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2030 roadmap on porous materials for energy and environmental applications

Kezhen Qi[a,b], Lan Ding[a,*], Pitcheri Rosaiah[c,*], Zhipeng Yu[d,*], Sofia Tikhanova[e,*], Vadim Popkov[e], Ahmed Ismail[f,*], Hui Dou[g,*], Derong Luo[g], Feng Liu[g], Yixue Xu[h], Shun-Qi Xu[i,*], Chunyang Dong[j,*], Ramin Hassandoost[k], Alireza Khataee[k,l,*], Ruiyang Zhang[m,n,*], Ying Zhou[m,n], Zijun Huang[o], Yongming Luo[o], Dedong He[o,*], Yunyun Ma[p], Zhuo Xing[p,*], Claudio Imparato[q,*], Aurelio Bifulco[q]

[a] College of Pharmacy, Dali University, Dali 671000, China
[b] School of Chemistry and Chemical Engineering, Yili Normal University, Yining 835000, China
[c] Department of Physics, Saveetha School of Engineering, Saveetha Institute of Medical and Technical Sciences (SIMATS), Thandalam, Chennai 602105, India
[d] International Iberian Nanotechnology Laboratory (INL), Braga 4715-330, Portugal
[e] Hydrogen Energy Laboratory, Ioffe institute, Saint Petersburg 194021, Russia
[f] School of Physics, University of Electronic- Science and Technology of China, Chengdu 610054, China
[g] Jiangsu Key Laboratory of Electrochemical Energy Storage Technologies, College of Materials Science and Technology, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China
[h] Research Center for Nano Photoelectrochemistry and Devices, School of Chemistry and Chemical Engineering, Southeast University, Nanjing 211189, China
[i] School of Energy and Environment, Southeast University, Nanjing 211189, China
[j] China-UK Low Carbon College, Shanghai Jiao Tong University, Shanghai 201306, China
[k] Research Laboratory of Advanced Water and Wastewater Treatment Processes, Department of Applied Chemistry, Faculty of Chemistry, University of Tabriz, Tabriz 51666-16471, Iran
[l] Department of Chemical Engineering, Istanbul Technical University, Istanbul 34469, Türkiye
[m] National Key Laboratory of Oil and Gas Reservoir Geology and Exploitation, Southwest Petroleum University, Chengdu 610500, China
[n] School of New Energy and Materials, Southwest Petroleum University, Chengdu 610500, China
[o] Faculty of Environmental Science and Engineering, Kunming University of Science and Technology, Kunming 650500, China
[p] Institute of Nanoscience and Nanotechnology, College of Physical Science and Technology, Central China Normal University, Wuhan 430079, China
[q] Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Naples 80125, Italy

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ABSTRACT

Porous materials, including metal-organic frameworks (MOFs), covalent organic frameworks (COFs), aerogels, and porous metal oxides, have been extensively explored as versatile platforms for energy conversion, storage, and environmental applications. Over the past five years, remarkable advances have been achieved in the design, synthesis, and functional optimization of these materials, opening new opportunities for practical implementation. In this roadmap, we focus on several key subtopics, including MOFs and COFs for supercapacitors and batteries, electrocatalysis and photocatalysis, heterojunction materials for charge separation, advanced electrocatalysts and photocatalysts based on aerogels, carbon aerogels for environmental remediation, and porous metal oxide nanomaterials for electrocatalysis. The current status, challenges, and opportunities in these areas are systematically summarized. Special attention is given to mechanistic insights, stability enhancement, conductivity improvement, and scalable fabrication strategies that are essential for bridging fundamental research and real-world applications. We believe this roadmap will provide valuable suggestions and updated knowledge for researchers, and offer useful inspiration to accelerate the development of porous materials for sustainable energy and environmental technologies toward 2030.

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* Corresponding authors.
E-mail addresses: dl912116@dali.edu.cn (L. Ding), dr.mosesrosaiah@gmail.com (P. Rosaiah), Zhipeng.yu@inl.int (Z. Yu), s.m.tihanova@mail.ioffe.ru (S. Tikhanova), ahmed.ics13@gmail.com (A. Ismail), dh_msc@nuaa.edu.cn (H. Dou), shunqi.xu@seu.edu.cn (S.-Q. Xu), chunyang_dong@sjtu.edu.cn (C. Dong), a_khataee@tabrizu.ac.ir (A. Khataee), ryzhang@swpu.edu.cn (R. Zhang), dedong.he@qq.com (D. He), xingzhuo@ccnu.edu.cn (Z. Xing), claudio.imparato@unina.it (C. Imparato).

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1. MOFs in supercapacitors and batteries

Rosaiah Pitcheri*

1.1. Status

Metal-organic frameworks (MOFs) have emerged as a unique class of crystalline porous materials constructed from metal nodes or clusters interconnected by organic linkers. Their modular architecture enables a wide variety of architectures, making them adaptable for different applications. In recent years, MOFs have shown significant attention within EEEEs, including supercapacitors (SCs) and rechargeable batteries (such as LIBs, SIBs, and ZIBs). MOFs exhibit exceptionally high surface areas (often exceeding 1000 m^2/g), highly porous nature, and chemically flexible frameworks, which allow for abundant exposure of electroactive sites and efficient mass transport [1–3]. These structural benefits extend opportunities to tailor charge storage mechanisms at both the surface and bulk levels. In SCs, MOFs can function either as active electrode materials or as precursors to porous carbons and metal oxides with high surface-to-volume ratios, enabling significant contributions from both electric double-layer capacitance (EDLC) and pseudocapacitance [4,5]. Their tunable pore structures further promote fast electrolyte ion diffusion for achieving high power densities and rapid charge-discharge cycles.

In rechargeable batteries, MOFs play vital roles depending on their structure and transformation. Pristine MOFs can act as host matrices that provide accessible channels for reversible ion insertion/extraction and mitigate structural collapse during cycling. More commonly, MOFs are converted into MOF-derived functional materials, such as porous carbons, metal oxides, sulfides, and phosphides, which exhibit enhanced electrical conductivity, high specific capacity, and robust structural stability compared to their parent frameworks. Such derivatives can serve as advanced anodes, cathodes, or conductive scaffolds in LIBs, SIBs, and ZIBs, offering improved rate performance and extended cycling life. Furthermore, pristine MOFs have several limitations for direct application in practical electrochemical devices. Their poor intrinsic conductivity restricts fast electron transport, while their relatively fragile crystalline frameworks may undergo collapse or degradation under harsh electrochemical cycling conditions. Moreover, large-scale synthesis and long-term operational stability remain challenging. As a result, significant research efforts are directed toward rational structural engineering, composite formation with conductive materials (*e.g.*, graphene, MXenes, CNTs), and controlled pyrolysis to yield robust MOF-derived architectures [6]. These strategies have propelled MOFs and their derivatives to the forefront of next-generation energy storage research, bridging the gap between traditional electrode materials and the performance demands of modern ESSs.

1.2. Current and future challenges

Despite the tremendous potential of MOFs and their derivatives for energy storage applications, several critical bottlenecks hinder their practical deployment in commercial supercapacitors and batteries. These limitations include poor intrinsic electrical conductivity, structural instability under prolonged cycling, challenges in scalable and cost-effective synthesis, and ensuring long-term device stability. Overcoming these obstacles is essential to translate the promising laboratory-scale performance of MOF-based materials into reliable, high-performance, and economically viable commercial energy storage devices.

Low electrical conductivity: The majority of pristine MOFs are composed of insulating organic linkers and isolated metal nodes, which leads to inherently poor electron transport. This low conductivity limits their ability to support fast redox reactions, resulting in sluggish charge storage kinetics and reduced rate capability. Overcoming this challenge requires strategies such as incorporating conductive linkers, forming composites with carbonaceous materials (graphene, CNTs, MXenes), or converting MOFs into conductive derivatives (*e.g.*, carbonized MOFs or metal sulfides).

