as an active material in hybrid supercapacitors (HSCs) with very high energy density and durability, which would enable two HSCs in series to power 12 LEDs. The approach of fusing together conductive polymers and 2D CMOFs presents new possibilities for high-performance electrode materials for HSC applications.

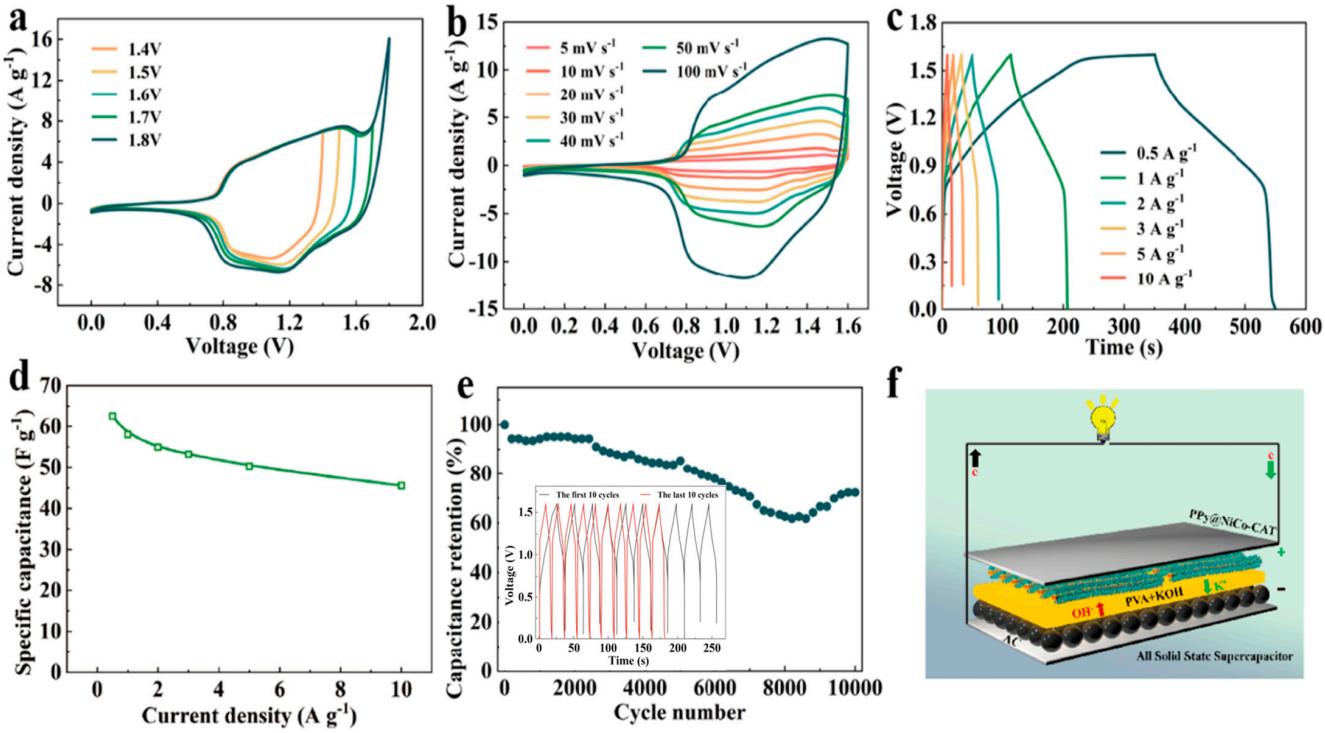


Figure 6. (**a**,**b**) CV curves of the HSC at different voltages and various scan rates, (**c**) GCD curves of the HSC, (**d**) specific capacity as a function of current density, (**e**) cycling performance of the HSC at a current density of 5 A g^{-1}, and (**f**) schematic illustration of the HSC. Inset of (**e**): comparison of the first 10 cycles and the last 10 cycles of HSC [50].

3.5. MOFs as Precursors for Electroactive Derivatives

One of the significant extensions of MOF chemistry is that they can be used as sacrificial templates or precursors to assemble porous inorganic and carbon-based materials. MOFs can be partially preserved and converted into metal oxides, hydroxides, sulfides, phosphides, carbides, nitrides, metal nanoparticles, and metal/carbon composites through thermal or chemical conversion methods.

MOFs can be combined with the conductivity and porous structure of carbon and the compositional advantages of transition-metal compounds to overcome some of the limitations of pristine MOFs to make MOF-derived materials [51]. For instance, a controlled conversion of bimetallic or trimetallic MOFs can be used to generate multiple redox-center multicomponent systems such as oxide, sulfide, phosphide, or metal/carbon systems, with heterogeneous interfaces. For instance, Yang et al. [52] have synthesized NC/Ni–Ni_3S_4/CNTs by carbonization and vulcanization of MOFs/CNTs, resulting in the uniform distribution of Ni_3S_4 in a 3D conductive network. This architecture allows for better conductivity and entry of electrolyte, thereby increasing the reaction kinetics. The material has a specific capacitance of 1489 F/g at 1 Ag^{-1} and an energy density of 39.8 Wh kg^{-1}, along with a power density of 749.8 W kg^{-1}, with excellent cyclic stability.

3.6. From MOF Materials to 3D Electrodes

One of the main problems with material approaches to 3D electrodes has been the difficulty of achieving efficient operation in a real electrode when the intrinsic properties are good. As the composition of MOFs has been growing more complex, an important limitation of material-centric approaches is that there is no guarantee that good intrinsic properties can be achieved within a practical electrode. After the MOFs have been included in the electrode, particle connectivity, distribution of active material, contact of the active material with the current collector, penetration of the electrolyte, electrode thickness, and mechanical stability all play a role.

This offers straightforward motivation for the combination of MOFs with 3D architecture. This conductive 3D scaffold can serve both as a current collector and mechanical support, as well as an electron-transport network and a structural template for the expansion of MOFs. The direct integration of MOFs in such architectures can help decrease the need for polymer binders and conductive additives and increase the utilization of the active material [53].

Hence, the creation of MOF-based HSCs is increasingly moving away from "Designing better MOFs" to "Designing electrochemical architectures with MOF materials". This transition is necessary for the next step, which is the design of 3D architectures to benefit from the chemical and structural benefits of MOFs and overcome transport, loading, and device limitations.

4. Three-Dimensional Electrode Architectures for MOF-Based Hybrid Supercapacitors

A step towards translating inherent MOF properties to practical hybrid supercapacitor devices is the shift from the powder-based electrodes to three-dimensional (3D) architectures. In a conventional electrode, the particles that are responsible for electrochemical reactions are combined with conductive additives and polymeric binders and are then deposited onto a current collector. While this is practical, the electrode created could include inactive components, discontinuous electron pathways, interparticle interfaces, and tortuous electrolyte-access pathways. These limitations can become even more severe if the thickness of the electrode increases or the loading of the active materials in the electrodes is increased. Such limitations can be further exacerbated by a thicker electrode and/or a higher loading of the active material(s) in the electrode. Three-dimensional architecture provides an alternative by spatial organization of the active material and conducting framework to create a network of interconnected pathways for electron transfer and electrolyte access [54].

4.1. Concept and Advantages of 3D Electrode Architectures

An electrode that can be described as a three-dimensional (3D) electrode is one that has a framework that is conductive or porous, and that embeds the active material throughout the structure. In contrast with traditional 2D electrode coatings, the 3D architecture introduces an extra dimension to control the distribution of active sites, electrolyte pathways, and electronic conduction networks. In the case of MOF-based electrodes, this structure can enhance electron- and ion-transport pathways, expand the electrode–electrolyte interface, and promote the penetration of the electrolyte and utilization of the active site. In addition, 3D structures can increase mechanical stability and decrease the use of polymeric binders and conductive additives and allow for a higher loading of active material. The advantages of 3D architectures render them particularly desirable for the enhancement of the electrochemical functionality and practical applications of MOF hybrid supercapacitors [55].