Structural instability: While the crystalline frameworks of MOFs are attractive for providing ordered pores and high surface areas, they are often mechanically fragile. During repeated charge–discharge cycles, volume expansion, ion insertion/extraction, or structural stress can cause partial collapse or amorphization of the framework. This leads to rapid capacity fading and limits long-term cycling stability. Designing flexible frameworks, introducing hierarchical porosity, or creating MOF-derived architectures with robust backbones are emerging solutions to mitigate these issues.

Limited energy density: A persistent trade-off exists between energy and power density in energy storage systems. Supercapacitors, though capable of delivering very high power, generally suffer from low energy densities. Batteries, on the other hand, offer high energy density but at the cost of slower kinetics. MOFs must bridge this gap by simultaneously enabling fast ion diffusion and providing sufficient redox-active sites. However, tailoring MOF structures to achieve this balance remains a challenge, especially under practical device-level conditions.

Scalability and cost-effectiveness: While laboratory-scale synthesis of MOFs with controlled morphology, porosity, and functionality has been demonstrated, translating these methods into large-scale, cost-effective production is still difficult. Many MOFs require expensive organic linkers, toxic solvents, or long reaction times, which are impractical for industrial-scale deployment. Furthermore, achieving uniform particle size and phase purity at scale remains a hurdle. Developing green, solvent-free, and continuous synthesis methods is therefore a key research direction.

Compatibility with electrolytes: Electrolyte stability is another critical factor influencing MOF performance. Many MOFs undergo degradation, ligand detachment, or framework collapse when exposed to aqueous or highly acidic/alkaline electrolytes, limiting their practical usability. In addition, mismatched pore sizes may restrict electrolyte ion access, further reducing electrochemical efficiency. Surface functionalization, post-synthetic modification, or encapsulation strategies are being investigated to improve chemical stability and broaden electrolyte compatibility.

In summary, MOFs hold great promise for developing high-performance electrodes because of their flexible structures and rich chemistry. But, their practical use is limited by several drawbacks such as poor conductivity and low stability. Therefore, development of special structural designs and smart functional composite materials is necessary to realize their potential in real-world energy applications.

1.3. Advances in science and technology to meet challenges

Recent advances in materials science have provided versatile strategies to overcome the intrinsic limitations of MOFs, including low conductivity, structural fragility, and electrolyte sensitivity, in supercapacitor and battery applications. The development of MOF-based electrodes can be categorized into three hierarchical levels:

(1) Architecture-level optimization, including hierarchical porosity, hollow and 2D morphologies, and strain engineering to improve ion diffusion and mechanical stability.
(2) Framework-level modification, featuring conductive linkers, redox-active nodes, and defect engineering to enhance intrinsic charge transport.

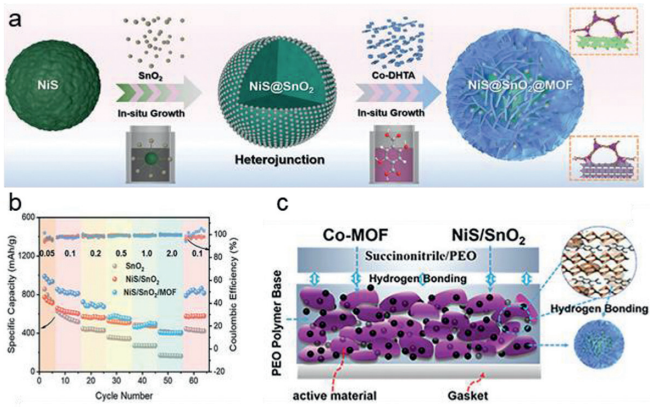


Fig. 1. (a) Schematic illustration of the synthetic route of NSM composite. (b) Rate performance of SnO_2, NiS/SnO_2, and NiS/SnO_2/MOF electrodes. (c) The schematic diagram of the interior structure in flexible electrode. Reproduced with permission [8]. Copyright 2023, Wiley.

(3) Hybrid or derived materials, such as MOF-carbon/MXene composites and MOF-derived sulphides or phosphides, which integrate high conductivity with robust frameworks.

These multiscale strategies collectively enhance charge transport, structural robustness, and active-site accessibility, marking a shift from macro- and microstructural design to molecular-level engineering and hybrid integration for high-performance energy-storage systems.

1.3.1. Architecture-level engineering

Architecture-level modifications bridge the gap between molecular framework design and macroscopic device performance. Hierarchical porous MOFs combine micropores for charge storage with meso–/macropores to facilitate rapid ion transport, enabling high capacitance and excellent rate performance. Hollow, tubular, and nanosheet architectures alleviate diffusion limitations, buffer volume expansion during cycling, and increase accessible surface area. For example, Co-based hierarchical porous MOFs exhibit enhanced capacitance and long-term cycling stability due to efficient ion diffusion pathways [7]. Strain engineering and alignment of MOF nanorods into ordered 3D arrays further improve lattice flexibility, mechanical robustness, and electrical conductivity, ensuring durable performance under realistic electrochemical conditions.

Building on these strategies, Zhang *et al.* [8] proposed a multi-templated synthesis to construct a Co-MOF-based heterostructure (NiS/SnO_2/MOF) composite (Fig. 1a). The composite electrode combines the porous MOF framework with the high-capacity NiS/SnO_2 heterostructure, yielding superior specific capacity and cycling efficiency (Fig. 1b). This architecture mitigates interfacial stress and volume changes during Li^+ insertion/extraction, while improving compatibility with poly(ethylene oxide) (PEO)-based solid electrolytes. The flexible NSM-PEO electrode maintains a stable voltage of 3.5 V under repeated bending, confirming excellent interfacial stability and low leakage risk (Fig. 1c). Such hierarchical design and mechanical optimization demonstrate the potential of architecture-level engineering for next-generation flexible energy-storage devices.

1.3.2. Framework-level innovations

At the molecular scale, the intrinsic conductivity and stability of pristine MOFs are often insufficient for electrochemical applications. To overcome this limitation, researchers have introduced π-conjugated linkers such as 2,3,6,7,10,11-hexaiminotriphenylene (HITP), which enable extended electron delocalization. For example, $Ni_3(HITP)_2$ has demonstrated metallic-like conductivity and excellent charge storage properties in supercapacitors. Incorporating redox-active metal nodes (Co^{2+}, Ni^{2+}, Fe^{3+}) further enhances faradaic activity, leading to higher capacitance and stability. Defect engineering offers another way to create additional active sites and improve ion accessibility. UiO-type MOFs with systematically tuned defect densities, have shown superior capacitance and enhanced ion diffusion. In addition, heteroatom-functionalized linkers containing N, S, or Patoms have been shown to modify the electronic environment, accelerate charge transfer, and maintain structural integrity. Collectively, these molecular-level design strategies transform MOFs from passive frameworks into intrinsically active electrode materials [9].