4.2. Conductive 3D Scaffolds

One of the most straightforward methods for the creation of MOF-based 3D electrodes is the use of conductive scaffolds. Typical substrates include nickel foam, stainless-steel mesh, carbon cloth, carbon felt, graphene aerogels, carbon foams, and other porous conductive structures [56]. Conductivity, mechanical strength, and commercial availability make metal foams very appealing. Intimate contact between the active phase and current collector can be achieved by direct growth of MOFs on the current collector, for instance on nickel foam, which removes some of the interparticle resistance of slurry casting the electrodes. The electrochemical contribution from the scaffold must, however, be separated from that of the MOF, especially when comparing performance between different electrode configurations [57].

When sustainability, flexibility and chemical stability are considered, carbon-based scaffolds offer other benefits. Carbon cloth and carbon felt can be used to create interconnected conductive pathways, and graphene and carbon aerogels can create high surface areas and hierarchical porous structures. Renewable precursors can be used to make porous conductive frameworks, which is an added layer of sustainability with biomass-derived carbon. Recently, our group [58] synthesized a ternary composite electrode by solvothermal and in situ oxidative polymerization techniques, which consisted of a zeolitic imidazolate framework (ZIF-67), polyaniline (PANI), and cellulose nanocrystals (CNCs) to improve the performance of supercapacitors. A high specific capacity of 414.05 C g^{-1} at 1 A g^{-1} was obtained for this ZIF-67/PANI/CNC composite, with a stability of 81.2% after 5000 cycles in a three-electrode configuration. Furthermore, an assembled asymmetric supercapacitor (ZIF-67/PANI/CNC//AC) with specific energy of 61.6 Wh kg^{-1} at 1.12 kW kg^{-1} was fabricated. The synergistic effects of ZIF-67 redox sites and efficient electron transport by PANI/CNC account for the improvements.

4.3. Direct Growth Versus Ex Situ Integration

The direct growth or ex situ assembly of MOFs into 3D architectures can be used. Direct growth methods involve the nucleation and growth of MOF crystals directly on the conductive scaffold. This approach allows for good interfacial adhesion and relatively even distribution of active material. It can also reduce the amount of polymer binders required. It is thus crucial to control the nucleation density, crystal size, growth orientation, and coverage for an effective electrode architecture [59].

Ex situ methods include the synthesis of the MOF out of the host structure before infiltration, assembly, and deposition. This route offers more flexibility for the optimization of the MOF synthesis process without regard to the scaffold. It is not necessarily easy to get uniform distribution in a thick 3D structure, however. Poor internal connectivity and particle aggregation can result in limited electronic and/or ionic access [60].

4.4. Hierarchical Porosity and Ion Transport

3D architecture is one of the most important factors in controlling ion transport. There are several length scales with which it is possible to consider the porosity of an electrode. There are micropores, which can offer high surface area and ion storage sites; mesopores, which can provide the electrolyte with a pathway for diffusion; and macropores, which can provide an electrolyte pathway through the electrode [61].

Hierarchical porosity can be achieved by MOF-based HSCs because of the intrinsic microporosity of MOFs, along with the porosity of the 3D scaffold at the larger scale. The structure can also help to address the disadvantages of using only the nanoscale pores of the MOF. The bigger channels enable fast electrolyte movement, and the smaller pores offer electrochemically active interfaces. For instance, Pan et al. [62] developed Ni-MOF

on a fishbone-like Ni_3S_2/Co_3S_4 (NCS), resulting in a 3D porous structure, NiMOF@NCS (Figure 7). This material exhibits excellent capacitive behavior due to its interconnected structure, showing significant active sites and electrical conductivity, with a specific capacity and 65% retention. A hybrid device, NiMOF@NCS/NF//AC, was fabricated and demonstrated good rate capability with consistent CV profiles. The GCD analysis indicated symmetrical triangles and minimal IR drop, retaining 61.5% capacity at increased current densities. A Nyquist plot illustrated rapid ion movement and enhanced conductivity. The device achieved an energy density of 58.8 Wh kg^{-1}, retaining 96.2% capacity after 10,000 cycles, showcasing a novel approach to modifying MOF morphology and electrochemical properties.

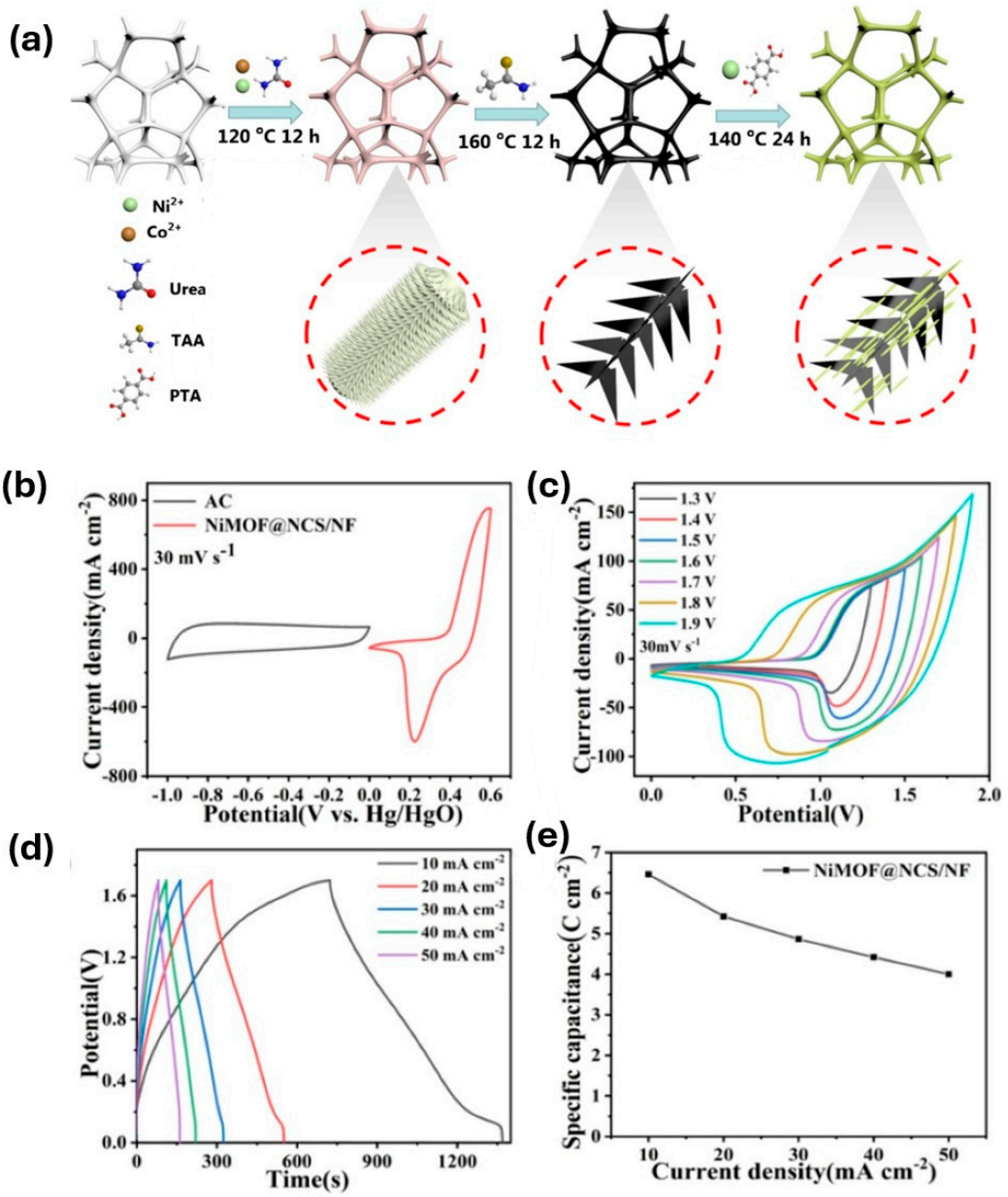


Figure 7. (**a**) Preparation scheme of NiMOF@NCS. (**b**) CV graph of AC and NiMOF@NCS/NF. (**c**) CV of the device. (**d**) GCD of the device. (**e**) Capacity at different current densities [62].

4.5. Three-Dimensional Architecture and Active-Material Loading

One of the main benefits reported for 3D electrodes is the ability to load the active material into the electrode without a corresponding increase in transport limitations. This is not something that is granted automatically, though. Pores may be blocked, and the thickness of the active layer can increase as MOF loading increases. However, architecture that is efficient at low loadings can exhibit significantly different performance at practical loadings [63].

This is especially significant for comparing literature reports. A very small amount of active material, deposited on a highly conductive scaffold, may lead to an increase in gravimetric capacitance, and the overall contribution of the current collector and inactive components may grow. Areal and volumetric performance should, therefore, complement gravimetric performance in practical HSCs.

All these factors, namely active-material loading, accessible surface area, ionic transport, electronic conductivity, and mechanical stability, should be rationally considered in the design of 3D architecture. Balancing these two is one of the key design considerations that high-performance 3D morphological electrodes based on MOFs need to achieve.

4.6. Freestanding and Binder-Free Architectures

Another significant direction for the development of 3D devices is in the form of freestanding electrodes. In such systems, electrodes can be used without the typical use of a metallic current collector or a polymeric binder, which may lead to lower inactive mass and interfacial resistance. Structures that can be engineered into freestanding electrodes include MOF-derived carbon networks, MOF/carbon aerogels, cellulose-derived frameworks, and self-supporting composite films [64].

Recently, Abbas et al. [65] achieved a redox-active Ni MOF (NiFc-MOF) on electrospun carbon nanofibers (CNFs) by an in situ method, which overcame the problem of MOF aggregation. This electrode is flexible and has high redox activity of ferrocene and carbon nanofibers, which gives it remarkable characteristics of high flexibility and lightness. It also achieved a specific capacity of 460 C g^{-1} in supercapacitors without the use of a binder, showing 82.2% cyclic retention after 25,000 cycles. The flexible asymmetric supercapacitor device achieved a high energy density of 56.25 W h kg^{-1} and had very good cycling performance (Figure 8). The electrode also exhibited high stability and flexibility in the flexibility device using various bending angles, having very little difference in cyclic voltammetry curves.

Eliminating the binder can increase electrical contact and decrease electrochemically inactive components. However, mechanical properties and flexibility, cyclic properties of the structure, and scalability for fabrication become more significant. A successful freestanding electrode, therefore, should not only offer high electrochemical activity but also ensure adequate mechanical strength to preserve its architecture during repeated charge–discharge operation. For instance, He et al. [66] created a metal–organic framework (MOF)-to-CNT-carbon cloth (VGC) hybrid structure by converting MOFs into CNTs on the VG surface. The structure was employed to develop electrode material for hybrid supercapacitors using a KOH electrolyte containing redox additives without a binder. The CNT-VG-CC electrode showed a superior areal capacitance of 1526.5 mF/cm^2 at 10 mA/cm^2, with almost 100% coulombic efficiency and excellent cycling stability, with 100% capacitance after 10,000 cycles. The supercapacitor device exhibited a high energy density of 179.7 μW h/cm^2 at a power density of 8000.0 μW/cm^2 and exhibited a high areal capacitance of 118.9% even after 10,000 cycles due to its unique structure, super-hydrophilicity, and high electronic conductivity.

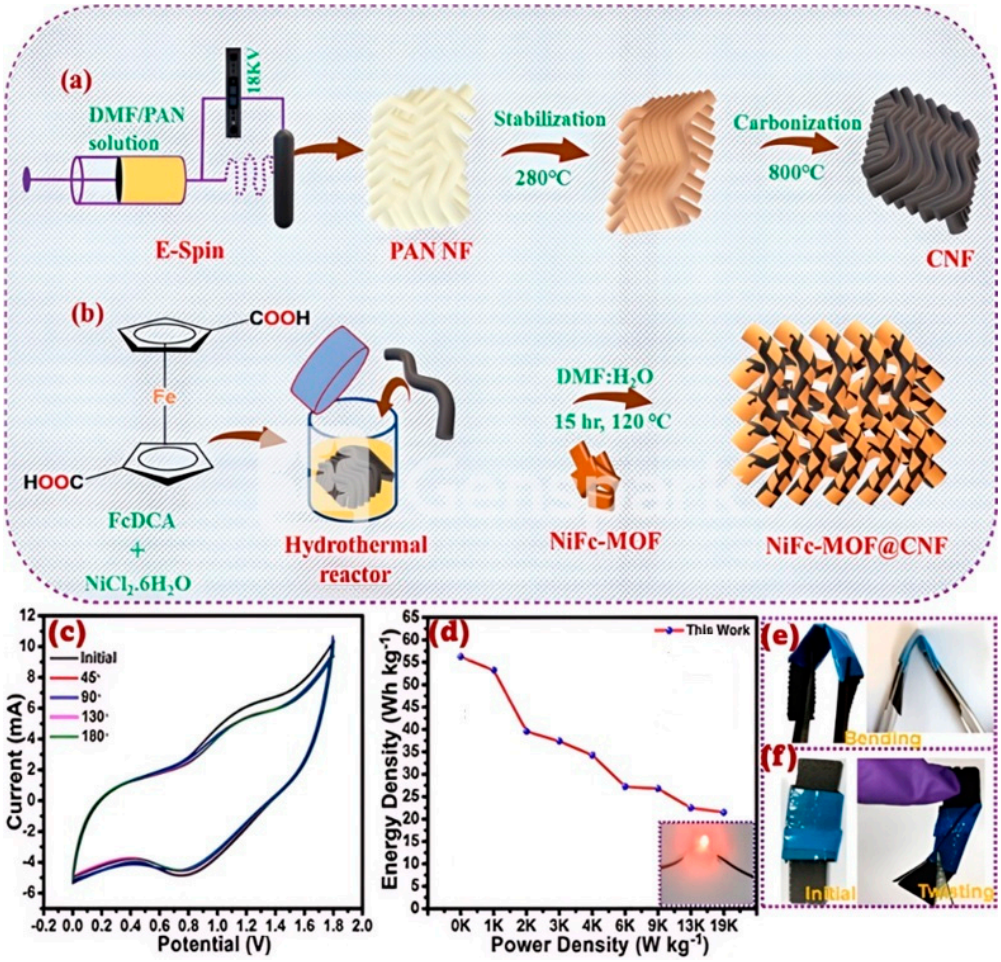


Figure 8. (**a**) Schematic illustration for the preparation of CNFs and the redox-active NiFc-MOF@CNF-based flexible electrode. CV curves of the flexible device at different angles. (**b**) Ragone plot showing a comparison of energy density vs. power density, and the inset shows a photograph of a lit LED. (**c**) Flexible device in bending mode. (**d**) Flexible device in initial and twisting modes (**e**) flexible device in bending mode. (**f**) flexible device in initial and twisting modes [65].

4.7. 3D Printing and Architecturally Controlled Electrodes

Additive manufacturing offers a new tool for controlling the geometry of the electrode, which is otherwise limited to natural porous scaffolds. 3D printing would also allow for the precise control of the thickness of the electrodes, the dimensions of the channels, the porosity, and the distribution of active material in space [67]. This is especially appealing for MOF-based systems as the architecture can be tailored to the anticipated ion and electron transport needs, instead of just randomly distributed pores.

Hierarchical channels that can be 3D printed onto MOF electrodes could act as dedicated electrolyte pathways but also provide continuous electronic pathways. These kinds of designs may also enable tailored electrode geometries for flexible, wearable, and microscale energy-storage devices [68].

Xie et al. [69] reported a 3D-printed micro-supercapacitor (MSC) based on a heterostructured COF@MOF-Ti_3C_2Tx electrode material with a designed nanoporous structure and enhanced electrochemical activity. The material achieves a capacitance value of 1078.0 F g^{-1}, and its material properties include rapid ion diffusion, high ion adsorption, excellent electron conductivity, and excellent pseudocapacitive reversibility. It also exhibits low ionic diffusion impedance and structural stability. The assembled micro-supercapacitor has an energy density of 60.3 μWh cm^{-2} and cyclic stability, which could be used for

powering intelligent traffic signals and alarm clocks, pointing towards the direction of high-performance micro-energy storage devices.