A representative example is the *quasi*-honeycomb conductive 2D c-MOF Cu-OHDDQP, which integrates dual-pore architecture and redox-active organic linkers to achieve high conductivity and capacitive performance [10]. In aqueous lithium electrolytes, Cu-OHDDQP delivers a record-high specific capacitance of 452 F/g while maintaining 90% capacity over 1000 cycles. Density functional theory (DFT) calculations (Fig. 2a) indicate that SO_4^{2-} and Li^+ ions act as active adsorbates, with pyridyl-N and bridging O atoms serving as Li^+ adsorption sites (−1.43 eV and −2.03 eV, respectively) and Cu nodes adsorbing SO_4^{2-} during charging (−1.55 eV). A four-electron pseudocapacitive mechanism (Fig. 2b) has been proposed, in which Cu(II) centers are sequentially reduced to Cu(I) and then oxidized back during cycling, accompanied by reversible Li^+ adsorption/desorption on the oxygen and nitrogen coordination sites. This multi-electron process yields a theoretical capacitance of 529 F/g, close to the experimental value, confirming the efficiency of the redox-active framework. The high performance is attributed to the quasi-honeycomb lattice providing enlarged surface area and the pyridyl groups offering additional Li^+ adsorption sites, demonstrating the synergy of ligand design and framework-level modulation.

1.3.3. Hybrid and derived materials strategies

Incorporating conductive materials or transforming MOFs into derivatives is highly effective for achieving practical device performance. MOF–carbon composites exploit the conductivity of carbon with the porosity of MOFs, yielding high capacitance and cycling stability, while MOF–MXene composites combine surface functionality and metallic conductivity, enhancing durability in flexible supercapacitors [11]. Thermal or chemical conversion produces functional derivatives, such as ZIF-8-derived N-doped carbons, which show high energy density and stable cycling. A notable example is the amorphous MOF (ZGB-MOF) developed as a solid-state electrolyte matrix. Constructed *via* Zn^{2+}/Ga^{3+} competitive coordination, ZGB-MOF contains O-Ga-Cl polyanionic clusters, nanopores, and abundant C=O/oxygen-vacancy sites [12]. These structural features facilitate $ZnCl^+$ carrier formation (Fig. 2c) and reduce the ion transport barrier to 0.12 eV. The derived ZGBC electrolyte exhibits high ionic conductivity (5.2×10^{-3} S/cm) and large interfacial ion diffusion ($D = 3.8 \times 10^{-11}$ – 9.5×10^{-10} cm^2/s) (Fig. 2d), along with a wide electrochemical window (up to 2.88 V) and high charging voltage (2.4 V). Full cells using a Zn-ferricyanide cathode retain stable cycling over 5000 cycles, demonstrating dendrite-free Zn deposition and excellent interface stability (Figs. 2e-g). This work highlights how amorphous MOF-derived matrices bridge electrode and electrolyte design, enabling flexible, dendrite-free zinc-based batteries.

At the device level, binder-free films and flexible current collectors have been achieved using MOF-derived nanofiber mats fabricated by electrospinning, which combine mechanical robustness with scalable processing routes [13]. These hybrid and derivative approaches not only enhance conductivity and stability but also bring MOF-based systems closer to practical application in next-generation electrochemical energy storage.

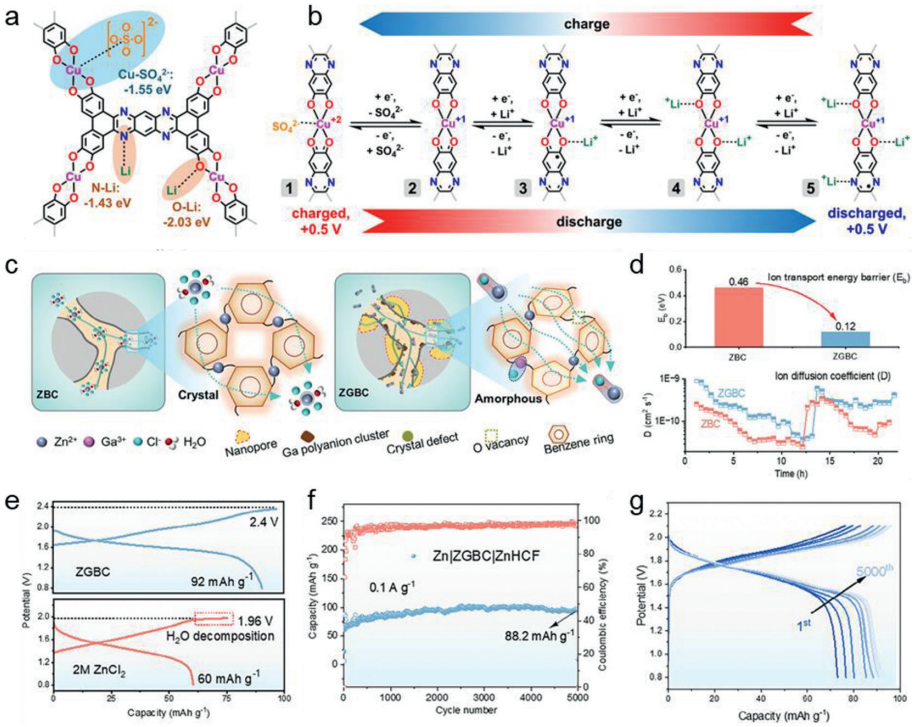


Fig. 2. (a) Possible active sites for the adsorption of electrolyte cations (Li^+) and anions (SO_4^{2-}) in a single repeating unit of Cu-OHDDQP. (b) Proposed energy storage mechanism for the Cu-OHDDQP electrode in aqueous Li_2SO_4 electrolyte. Reproduced with permission [10]. Copyright 2025, Wiley. (c) Transport schematic of Zn-ion groups in ZBC and ZGBC electrolytes. (d) Ion transport energy barrier and ion diffusion coefficient of ZBC and ZGBC electrolytes. (e) The charge and discharge curves with different charging voltage. (f, g) The long-term cycling performance and charge and discharge curves of Zn|ZGBC|ZnHCF full cell, running at 0.1 A/g and room temperature. Reproduced with permission [12]. Copyright 2025, Wiley.

1.4. Concluding remarks and prospects

MOFs and their derivatives have emerged as multifunctional and highly tunable platforms for advancing electrochemical energy storage systems. The ordered porosity, structural diversity, and chemical modularity of MOFs enable them to serve as active electrode materials, conductive hosts, and precursors for complex nanostructures. Over the past decade, extensive research has demonstrated their potential to significantly enhance the performance of both supercapacitors (high power, rapid charging) and rechargeable batteries (high energy, long-term storage). Despite these advances, substantial challenges remain before MOF-based systems can transition from laboratory studies to widespread commercial applications.

Several directions are expected to shape the future research landscape. One promising strategy involves the design of intrinsically conductive MOFs by incorporation of π-conjugated ligands, redox-active linkers, and mixed-valence metal nodes to impart both electrical conductivity and robust electrochemical activity, thereby minimizing the need for post-pyrolysis treatments or external carbon additives. Equally important is the development of scalable and sustainable synthesis strategies, such as mechanochemical and solvent-free methods or the 3D printing of MOF-based inks, to enable cost-effective and continuous production with high crystallinity, uniform particle size, and phase purity. Another key direction lies in the creation of hybrid and tandem architectures, for example, MOF-MXene, MOF-graphene, MOF-polymer, or MOF-single-atom catalyst composites, which can synergistically integrate conductivity, redox activity, and mechanical stability to achieve high energy and power densities with long cycling lifetimes.

Finally, stronger emphasis on device-level validation—progressing beyond half-cell testing toward full-cell prototypes, flexible and wearable devices, and pouch-scale demonstrations under realistic conditions such as high areal loading, thick electrodes, and safe electrolytes, will be indispensable for commercialization. In summary, MOFs represent a transformative materials platform at the intersection of chemistry, nanotechnology, and electrochemistry. With sustained advances in structural engineering, sustainable manufacturing, and mechanistic understanding, MOF-based electrodes are poised to play a vital role in the development of next-generation supercapacitors and rechargeable batteries, bridging the gap between laboratory-scale innovations and practical energy storage technologies.

2. MOFs in electrocatalytic applications

Zhipeng Yu*

2.1. Status

MOFs have emerged as highly versatile platforms for electrocatalysis owing to their structural regularity, permanent porosity, and tunable metal-ligand chemistry [14]. Unlike conventional porous catalysts, MOFs provide atomically well-defined coordination environments that can be precisely tailored to create active sites for key electrochemical reactions, including the oxygen evolution/reduction reactions (OER/ORR), hydrogen evolution reaction (HER), and carbon dioxide reduction reaction (CO_2RR) [15]. Two complementary application modes have been widely explored: (1) Pristine MOFs as electrocatalysts, where the metal nodes and organic linkers act as catalytic centers [16], and (2) MOFs as precursors or templates, in which high-temperature or electrochemical transformations yield conductive carbons, atomical dispersion catalysts, or metal-based nanoparticles embedded in porous matrices [17]. Recent advances in conductive and conjugated MOFs,

such as $Co_3(HADQ)_2$ (HADQ = hexaamine dipyrazino quinoxaline), $Cu_3(HHTP)_2$ (HHTP = hexahydroxytriphenylene), and other 2D π-d frameworks, have demonstrated superior intrinsic conductivity and redox activity, making them competitive with traditional carbon- or metal-based catalysts [18]. Meanwhile, MOF-derived nanostructures (*e.g.*, metal-nitrogen-carbon, M-N-C) have achieved state-of-the-art activity and durability in fuel cell and electrolyzer applications.

Importantly, the structural features that enable high electrocatalytic performance, including tunable metal nodes, conjugated linkers, and permanent porosity, also provide fast ion transport pathways and abundant redox-active sites for energy storage. Consequently, many conductive MOFs and MOF-derived materials are multifunctional and capable of serving both as electrocatalysts and as electrodes in supercapacitors or batteries [19].

2.2. Current and future challenges

Although MOFs have shown remarkable promise in electrocatalysis, their path to large-scale deployment remains constrained by several interrelated issues. The foremost challenge is electronic conductivity: Most pristine MOFs are insulating due to localized frontier orbitals, which hinders charge transport and limits catalytic turnover [16]. Recent progress in conductive frameworks such as $Cu_3(HHTP)_2$ has alleviated this limitation, but their conductivity is still far below that of benchmark 2D materials like graphene [16,18].

Equally critical is structural and chemical stability. Under practical reaction conditions, strong alkaline or acidic electrolytes, high current densities, and long-term cycling, many MOFs undergo node reconstruction, linker decomposition, or metal leaching. Such instability not only degrades activity but also complicates mechanistic interpretation. Furthermore, scalable processing and device integration remain underdeveloped: Current approaches (powder catalysts, slurry-coated electrodes) obscure active sites and rely on binders, reducing intrinsic advantages. Achieving wafer-scale thin films, freestanding membranes, or binder-free electrodes is essential but technically demanding [20].

Finally, mechanistic understanding lags behind. Although MOFs provide atomically defined sites, their dynamic transformations during operation are poorly captured, limiting predictive design. For multifunctional applications, balancing electrocatalytic activity with energy storage performance presents a challenge. Optimization strategies must consider both electron/ion transport for catalytic turnover and reversible redox activity for charge storage, highlighting the need for integrated design approaches that unify electrocatalysis and energy storage functionalities.

2.3. Advances in science and technology to meet challenges

Progress toward overcoming these limitations is emerging along four interconnected directions:

(1) Rational design of conductive frameworks. By incorporating extended π-conjugated linkers (HHTP, HADQ) and electron-rich metal centers (Ni^{2+}, Cu^{2+}, Co^{2+}), researchers have developed conductive MOFs with band-like charge transport. $Co_3(HADQ)_2$, for example, exhibits ORR activity rivaling Pt (Figs. 3a and b) [21], while Cu–HHTP-based frameworks have demonstrated satisfactory HER catalytic activity, with an overpotential of 112 mV at a current density of 10 mA/cm^2 (Figs. 3c and d) [22]. Critically, these same design principles also enhance ion adsorption and reversible redox processes, enabling these MOFs to serve simultaneously as high-performance electrodes for supercapacitors and batteries [18].

(2) MOF-derived and hybrid catalysts. Pyrolysis and controlled transformation convert MOFs into porous carbons, M-N-C atomical dispersion catalysts, and heterostructured composites [23,24]. These derivatives retain the spatial dispersion of active sites while gaining conductivity and robustness. MOF-derived Co-N-C materials have shown state-of-the-art ORR activity (Figs. 3e and f) [25], while hybridization with MXenes or graphene improves interfacial electron transfer (Figs. 3g and h) [26]. Such hybrid structures are also highly effective for energy storage, providing interconnected conductive networks and abundant electroactive sites for rapid ion diffusion and storage. By 2030, the convergence of MOFs with other 2D conductive platforms is expected to yield hybrid catalysts with synergistic advantages in both stability and activity.

(3) Advanced fabrication and integration. Emerging strategies such as interfacial growth, vapor-assisted deposition, and direct electrochemical assembly now allow the formation of thin, crystalline MOF films or freestanding electrodes (Fig. 3i) [27]. These binder-free architectures maximize utilization of active sites and ensure seamless contact with current collectors. Scalable roll-to-roll deposition and 3D printing are envisioned as transitional steps toward industrial adoption, enabling MOFs to be deployed in practical electrolyzers, fuel cells, and energy storage devices such as supercapacitors or battery electrodes [20].

(4) *Operando* characterization and theory-guided design. *In situ* X-ray absorption, Raman spectroscopy, and electrochemical microscopy have begun to reveal how MOF active sites evolve under working potentials, capturing dynamic metal-ligand reconstruction and local environment changes [28]. Coupled with density functional theory and machine learning, these insights guide the rational design of multifunctional MOFs capable of balancing electrocatalytic efficiency with energy storage capacity, highlighting structure–function relationships that unify the two applications.

Collectively, these advances set the stage for MOFs to transition from proof-of-concept systems to multifunctional materials that integrate electrocatalysis and energy storage. By 2030, enhanced conductivity, stability engineering, scalable fabrication, and mechanistic clarity are expected to enable MOFs as cornerstone materials for integrated energy technologies.

2.4. Concluding remarks and prospects

Looking toward 2030, MOFs are expected to evolve from model catalysts to integral components of advanced energy and environmental systems. Rational design of conductive MOFs and robust MOF-derived catalysts can simultaneously enhance electrocatalytic activity and energy storage performance. Future research should focus on:

(1) Multifunctional material design: Heteroatom doping, defect engineering, and hybridization with conductive matrices can be strategically applied to simultaneously enhance electrocatalytic activity and energy storage performance. Such approaches exploit the inherent porosity and tunable metal-ligand chemistry of MOFs to maximize active site accessibility and charge transport efficiency.

(2) Scalable fabrication and integration: Developing wafer-scale thin films, freestanding electrodes, and 3D-printed architectures will facilitate the translation of MOFs from lab-scale proof-of-concept to practical devices. Binder-free and interfacially optimized electrodes can further enhance utilization of active sites and device stability.