Mokhtarnejad et al. [70] synthesized a bi-MOF-rGO hybrid nanocomposite (HNC) efficiently using laser ablation synthesis in solution (LASiS) (Figure 9). The specific capacitance of this material is very high (1092 F g^{-1}) at 1.0 A g^{-1} in 0.5 M Na_2SO_4 electrolyte. Besides this, the composites have been successfully mixed with different solvents, viscosity modifiers, in-house synthesized porous carbon (PC), and commercially available graphene and binders to develop a high-performance 3D-printed active layer ink for sequential inkjet printing of high-performance supercapacitors.

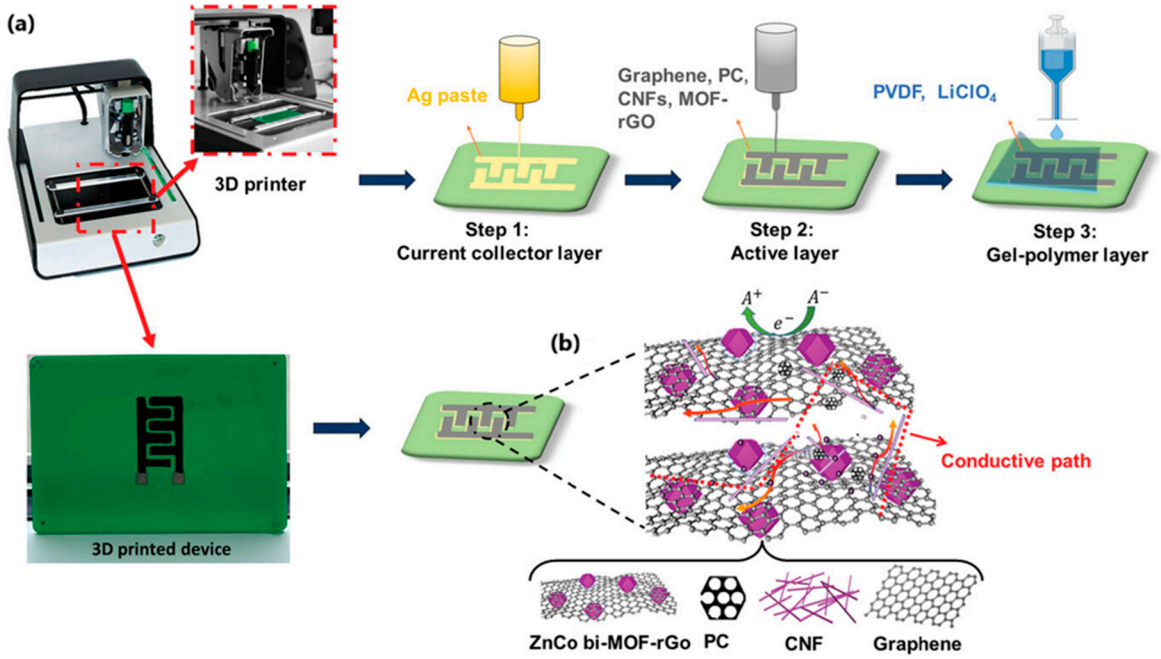


Figure 9. Schematic illustrations of (**a**) sequential layer-on-layer method for the 3D-printed SC electrodes; (**b**) percolated conductive network of PC, CNFs, MOF-rGO, and graphene nanosheets [71].

Nevertheless, the challenges of 3D printing for sustainability and scalability are also present, such as formulation of printing inks, requirement of binders, printing energy consumption, solvent use, post-treatment, and reproducibility. Therefore, it is not only the architectural precision but also the overall environmental and processing impact that should be considered when assessing additive manufacturing.

5. Sustainable MOFs and Green Strategies for Hybrid Supercapacitors

The development of advanced MOF-based electrodes traditionally has been fast due to electrochemical performance, quantified by specific capacitance, energy density, power density, and cycling stability. The shift to sustainable energy-storage technologies, however, calls for a more comprehensive evaluation that extends beyond the material's electrochemistry to include its production processes, its resource use, installation in a device, and its final fate. Such a point of view is more important for MOFs, which can employ organic linkers, metal salts, modulators, organic solvents, repeated washing, activation, and energy-intensive post-treatment in their synthesis process. Therefore, the sustainability of an MOF-based HSC cannot be justified using a porous or chemically efficient material only [72].

The production of environmentally friendly and scalable 2D conductive metal–organic frameworks (MOFs) is of vital importance to compete with the cheaper carbon-based electrodes. It has been demonstrated to be effective in reducing the amount of solvents

used and in providing high yields of products, frequently with multiple polymorphs and topologies not found in solution processes. This synthesis method also has potential to offer a sustainable method for MOF preparation, to give rapid access to precise reticular materials, to be scalable, and to reduce energy consumption and waste compared with the conventional methods. This approach avoids the use of volatile organic solvents such as dimethylformamide (DMF), which are widely used in conventional MOF preparation but are harmful to the environment and human health [73].

For instance, the mechanochemical synthesis of Cu-HHTP is linked to the MOF connectivity inferred from the 3D ED-solved crystal structure by Marchetti et al. [74]. The material obtained was incorporated into composite electrodes and tested in symmetric coin cell format using an aqueous electrolyte of 1 M Na_2SO_4 as shown in Figure 10. The electrochemical performance was compared with a solvothermally synthesized one by using two-electrode cyclic voltammetry, galvanostatic charge–discharge, and electrochemical impedance spectroscopy, which were used to determine the capacitance, energy/power output, and the equivalent series resistance. Further, three-electrode cyclic voltammetry in the same electrolyte was also performed to remove the intrinsic redox/electric-double-layer response of Cu-HHTP from the overall cell response. Specifically, the Cu-HHTP prepared by mechanochemical synthesis provides improved supercapacitor performance with a slight increase in series resistance, such as higher reversible specific capacitance, higher energy density, higher power capacity, and cycling stability.

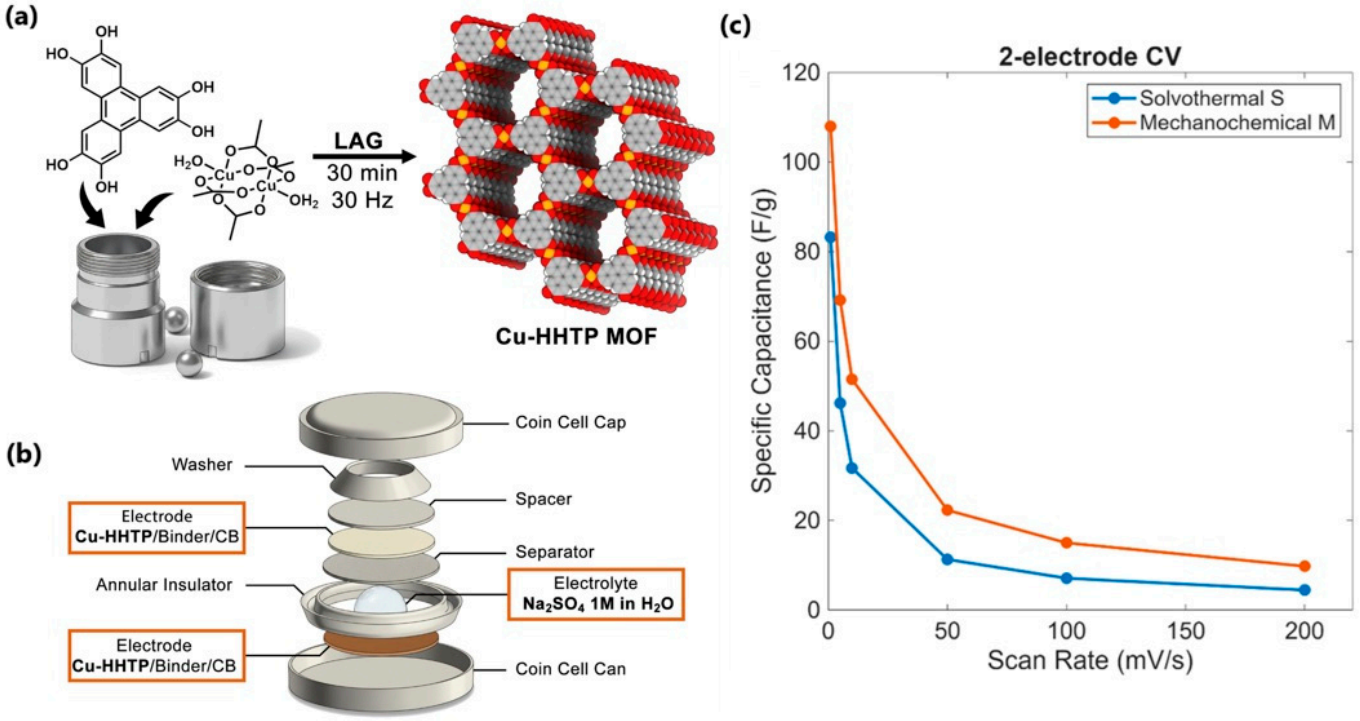


Figure 10. Schematic illustration of the general workflow from MOF synthesis (**a**) device preparation (**b**), and specific capacitance of Cu-HHTP(S) and Cu-HHTP(M) electrodes computed from two-electrode measurements, and (**c**) a plot of specific capacitance against scan rate [74].