(3) Mechanism-guided discovery: Combining *operando* characterization, high-throughput computation, and machine learning enables predictive design of MOFs with optimal conductivity, stability, and catalytic efficiency. Integrating experimental and computational insights can accelerate the identification

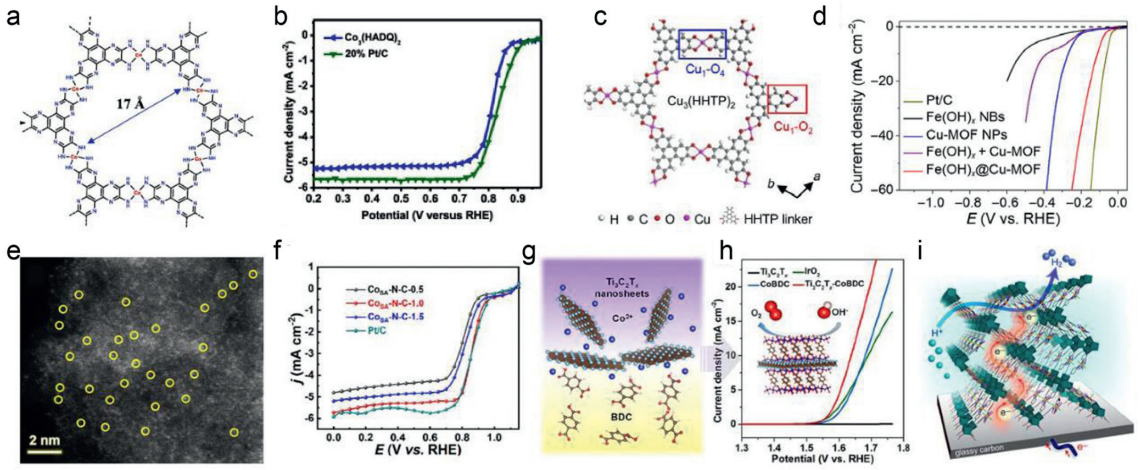


Fig. 3. (a) Crystal structure and (b) electrocatalytic ORR activity of $Co_3(HADQ)_2$. Reproduced with permission [21]. Copyright 2022, Elsevier. (c) Crystal structure and (d) electrocatalytic HER activity of Cu–HHTP. Reproduced with permission [22]. Copyright 2021, AAAS. (e) Atomic-resolution electron microscope images and (f) electrocatalytic ORR activity of Co-N-C catalyst. Reproduced with permission [25]. Copyright 2025, Elsevier. (g) Schematic illustration of the MXenes/Co MOF hybrid and (h) its electrocatalytic OER activity. Reproduced with permission [26]. Copyright 2017, American Chemical Society. (i) Schematic illustration of the MOF thin film for HER. Reproduced with permission [27]. Copyright 2019, American Chemical Society.

of MOFs suitable for both electrocatalysis and energy storage, while revealing structure-function relationships.

(4) Exploration of multifunctional applications: Beyond traditional electrocatalysis and batteries, MOFs can be tailored for hybrid applications such as CO_2 reduction coupled with energy storage, solar-to-chemical conversion, or environmental remediation, leveraging their structural precision, tunability, and multifunctionality.

Collectively, these strategies provide a concrete roadmap for next-generation MOFs, ensuring that by 2030, they can serve as cornerstone materials for integrated energy and environmental technologies, encompassing sustainable catalytic transformations and efficient electrochemical energy storage.

3. MOFs for photocatalysis

Sofia Tikhanov*, Ahmed Ismail*, Vadim Popkov

3.1. Status

MOF photocatalysts are highly promising platforms for light-powered catalysis due to the core design tunability of MOFs [29,30]. Tailorable porosity and large surface area allow for numerous adsorption and reaction sites, and the richness of metal nodes and organic linkers permit site-selective tuning of the coordination sites and active sites [31–34]. These attributes set MOFs apart from the usual semiconductors and allow for the co-integration of functions of catalysis, adsorption, and separation as one framework.

However, pure MOFs bear narrow optical absorption bands, poor charge-carrier mobility, as well as structural instability upon operating conditions, and as a whole, leave much to be desired in photocatalytic activity. To compensate for these deficiencies, MOF-based composites have been synthesized as an integration of MOF structural merits with photophysical characteristics of semiconductors. Visible-light-responsive heterostructures have been achieved by integrating MOFs with TiO_2, Fe_2O_3, Fe_3O_4, $BiVO_4$, g-C_3N_4, Ti_3C_2, and CdS [35–38]. These semiconductors widen the light-response band of MOFs and promote interfacial charge transfer, and the porous MOF framework supports dispersion and stability enhancement.

The rational integration of MOF composites facilitates the harvesting of a wider range of solar irradiation, diminishes the recombination of photogenerated electron–hole pairs, and reveals further catalytically active sites [39]. This synergistic design approach has emerged as a pivotal element in enhancing MOF photocatalysis for efficient and sustainable applications in and energy sectors.

Beyond energy conversion, MOFs have recently demonstrated strong potential in environmental remediation. Their hierarchical porosity facilitates pollutant adsorption, while photoactive metal nodes and linkers drive degradation under light irradiation. Visible-light-responsive MOFs such as NH_2−MIL-125(Ti), UiO-66-NH_2, and MIL-100(Fe) effectively remove dyes, antibiotics, and heavy-metal ions *via* integrated adsorption-photodegradation processes [40–42]. This coupling of molecular capture and photocatalytic degradation offers a sustainable strategy for water purification and air detoxification.

3.2. Current and future challenges

In recent years, extensive efforts have been devoted to exploring MOFs and their derivatives for photocatalytic and related reactions, with abundant experimental evidence highlighting their promise [43–46]. Owing to their crystalline porous nature, composed of metal nodes/clusters and organic linkers, MOFs feature low framework density, high surface area, tunable pore structures, and abundant active sites, making them attractive for photocatalytic processes such as CO_2 reduction, H_2O_2 synthesis, hydrogen evolution, and organic conversion.

Although considerable progress has been achieved, MOF-based photocatalysts still face intrinsic limitations in light absorption, charge transport, and long-term durability. The first large-scale challenge arises from the restricted light absorption across the visible spectrum, which directly lowers the efficiency of solar-to-chemical energy conversion. Most pristine MOFs exhibit wide bandgaps, low active-site density, and rapid electron-hole recombination. To counteract these drawbacks, MOF-semiconductor heterostructures have been extensively explored to enhance visible-light harvesting and interfacial charge transfer [47,48]. However, these systems typically require multi-step synthesis and complex post-treatments that impede scalability and cost-effectiveness.

A second limitation concerns the reliance on sacrificial reagents in photocatalytic half-reactions such as water splitting or pollutant degradation. Although these reagents facilitate charge separation by trapping holes or electrons, they compromise atom economy

and sustainability. The use of cocatalysts provides a more practical alternative by accelerating redox kinetics and stabilizing charge carriers. Yet, the widespread dependence on noble-metal cocatalysts remains a barrier to large-scale deployment, emphasizing the urgent need for low-cost, earth-abundant substitutes such as transition-metal phosphides, sulfides, and carbides.

In the field of environmental remediation, achieving selective and complete degradation of pollutants with minimal by-product formation remains a formidable challenge. The photocatalytic efficiency of MOFs is largely governed by pore architecture and mass diffusion. While host–guest synergistic stabilization of active sites within MOF channels can enhance reaction coverage and selectivity, intrinsic microporosity often restricts reactant diffusion and slows catalytic turnover. Rational design of hierarchically porous structures or defect-engineered frameworks could alleviate these diffusion bottlenecks and improve reaction kinetics.

Furthermore, metal-ion leaching and framework instability during photocatalysis raise concerns about secondary contamination and environmental safety. The toxicity and biocompatibility of MOFs depend strongly on composition, particle size, surface chemistry, and degradation behavior. Although certain frameworks such as FeBDC have demonstrated low cytotoxicity and stable photocatalytic activity [49], the long-term ecological impact and safety of degradation products require comprehensive evaluation before practical application in water treatment.

Beyond the material-level issues, scale-up from laboratory research to industrial application is hampered also by cost, energy efficiency, and environmental concerns. All of these need to be overcome if the potential of MOF-based photocatalysis for sustainable energy generation and clean water is to become a reality.