Furthermore, costly organic ligand molecules are recycled, reducing the materials cost as well as the environmental cost for the synthesis process. For instance, Xiao et al. [75] synthesized a Co-based metal–organic framework (UPC-9) with the help of 2,3,5,6-tetramethyl-1,4-diisophthalate and prepared ultrathin Co_3O_4 hexagonal nanosheets by hydrolysis and calcination, with a thickness of only 3.5 nm. The nanosheets have an ultrathin 2D structure

and many macroporous and mesoporous features, resulting in a high specific capacitance for the asymmetric supercapacitor demonstrated at 1 Ag^{-1} (1121 Fg^{-1}) and 25 Ag^{-1} (873 Fg^{-1}).

It is important to reduce energy use during MOF synthesis to reduce environmental impact. Traditional methods are energy-consuming, and other technologies such as low-temperature synthesis, microwave-assisted synthesis, and ultrasonic synthesis can reduce energy consumption and ensure or improve the quality of MOFs [76]. However, challenges such as high equipment costs and scalability issues hinder their industrial application, indicating a need for further research. Because BDC is a key component for synthesis of different MOFs, it is logical to think that formulating an efficient approach to extract BDC from plastic waste and using it in the synthesis of MOFs would be economically viable recycling of plastic waste.

Scaling up the eco-friendly synthesis of the green MOF for HSC is critical, especially in terms of maintaining the quality of the material. The challenges are uniformity of properties, efficiency, and cost [76]. New developments in solvent-free mechanochemical processes, microwave-assisted technology, and ultrasonic synthesis techniques have successfully demonstrated the synthesis of MOFs such as ZIF-8 and HKUST-1, maintaining purity and integrity. However, there are some problems, such as batch uniformity, equipment design, etc. Optimized conditions have been demonstrated to improve quality and lower costs in some successful examples such as MIL-101 and UiO-66 and thus make sustainable applications in energy storage possible.

5.1. Defining Sustainability in MOF-Based Energy Storage

Several stages in the life cycle of the material and device should be considered when considering sustainability in MOF-based HSCs. These range from raw-material sourcing, MOF synthesis, post-synthetic treatment, electrode fabrication, electrochemical operation, to end-of-life management. A material with many relatively harmless inputs may have a significant environmental impact if it requires a lot of solvents, high temperatures, and/or washing and/or activation. This demands, however, a deeper evaluation that takes into account raw materials, synthesis, electrode production, operation of the device, and recycling [77].

This overall approach is significant because there is a possibility of disadvantages appearing at one stage and advantages at another. For instance, higher porosity in an MOF can lead to better electrochemical performance but also add extra solvent or activation procedures. Likewise, the addition of a highly conductive component may lead to a better rate capability and make the fabrication of electrodes more complex and environmentally harmful.

Therefore, sustainable MOF development should aim not only to increase the electrochemical functionality but also to increase the ratio between this functionality and the consumption of resources.

5.2. Earth-Abundant and Environmentally Preferable Metal Centers

Metal is another aspect of MOFs that needs to be considered for sustainability. Transition metals like Fe, Mn, Cu, Zn, Mg, Al and Ni are interesting for several reasons, including their availability, their varied coordination chemistry, and their possible redox activity. Their incorporation can lead to MOFs of various oxidation states and coordination environments, which are appropriate for Faradaic charge storage [78].

By incorporating complementary redox processes in the same framework, multimetallic MOFs can further enhance the functionality. The bimetallic and trimetallic systems may offer an additional capacity for charge storage, altered electronic structures, and enhanced structural stability. More metal species added, however, should be supported by tangible

functional advantages. More complex composition could lead to higher synthesis demands and make material recovery/recycling more difficult [79].

5.3. Green Solvents and Aqueous Synthesis

One of the largest factors determining the environmental impact of MOF synthesis is the choice of solvents. Typical MOF synthesis methods using conventional solvothermal synthesis often use organic solvents, such as dimethylformamide, dimethylacetamide, or other high-boiling solvents. Solvents may also be used in large amounts for washing and for solvent exchange [80].

Where the MOF chemistry allows, water and less-toxic solvents like ethanol are desirable alternatives. Use of aqueous or hydroalcoholic synthesis may decrease environmental issues associated with solvents and may even allow for easier processing downstream.

Muzaffar et al. [81] reported the efficient one-pot preparation of $CoVS_2$-MOF using bioactive chemicals of *Euphorbia cognata Boiss*. This is a cost-effective way to use organic compounds without any harmful substances. $CoVS_2$-MOF@CMS exhibited a specific capacity of 820 Cg^{-1} at 3 $mVs^{-1,}$ which is much better than the reference materials. The supercapacitor device attained an impressive Qs of 300 Cg^{-1} at 1.5 Ag^{-1} with a coulombic efficiency of 89%, specific energy of 36.68 $Whkg^{-1}$, and specific power of 2218.53 Wkg^{-1}.

5.4. Bio-Derived Ligands and Renewable Molecular Building Blocks

Another research avenue towards potentially more sustainable MOFs is the use of renewable organic ligands. Naturally occurring molecules or compounds obtained from biomass with carboxylate, hydroxyl, amino, or other coordinating groups may be used as linkers or as precursors for the preparation of linkers [82].

These can be organic acids, amino acids, polyphenolic compounds, carbohydrates, and molecules derived from lignocellulosic biomass. These materials can incorporate other functional groups and may enable the incorporation of both renewable feedstock and tunable MOF chemistry.

Bio-derived does not always mean environmentally friendly, though. Significant energy and/or chemical inputs may be required for the extraction, purification, chemical modification, or processing of a biomass-derived ligand. Hence, the sustainability of links that are made from biomass needs to be evaluated throughout their production chain [83].

5.5. Biomass-Derived Carbon and Sustainable MOF Composites

Biomass carbon is especially promising for MOF-based HSCs, as it can overcome two critical challenges: electrical conductivity and sustainability. Porous carbon frameworks with tunable surface chemistry and hierarchical structures can be obtained by converting biomass precursors like cellulose, lignin, agricultural residues, and other renewable carbon sources. MOFs can be integrated with carbon materials derived from biomass, which can generate multifunctional electrodes where carbon materials offer an interconnected electronic network and MOFs offer redox centers and structural functions [83].

Nanda et al. [84] invented an asymmetric supercapacitor (ASC) using a nickel metal–organic framework (Ni-MOF) and a nickel–MXene composite (MXAC) composed of biowaste-derived brinjal carbon. The results from morphological analysis showed that Ni-MOF has a nanosheet-like structure, and MXAC has a porous carbon skeleton. Ni-MOF showed a specific capacitance of 536 F g^{-1} at 1 A^{-1} (0–0.4 V), and MXAC showed a specific capacitance of 298 F/g at −1–0 V (in 2 M KOH). The Ni-MOF//MXAC ASC device exhibited 146 F/g at 1 A/g (1.5 V) with 2 M KOH, 45.6 Wh/kg energy density at 750 W/kg, and 85% capacitance retention after 10,000 cycles, which was excellent stability.

However, biomass-derived carbon has its inherent variability. The composition of biomass, pretreatment, activation, carbonization temperature, atmosphere, and post-processing can all have a significant effect on the surface area, pore structure, conductivity, and heteroatom content of the biomass. It is therefore a prerequisite to characterize and replicate the process of its synthesis.

5.6. Sustainability and MOF-Derived Materials

The energy-intensive thermal treatment and controlled atmosphere required to convert MOFs into metal oxides, sulfides, phosphides, or metal/carbon composites represent an important sustainability challenge since the resulting materials exhibit much higher electrical conductivity and electrochemical activity. While processes like pyrolysis can create highly conductive and porous structures, the environmental impact of the entire process, including the MOF synthesis, needs to be considered [85].

This is especially significant if the MOF is used mainly as a template and a large amount of the MOF structure is destroyed in the process of conversion. Under these circumstances, any extra material and energy needs can cancel out the functional benefits achieved.

Thus, to assess the sustainability of MOF-based electrodes, it is necessary to consider not only their electrochemical properties but also the added value of the electrode fabrication process, with respect to its energy, material, and environmental footprint. To compare the pristine MOFs, MOF composites, and MOF-derived materials for sustainable hybrid supercapacitors, this point of view is crucial.

5.7. Sustainability of Three-Dimensional Electrode Fabrication

The sustainability assessment needs to encompass MOF synthesis and the fabrication of the 3D electrode. Conductive substrates like nickel foam, metal meshes and carbon cloth offer high electrochemical performance but add to material and production costs. Similarly, complex aerogels, freeze-dried materials and 3D-printed electrodes may need several fabrication steps [86]. Ideally, the 3D structure should, therefore, have a minimum number of inactive components and a maximum number of active components. It is possible to minimize the quantity of polymeric binder and conductive additive if the growth is binder-free and direct deposition and freestanding architectures are used (Figure 11). Meanwhile, scalable fabrication processes are preferred to very elaborate laboratory processes, which are difficult to reproduce or industrialize [87].

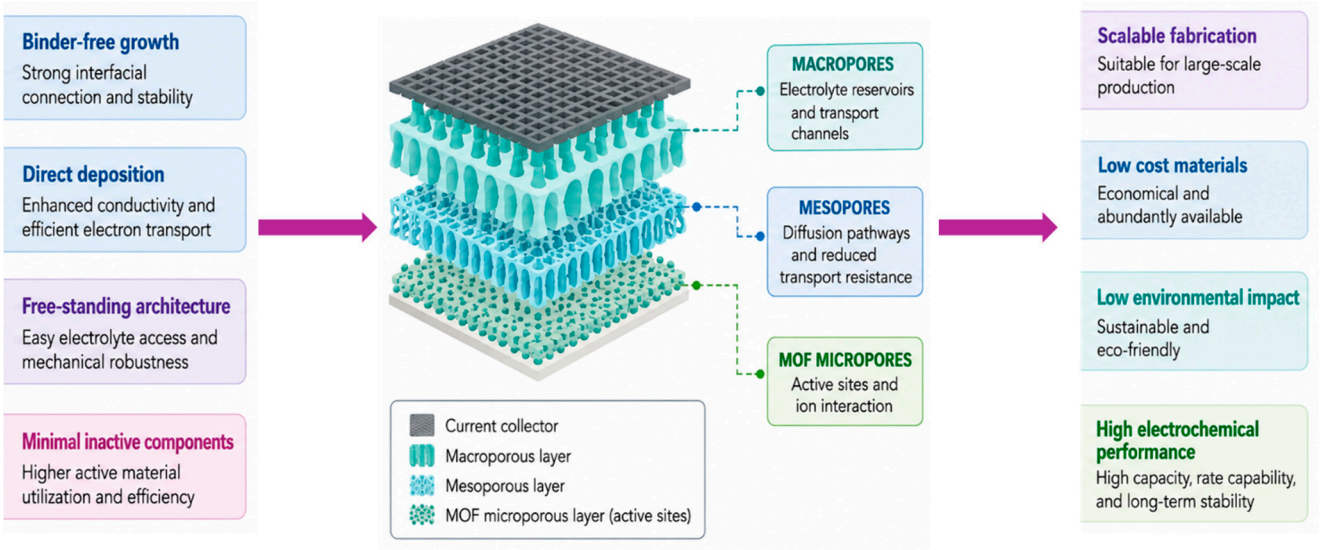


Figure 11. Design principles of 3D hierarchical electrode architecture and their benefits.

5.8. Sustainability–Performance Trade-Offs

Currently, the literature poses one major challenge because individual green features are often identified as overall sustainability. For instance, an MOF that is synthesized in water might at first look like it is environmentally friendly because water is employed instead of traditional organic solvents [88]. However, the overall environmental impact of its synthetic method could be significant if the reaction is lengthy, the yield is low, several washing and purification steps are required, or if significant amounts of solvents are used or energy-intensive drying and activation processes are employed. Likewise, the application of plenty of metal ions or less toxic metal ions does not necessarily mean a sustainable material when the extraction, purification, and incorporation into the MOF consume a lot of energy and chemical resources. In conclusion, the sustainability of an MOF should be assessed in terms of the whole material and device life cycle and not just one aspect of the MOF synthesis.

The same is true with the biomass-based electrodes. Whilst biomass is a renewable source of carbon and can help to lower the reliance on fossil-based precursors, the biomass itself does not necessarily have to be sustainable. It could not be so environmentally friendly if the biomass activation involves extensive chemical activation, corrosive or hazardous reagent usage, washing and purification steps, or high-temperature carbonization. The sustainability aspects of renewable feedstocks thus need to be considered in conjunction with the energy, water, chemical, and material needs for their transformation into a functional electrode material.

However, the existence of a non-renewable part does not imply that a material has negative overall sustainability. The material may have a longer operating life, greater energy-storage efficiency, rate capability, or structural stability than the alternative material. Where this performance improvement means that less material is required to be used or that the service life of the device produced is significantly increased, the environmental consequences of the production of the material may be offset by the reduction in material and fewer device changes. This is an example of the need to consider the link between material content, performance, durability, and consumption in the context of sustainability assessment.

The sustainability of MOF-based hybrid supercapacitors should therefore be measured based on multiple dimensions such as electrochemical performance, resource efficiency, energy consumption, toxicity, scalability, durability, and end-of-life recovery [89]. Electrochemical performance continues to be relevant since high energy density, power density, rate capability, and cycling stability are the key factors for the practical utility of the device. These parameters, however, must be considered in conjunction with the amount and importance of the resources needed to make the electrode. Resource efficiency should include the availability and origin of metal centers, organic linkers, solvent use, material yield, and the necessary amount of active material to obtain a certain performance. The energy demand should also be accounted for in the whole production process and not only during the MOF synthesis step. Thermal treatments such as drying, activation, calcination, carbonization, and others can make a significant contribution to the overall energy demand of electrodes based on MOFs. In the same way, the toxicity of solvents, metal precursors, linkers, activating agents, and other chemicals used in the synthesis and processing should be considered.

Another crucial aspect of sustainability is scalability. A small-scale synthesis process that works well may not necessarily be economical for a pilot or full-scale production. If implemented on a large scale, it could lead to the use of more solvents, more energy, more waste, or a more complicated process. Therefore, a simple, repeatable, high-yield, and potentially scalable method for the synthesis of MOFs that uses very few hazardous chemicals and does not include unnecessary processing steps should be preferred [25].

Durability is also crucial because the life of an energy storage device has a significant effect on its environmental impact. If the electrochemically active surface is very efficient and quickly loses its activity, frequent replacement is necessary, which means more raw material usage, energy consumption, and waste management. However, another electrode may offer significantly greater service life if it has an acceptable electrochemical performance but much better cycling stability. Therefore, long-term cycling stability is not just an electrochemical performance criterion but also a very significant indicator of sustainability.