3.3. Advances in science and technology to meet challenges

In light of the aforementioned deficiencies, researchers have attempted numerous means such as doping, heterojunction construction, and morphology adjustment to improve MOF-based photocatalysts' performance. Among those, the construction of bimetallic MOFs is the most successful. Due to the incorporation of two kinds of metal ions into one framework, bimetallic MOFs are generally more catalytically active and structurally stable than monometallic MOFs are. A typical case is the interface modification between MoS_2 and a bimetallic MOF represented by Masekela *et al.*'s work, whereby the performance of both the production of hydrogen and the removal of pollutants was significantly optimized [50].

Another promising research field is related to the tuning of carrier dynamics through conductive additives. In La-MOF systems, the inherently poor charge-transfer capacity was dramatically upgraded with the introduction of two-dimensional MXene. MXene's electron-withdrawal nature adjusted the configuration of the valence band, facilitating hole transfer and rapid carrier separation. Besides expanding the photocatalytic usages of MOFs, it also shows a case of applying valence-band engineering to overcome intrinsic transport barriers [51]. Wang *et al.* [52] successfully encapsulated C_{60} and C_{70} within ZIF-8 forming a unique solid solution structure, that enhances host-guest connection and promotes charge separation through electron density movement from ZIF-8 to the fullerenes (Fig. 4a). Under visible light irradiation, C_{60}@ZIF-8 and C70@ZIF-8 displayed excellent photocatalytic performance with 6.01 and 24.95 mmol g^{-1} h^{-1} H_2 production, which is 319.4 and 59.2 times higher than pure fullerenes. This was due to extended light absorption, improved charge separation and strong built-in electric field in C_{60}@ZIF-8 and C70@ZIF-8.

In order to decrease dependence on sacrificial agents, piezo-photocatalysis as an emergent alternative method has been developed. In this system, the integration of step-scheme heterojunctions allows for the development of a dual internal electric field, promoting noteworthy enhancement of photogenerated charge carrier separation and transfer. An illustrative example is the heterostructure of Zn-MOF-74@g-C_3N_4, showcasing the potential that the combined effect between piezoelectricity and step-scheme transfer of charges may drive the enhanced photocatalysis activity even under visible light irradiation [53].

In addition to strategies such as structural tuning and heterostructure construction, recent progress has also focused on expanding the functional applications of MOF-based photocatalysts in energy conversion and environmental remediation. Hydrogen peroxide (H_2O_2), with its high energy density, storability, and transportability, is emerging as a promising liquid fuel alternative to H_2 or fossil fuels, and its global demand is projected to reach 5.7 million tons by 2027 [54]. Since Yamashita and co-workers first reported MOF-based photocatalytic H_2O_2 production in 2018 [55], significant efforts have been made to overcome limitations such as poor charge separation and sluggish mass transfer (Fig. 4b). Huang *et al.* [56] developed MOF@MOF hierarchical heterostructures (PCN-134@MOF-525), achieving a 4.5-fold enhancement in H_2O_2 yield owing to improved charge separation and light harvesting, while Meng *et al.* [57] constructed redox-active single-atom centers in NH_2−MIL-101(Fe), with Cu-linked sites showing the best activity due to synergistic charge mobility and radical generation.

MOFs have also demonstrated remarkable potential in environmental remediation. For instance, MIL-100(Fe) and NH_2-UiO-66(Zr) composites efficiently degrade antibiotics and phenolic compounds under visible light through a synergistic adsorption-photocatalysis mechanism. Guo *et al.* [58] synthesized a dehydrated MIL-100(Fe)-250 *via* thermal treatment, which exhibited outstanding efficiency in removing emerging organic contaminants. The dehydrated MIL-100(Fe)-250 showed selectively accelerated removal of quinolone and phenolic pollutants, effectively resisting interference from coexisting substances. *In situ* infrared spectroscopy and DFT calculations revealed that the dehydration process modulated the steric hindrance and local electron density of Fe centers. This modulation enhanced charge transfer, facilitated Fe(III) reduction, and promoted H_2O_2 activation and conversion efficiency. Moreover, the dehydrated MIL-100(Fe)-250 maintained excellent adaptability over a wide pH range. This study highlights the enormous potential of spatially confined porous MIL-100(Fe)-250, where size-selective effects contribute to its superior catalytic performance. Therefore, this material represents a promising candidate for pollutant elimination in real aqueous environments, offering a viable strategy for advanced water purification. Zheng *et al.* [59] constructed a PDINH/NH_2-UiO-66 (PNU) Z-scheme heterojunction for simultaneous Cr(VI) reduction and antibacterial applications. The PNU-1 composite exhibited significantly enhanced photocatalytic activity, achieving a 97 % Cr(VI) reduction rate within 60 min, outperforming pristine PDINH and NU-66. In addition, PNU-1 showed excellent antibacterial properties. Comprehensive characterization and radical trapping experiments confirmed that the Z-scheme heterojunction effectively broadened light absorption, promoted charge-carrier separation through an internal electric field, and generated abundant reactive oxygen species, collectively improving photocatalytic degradation efficiency. Similarly, Cu-TCPP MOFs have been successfully applied for Cr(VI) reduction and dye degradation in wastewater, further demonstrating the feasibility and versatility of MOF-based photocatalysts for practical environmental remediation.

To situate MOF-based photocatalysts within the broader landscape of porous materials, it is important to consider cross-material synergies. Compared with MOFs, COFs generally provide enhanced π-conjugation and chemical stability, facilitating charge transport and structural robustness. MOF- and COF-derived carbons inherit the tunable architectures of their precursors while offering superior electrical conductivity and long-term stability, bridging the gap between crystalline frameworks and practical

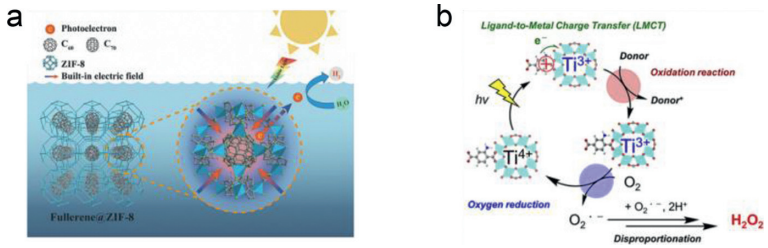


Fig. 4. (a) Mechanistic scheme of MOF-encapsulated fullerene solid solution for photocatalytic H_2 evolution. Reproduced with permission [52], Copyright 2025, Elsevier. (b) Schematic presentation of MOF for photocatalytic H_2O_2 production. Reproduced with permission [55]. Copyright 2022, Elsevier.

electrodes or catalysts. Carbon aerogels and graphene-based scaffolds can further complement these systems by improving mass transport, electron percolation, and mechanical resilience. Collectively, these observations highlight the value of integrated design strategies that combine the strengths of each class, offering multifunctional performance for energy conversion and environmental remediation.

3.4. Concluding remarks and prospects

MOFs continue to exhibit unprecedented potential as photocatalysts mainly owing to their very high surface area, tunable structural property, and functional capability. Coupling MOF composites with semiconductors or other functional units can compensate for major shortcomings of bare frameworks, including obstructed light absorption and rapid charge recombination, and allow for greater efficacy in energy and environmental applications. In particular, future efforts should focus on developing MOF-based photocatalysts that simultaneously achieve efficient energy conversion and effective pollutant degradation, aligning with the dual goals of carbon neutrality and environmental sustainability. Despite these advances, a number of crucial challenges remain against large-scale application. Narrow bands for optical absorption, requirements for sacrificial agents, low diffusivity through pores, and potential toxicity limit efficiency, long-term stability, and safety. These demands necessitate prudent selection of materials and vigorous testing of environmental compatibility.