Finally, there is a need to consider end-of-life recovery by evaluating MOF-based HSCs. Removal of valuable metals, carbon materials, or other functional constituents may decrease the need for virgin resources and waste production. If the design of electrodes and devices could include recovery, separation, recycling, and/or safe disposal, their overall sustainability might be enhanced. Table 1 summarizes the performance of some representative 3D MOF-based architectures in hybrid supercapacitor devices.

Table 1. Summary of the electrochemical performance of 3D MOF-based architectures designed via a green synthesis approach.

Hybrid MOFs/MOF-Derivatives	Green Strategy	Electrolyte	Capacitance (Two-Electrode) Fg^{-1}	Energy Density (Wh/kg)	Power (W/kg)	Stability (%/C.N.)	Ref.
MOF/CNT	In situ physical mixing	PVA-Li_2SO_4 gel	64.5	20.2	750	89.4/8000	[90]
HNC-900	Self-activation	1 M Na_2SO_4	26.3	9.3	800	99.9/10,000	[91]
NC-MOF@NMSe	Co-precipitation and annealing	3 M KOH	00.7 mF cm^{-2}	108.2 μWh cm^{-2}	2415 μW cm^{-2}	127/10,000	[92]
$Mo_2TiC_2T_x$@NiCo-MOF@AC	Hydrothermal	1 M KOH	278.89 Cg^{-1}	35.95	2050.35	89/8000	[93]
Cu-MOF/rGO	Ultrasonication	1 M Na_2SO4	156	14.66	2110	83.3/4000	[94]
Co/ZIF-67–rGO	Laser ablation	0.5 M Na_2SO_4	875 F g^{-1}	77.7	399.6	-	[70]
Cellulose/graphene oxide/ZIF-8-derived carbon film (CCGZ)	Room-temperature synthesis, film formation and carbonization	3 M KOH	432.8 F g^{-1} at 5 mV s^{-1}	14.92	200	87.3/10,000	[95]
La-MOF@DAC	Ultrasonication	PVA-H_2SO_4	123	14.4	640	113/5000	[96]
MOF-derived $CoSe_2$@N–C	Microwave	1 M KOH	121.3	42.2	791.6	94.4/5000	[97]
CoV-Trimesic-H3IMDC MOF	Ultrasonication	1 M KOH	187.5	70.65	835	92.1/10,000	[98]
Ln-MOF@FCNT	Solvothermal	3 M KOH	249.9	87.6	750.2	91.4/5000	[99]
$Co_{0.8}$-$Ru_{0.08}$-MOF composite	Ultrasonication	3 M KOH	54.93	17.3	750	-/-	[100]
MXene/HKUST-1	Ultrasonication	PVA/LiOH gel	19.0 Cg^{-1}	4.0	348	65/10,000	[101]
ZIF-67: Ni hybrid MOF	Room temperature	6 M KOH	45 $mhAg^{-1}$	36.97	416.3	99.8/10,000	[102]

C.N. = cycle number, -/- = Not available.

6. Sustainability–Performance Trade-Offs and Life-Cycle Considerations

Moving from a traditional approach that considers sustainability as an additional property to using it as a core ingredient of materials and device design is essential for the development of sustainable hybrid supercapacitors. This is especially relevant to MOF-based 3D architectures as electrochemical performance enhancements often in-

volve introducing other metals, linkers, solvents, conductive scaffolds, post-processing, or energy-intensive processing.

6.1. Raw Material Sustainability

The choice of appropriate metal centers and organic linkers is an important initial step towards developing sustainable MOFs. Considering the monometallic, bimetallic, and multimetallic frameworks, earth-abundant metals like Fe, Mn, Zn, Cu, Mg, and Al have the attractive properties of availability, their various coordination chemistry, and ability to form mono-, bi-, and multimetallic frameworks, which makes them attractive alternatives to relatively scarce or expensive elements [103]. But it is not enough to simply have a lot of a metal to make it a sustainable metal. Other factors to consider include the ability to be mined, refined, transported, the production of precursors, and possible toxicity.

Likewise, organic linkers can have an impact on the environment, especially if they are prepared through several steps of different chemicals and/or if they need to be purified with harmful chemicals or energy-intensive processes. Thus, selection should consider both the structural stability and the electroactivity of the frameworks that can be obtained from the selected linkers, as well as the origin of the linkers, synthesis requirements, toxicity, and feasibility of recovery and re-use/reuse [104].

This sustainable-by-design approach should therefore consider the entire material life cycle, from raw material availability to precursor toxicity, extraction and processing needs, synthesis yield, recyclability and end-of-life recovery (Figure 12). This holistic approach can help ensure that the enhancements in MOF electrochemical performance do not come at an undue environmental and resource cost.

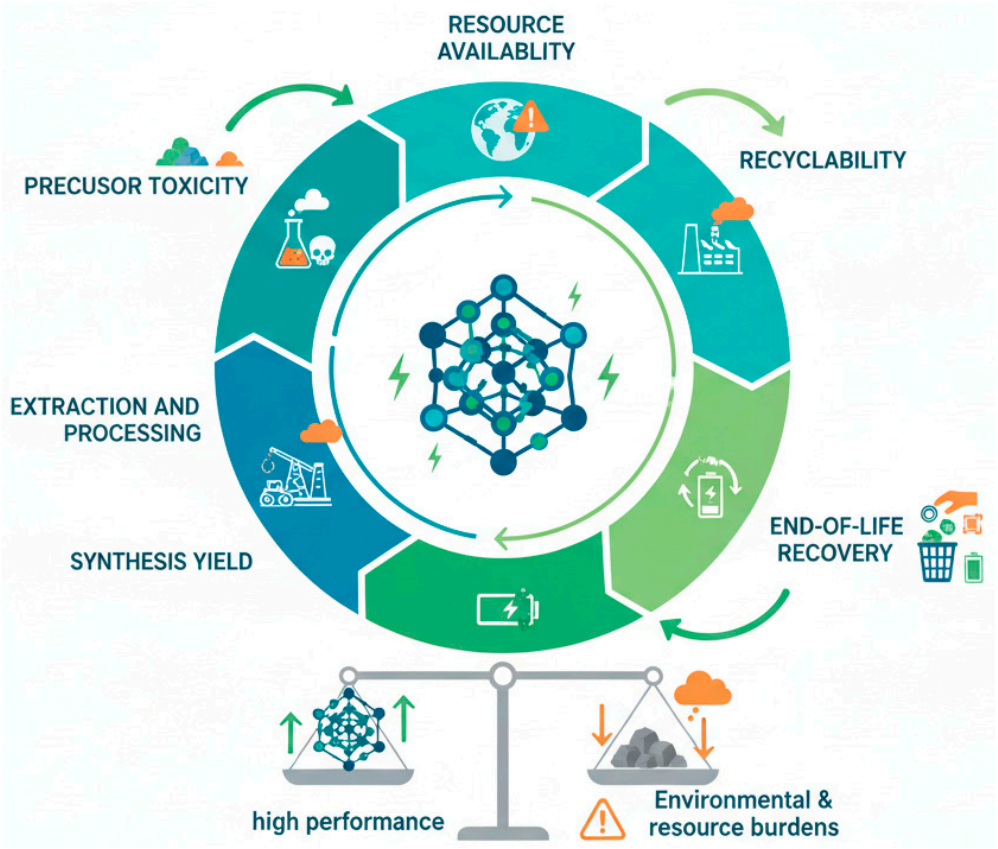


Figure 12. Factors that govern raw materials sustainability in the design of 3D electrode architecture.

6.2. Energy Consumption During Synthesis and Conversion

Another key but often overlooked aspect of MOF sustainability is energy demand [105]. Typically, conventional solvothermal synthesis involves extended heating times, and activation and conversion of MOFs to oxides, sulfides, phosphides, or composites with carbon may involve further thermal treatments. The overall energy use for producing the electrodes can be significantly higher for these processes.

Hence, the overall energy demand for 3D electrodes made using MOFs can be regarded as the sum of the energy needed for MOF synthesis, activation, converting MOF to the material, and forming the electrodes. What can also be observed is that a material that has good electrochemical properties after long-term thermal processing may have a higher environmental impact than its electrochemical performance.

Therefore, the energy efficiency of the synthesis route should be quantified by considering parameters like energy required for the preparation of a given amount of an electrode or energy required to obtain a given electrochemical performance. This can be useful to determine synthesis and processing strategies that give high electrochemical performance at low energy consumption.

6.3. Sustainability of Conductive Scaffolds

The scaffold is an integral element of a three-dimensional electrode and needs to be part of sustainability assessment. Nickel foam, for instance, has a very high electrical conductivity, mechanical strength, and structural integrity but has a significant inactive mass of the electrode. Lighter options are carbon cloth and carbon felt, but they are also energy- and resource-intensive to make and process.

Another promising alternative is carbon from biomass because renewable raw materials can be used to form conductive porous structures [106]. But this advantage to the environment is highly subject to the energy, chemicals, and processing conditions used in carbonization and activation. Therefore, the use of a renewable precursor is not necessarily associated with reduced environmental impact.

To evaluate the sustainability of the electrode system, it is important to consider the MOF, scaffold, binder, conductive additive, and current collector, not just the MOF material. This view of the whole electrode gives a realistic picture of material usage, environmental impact, and practical sustainability.

6.4. Durability and Lifetime Energy Storage

Electrochemical durability, or cycling stability, is a traditional metric for reporting stability. However, from a sustainability point of view, durability has a wider meaning: if the device lasts longer, the need to replace materials can be decreased and thus the amount of resources, energy, and waste produced during the operational life of the device [105].

One useful conceptual measure is therefore electrochemical performance as delivered over the operational lifetime, considering initial performance and the capability of the electrode to maintain its performance over many cycles. This might be a more useful measure of sustainability than the first energy density or specific capacitance [107].

While the initial performance of an electrode might be slightly worse, it could still last much longer in service than a high-performance electrode that quickly ceases to be electrochemically active. Therefore, future research should be accompanied by the long-term cycling stability of the MOF-HSC to evaluate the sustainability of the MOF-based HSC.

6.5. Scalability and Manufacturing Considerations

Synthetic methods used in the laboratory are often not readily amenable to being scaled up to a production process for the synthesis of commercial quantities. High solvent-to-

product ratios, long reaction times, and careful crystal growth may not be feasible in large-scale reactors, and small reaction volumes and extensive washing may be impractical [108].

3D architecture presents extra manufacturing challenges. Control of the pore structure and MOF deposition uniformity and control in a thick scaffold can be challenging in scaling up the process for direct growth on complex substrates.

It must therefore be recognized that the entire development process should be considered early on, from laboratory synthesis to scalable synthesis, electrode manufacturing, and device integration, instead of scale-up being considered as a last resort.

7. Challenges and Outlook

Despite significant progress in MOF-based three-dimensional (3D) hybrid supercapacitors, a substantial gap remains between laboratory-scale electrochemical performance and the requirements of practical energy-storage devices. Current studies often emphasize high specific capacitance or energy density under idealized conditions, with less attention to realistic mass loading, electrode-level performance, durability, scalability, and sustainability. Future research should therefore integrate MOF chemistry, 3D architecture, charge-transport kinetics, device performance, and environmental considerations.

1. Practical device evaluation. Future studies should employ realistic active-material loadings and report areal and volumetric capacitance, capacity, energy density, and power density alongside gravimetric metrics. Three-electrode measurements should be complemented by balanced two-electrode full-cell evaluations to provide a more realistic assessment of hybrid supercapacitor performance.
2. Transport-informed 3D electrode design. 3D MOF architecture should be designed based on mass and charge transport. Pore connectivity, pore-size distribution, tortuosity, electrode thickness, interfacial contact, and conductive-network continuity should be correlated with ion diffusion, electrolyte accessibility, electron transport, and active-site utilization. Combining electrochemical measurements with impedance spectroscopy, imaging, modeling, and operando characterization could establish these relationships quantitatively.
3. High-loading electrodes. Increasing active-material loading can introduce significant ion- and electron-transport limitations. Future architectures should maintain interconnected transport pathways at practical loadings while minimizing inactive components. Hierarchical porosity and continuous conductive frameworks offer promising strategies.
4. Long-term structural and electrochemical stability. Stability studies should extend beyond capacitance retention to investigate structural and chemical changes during cycling. Post-cycling XRD, XPS, microscopy, Raman spectroscopy, and surface-area analysis can identify phase transformations, linker degradation, metal dissolution, pore collapse, and surface reconstruction, helping establish structure–stability relationships.
5. Standardized reporting and benchmarking. Meaningful comparison requires consistent reporting of mass loading, electrode composition, binder and conductive-additive content, current collector, electrolyte, voltage window, testing configuration, current density or scan rate, electrode dimensions, and calculation methods. Both gravimetric and electrode-level metrics should be reported to minimize ambiguity.
6. Quantitative sustainability assessment. Sustainability should be incorporated into electrode development through quantitative evaluation of solvent consumption, synthesis energy, reaction time, precursor utilization, material criticality, waste generation, solvent recovery, and recyclability. Coupling life-cycle assessment with electrochemical evaluation can identify routes that balance performance and environmental impact.

7. Advanced computational and manufacturing approaches. DFT, machine learning, multiscale modeling, and additive manufacturing could support rational electrode design. These approaches can provide insights into electronic and ion–surface interactions, accelerate material and architecture discovery, bridge molecular and device scales, and enable precise control of electrode geometry, pore structure, thickness, and conductive pathways.
8. Circularity and scalable technologies. Practical development requires scalable synthesis, low-solvent or solvent-free processing, continuous manufacturing, and greater use of abundant elements. Recovery of valuable metals and carbon components, material regeneration or recycling, and safe end-of-life management should also be considered from the early stages of design.

Overall, the field should shift from isolated "high-performance MOF materials" toward sustainable, transport-optimized 3D devices. This requires understanding how MOF chemistry determines electrode architecture, how architecture controls ion and electron transport, and how transport influences active-site utilization and device performance.

8. Conclusions

MOF-based hybrid supercapacitors offer significant opportunities for advanced energy storage because of their structural tunability, high porosity, diverse metal chemistry, and multiple electrochemically active centers. However, practical implementation requires more than increasing surface area or redox activity. Integrating MOF chemistry with 3D electrode architectures can simultaneously improve electronic transport, ion diffusion, active-site accessibility, mechanical stability, and electrode-level performance.

This review highlights the importance of the composition of MOFs and 3D electrode structure as key parameters. The charge-storage properties are directly influenced by the choice of metals and linkers, defect chemistry, pore properties, and redox behavior. In the meantime, the design of the electrode affects the availability of electrolytes, transport pathways, and utilization of active material. Conductive scaffolding, hierarchical porous networks, MOF/carbon composites, and 3D printing are all strategies that can be used to improve the performance of the electrodes. These methods are successful in achieving a proper balance between porosity, conductivity, mechanical strength, and quantity of electrochemically active materials.

For practical applications of MOF-based 3D architectures, sustainability is vital, and factors like availability of abundant metals, use of renewable materials, use of environmentally friendly solvents, energy-efficient production processes, and reducing the inactive components are taken into consideration.

In conclusion, the synergistic interplay between the material structure, electrochemical performance, sustainability, and electrode configuration is comprehensively captured by combining MOF chemistry with 3D architectures. This review asserts that these factors should be viewed collectively, as an integrated strategy enhances the assessment of MOF-based hybrid supercapacitors regarding efficiency, performance, and environmental impact.

Sustainability must also become an integral component of electrode design. Earth-abundant metals, renewable components, greener solvents, energy-efficient synthesis, reduced inactive materials, scalable fabrication, and end-of-life recovery can contribute to more sustainable devices. Importantly, sustainability should be evaluated across the complete material-to-device life cycle rather than from a single favorable synthesis parameter.

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Abbreviations

The following abbreviations are used in this manuscript:

HSC	Hybrid supercapacitors
MOFs	Metal–organic frameworks
3D	Three-dimensional
EDLC	Electric double-layer capacitors
CNT	Carbon nanotubes
ASC	Asymmetric supercapacitors
rGO	Reduced graphene oxide
DFT	Density functional theory
ZDPC	ZIF-derived porous carbon

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