New dimensions that borrow from the chemistry of bimetallic MOFs, interface engineering with conductive fillers, and embedding of piezo-photocatalytic conversions have come a long way, with activity increasing with increasing stability. There is still much work ahead of us as we try and span the gap between research prototypes and implementation.

Future work must be directed toward scalable and cost-effective synthesis routes, non-toxic, biocompatible materials, and long-lasting structural engineering capable of withstanding long-term operation. The research will involve interdisciplinary work between chemists and materials scientists and engineers as well as environmental experts such that seminal advances are brought to bear as practical technologies.

The conceptual framework of such perspectives is outlined in Fig. 5, showing the interface between challenges, strategies, and future prospects of MOF-based photocatalysts in the bigger picture of the roadmap.

Lastly, MOFs are an immensely versatile class of porous materials with the potential to become the key bearer of sustainable energy supply and environmental clean-up, if issues of scale, safety, and ecological compatibility are overcome.

4. COF for supercapacitors and batteries

Hui Dou*, Derong Luo, Feng Liu

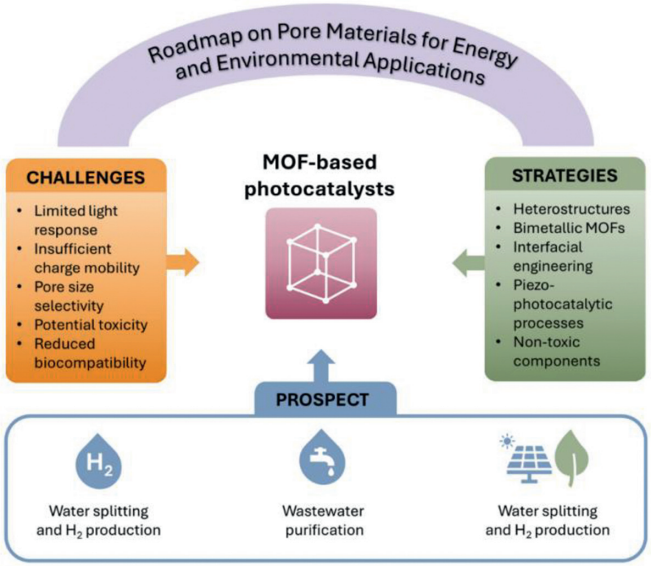


Fig. 5. Schematic roadmap of MOF-based photocatalysts, summarizing challenges, representative approaches, and future prospects.

4.1. Status

COFs are a novel class of organic crystalline materials developed from reticular chemistry concepts based on dynamic covalent chemistry, with the characteristics of long-range order, low density, great stability and high specific surface area [60–62]. COFs have garnered the wide attention in the field of energy storage because of the strong designability of the structure and functionality [63–65]. Despite a development history of <20 years, COFs have demonstrated promising potential for application in the energy storage technologies such as rechargeable metal ion batteries (Li^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, Zn^{2+}, Al^{3+}), Li-air batteries and capacitors (Fig. 6) [66,67]. This paper mainly reviews the recent research progress of COFs in the aforementioned fields.

4.2. Current and future challenges

The structural and functional diversity enable the wide applicability of COFs in energy storage technologies. The well-defined pore structures provide rapid channels for ion transport [68], and the strong covalent bond enables COFs to maintain a stable chemical structure, which can improve the resistance of solubility and alleviate the decomposition during electrochemical reactions. The designability of the structures allows the introduction of various functional groups into the framework, thereby creating a wide range of different active sites [69]. Besides, the electronic structure of COFs can be optimized by controlling building blocks, substituting groups and modification [70].

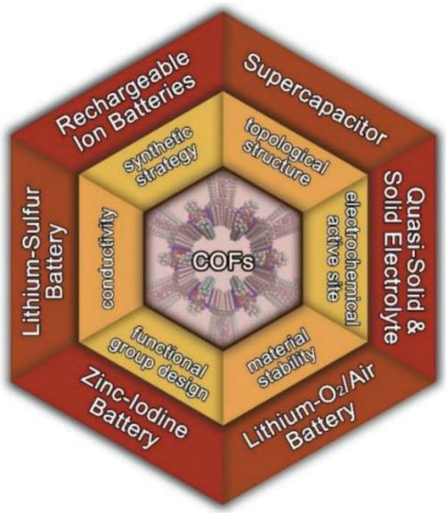


Fig. 6. Scientific challenges of COFs in the energy storage application.

However, the application of COFs still faces significant challenges, and the most important puzzle is that the internal relationship between material structure and electrochemical performance is not clear. Currently, the design of COFs primarily employs molecular engineering strategies focusing on the following aspects: (1) Customizing pore structures and topologies through the design of building units; (2) constructing specific redox-active sites through the selection of functional groups and covalent bonds, (3) enhancing the conjugation degree of the framework or fabricating composites to improve the conductivity. Additionally, the scalability and environment-friendly transformation of the synthesis process of COFs are essential issues that must be addressed for the practical application of COFs [71].

4.3. Advances in science and technology to meet challenges

The specific capacity and operating voltage of electrodes are critical parameters that impact the energy density, making the development of high-performance electrode materials a top priority in battery research [72]. The energy storage mechanism of COF electrodes is mainly achieved through redox reactions at the active sites. According to the types of active sites, COFs can be divided into n-type (containing carbonyl, imine, azo, disulfide bond, *etc.*), p-type (triphenylamine, carbazole, phenazine, phenoxazine, free radical, *etc.*) and bipolar materials. The n-type COFs follow an electron transport mechanism, which combines with metal ions in the discharging process (reduction reaction), while occurs the reversible reaction in the charging process (oxidation reaction). On the contrary, p-type COFs follow the hole transport mechanism, which loses the electrons and combines with the anions in electrolyte, thus exhibits higher redox potential and better reaction kinetics compared with n-type COFs. However, p-type COFs usually contain a large number of inactive groups and thus deliver low theoretical specific capacity, limiting their further development. It should be pointed out that the redox potential of the active site determines the suitability of the COFs as a positive or negative electrode. For example, the BQ1-COF cathodes based on C=N and C=O functional groups have the lowest proportion of inactive groups, indicating a high theoretical specific capacity of 773 mAh/g by the redox reactions involving 18 electrons (Figs. 7a-c) [73]. The p-type highly porous and crystalline azatruxene-based TAT-TA COF cathode obtains a discharge potential of up to 3.9 V (Figs. 7d and e) [74]. In addition, the COF@CNT as a LIBs anode usually have the superlithiation mechanism, which can form C_6Li_6 structure to provide a considerable discharge capacity (1536 mAh/g at 100 mA/g) (Figs. 7f and g) [75].

The stable porous structure and designable redox active sites also make COFs-based electrodes excellent performance in other rechargeable ion batteries (Na^+, K^+, Ca^{2+}, Mg^{2+}, Zn^{2+}, Al^{3+}, *etc.*). Moreover, by introducing specific active sites (carbonyl, imide, metal center, *etc.*), COFs can be applied to the sulfur positive electrode or separator modification of lithium sulfur batteries (LSBs) to effectively inhibit the polysulfide shuttle and promote polysulfide transformation. Similarly, the nitrogen-rich COFs can be used to improve the capture of iodine, thus build high performance zinc-iodine batteries. COFs are potential electrode materials for capacitors because of their high specific surface area, regular pore structure and modifiable functional groups. Moreover, COF can be used as cathode material and gas diffusion layer for lithium-air batteries with extremely high theoretical energy density, which helps address the challenges of sluggish ORR/OER kinetics and rapid degradation in ambient air [76]. However, their low electrical conductivity remains the primary obstacle limiting their electrochemical performance. Therefore, enhancing the conjugated structure of COFs or combining them with highly conductive materials (*e.g.*, CNT, graphene, MXene) is necessary to improve conductivity. Besides, the formation of the composite materials can also avoid excessive π-π stacking of the COF layers and improve the utilization of the active sites. More importantly, the strong structural designability of COFs enables their application as solid-state or gel electrolytes, providing a potential solution to the safety concerns associated with liquid electrolytes. For example, Liu *et al.* reported a rigid COF skeleton bonded with flexible, lithiophilic polyethylene glycol (PEG) chains to create a solid electrolyte using an electrospinning strategy [77]. The resulting PEG-COM electrolyte demonstrated improved dendrite inhibition and a high conductivity of 0.153 mS/cm at 30 °C.

4.4. Concluding remarks and prospects

Thanks to the robust foundation of organic chemistry, COFs have rapidly evolved into a burgeoning material discipline within a span of <20 years. COFs have demonstrated promising applications in various fields, particularly in energy storage. The research of COFs primarily involves the design and synthesis of materials, as well as performance analysis. This includes principles of structural design, synthesis strategies, material characterization techniques, comprehensive electrochemical performance testing, and the energy storage mechanism analysis by various *in-situ*/*ex-situ* techniques. While the application of COFs in the battery field is still in its early stages and encounters numerous challenges, we are confident that COFs will undoubtedly play an indispensable role in the realm of organic batteries with advancements in theoretical and experimental methods and a deeper understanding of these materials.

5. COF in electrocatalytic applications

Yixue Xu, Shun-Qi Xu*

5.1. Status

COFs, emerged as a novel class of crystalline porous polymers, have attracted extensive attention in catalysis due to their unique combination of modularity, porosity, and chemical functionality. Their reticular design enables precise control over pore size, topology, and functional site distribution, offering a rational platform for catalytic performance optimization [78]. The ordered channels within COFs promote efficient mass transport of reactants, intermediates, and products, while their chemically modular structure

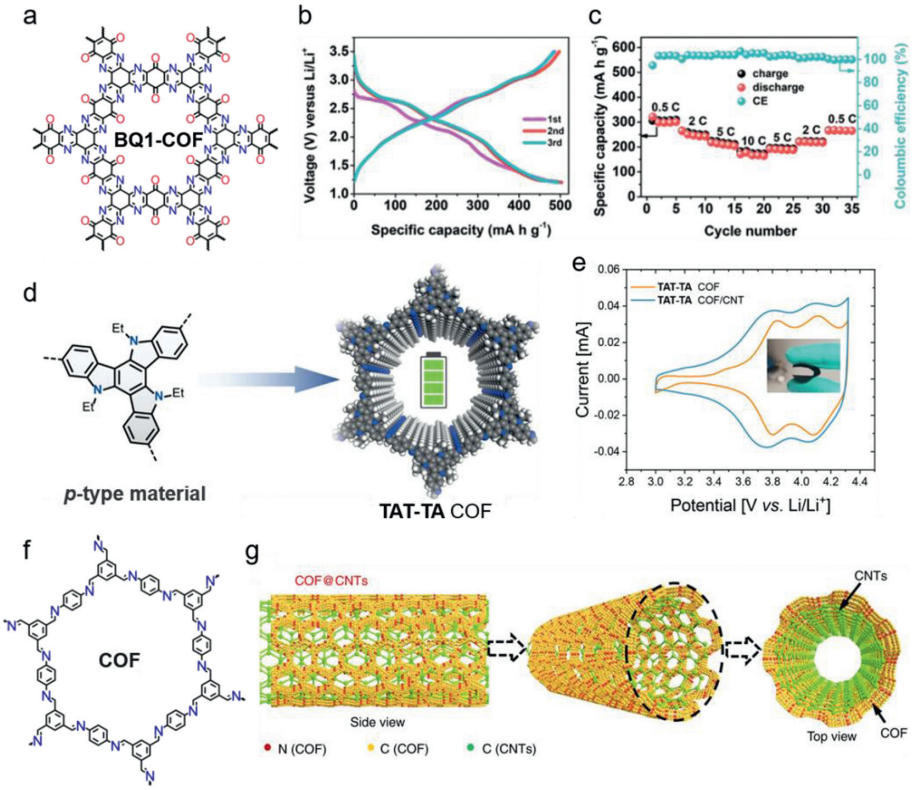


Fig. 7. COFs as electrode materials of LIBs. (a) Chemical structure of BQ1-COF. (b) The initial three-cycle capacity-voltage profile of BQ1-COF at a current of 0.05 C. (c) Rate capabilities of BQ1-COF at various current rates. Reproduced with permission [73]. Copyright 2020, Elsevier. (d) The azatruxene-based p-type TAT-TA COF and (e) CV curve at 0.2 mV/s of TAT-TA COF/CNT. Reproduced with permission [74]. Copyright 2024, American Chemical Society. (f) Chemical structure of COF and (g) graphical representation of COF@CNTs with few COF layers covered on the exterior surface of CNTs. Reproduced with permission [75]. Copyright 2018, Springer Nature.

allows heteroatom doping, extended π-conjugation, and incorporation of transition metals to tailor the electronic microenvironment of catalytic centers [79]. These merits make COFs particularly suited for electrocatalytic reactions where both electron transfer and molecular diffusion are critical, such as HER, OER, ORR, N_2 reduction reaction (NRR) and CO_2RR [64,80,81].

Beyond intrinsic design, COFs provide a platform to systematically study structure–activity relationships, since their well-defined pore environments and uniform distribution of active sites facilitate mechanistic investigations. Layer-by-layer growth, post-synthetic functionalization, and hierarchical porosity engineering have been explored to enhance performance by improving accessibility of catalytic centers, accelerating charge transport, and promoting reactant diffusion [82]. For example, 2D COFs with π-conjugated frameworks enable faster electron transport along the layers, while three-dimensional (3D) COFs with interconnected pores improve mass transfer. Furthermore, the tunable chemical environment of COFs enables precise regulation of the adsorption energy for reaction intermediates, providing a pathway to balance activity and selectivity. Collectively, these design principles establish COFs as highly modular and versatile platforms for next-generation electrocatalysts, bridging molecular precision and practical performance requirements.

5.2. Current and future challenges

In recent years, considerable progress has been achieved in the development of COFs for electrocatalysis. However, several intrinsic and practical limitations have highly hindered their application. Especially, the chemical stability remains a major concern. The construction of most COFs rely on dynamic covalent bonds, such as imine, boronate ester, or hydrazone linkages, which are prone to hydrolysis or oxidation under acidic, alkaline, or oxidative conditions encountered during HER, OER, ORR, NRR and CO_2RR [83]. Such degradation can lead to the collapse of crystallinity, loss of porosity, and rapid deactivation during prolonged operation. In addition, poor intrinsic conductivity limits charge transport, particularly in high-current-density reactions such as OER and ORR, where sluggish electron transfer can dominate overall kinetics [84]. Moreover, active site accessibility is sometimes insufficient; although COFs are inherently porous, catalytic centers can be partially buried within the framework or form aggregates, reducing effective utilization, especially in reactions like CO_2RR [85].

Another challenge lies in reaction selectivity, particularly for multi-electron processes. For instance, CO_2RR involves competing pathways producing CO, formate, or hydrocarbons, and uncontrolled binding of intermediates often lowers Faradaic efficiency [86]. Similarly, ORR can yield undesired peroxide species if electron transfer pathways are not properly modulated. Additionally, scalability and device integration remain obstacles: Conventional solvothermal synthesis often produces microcrystalline powders, which are difficult to process into mechanically robust, conductive electrodes. The absence of standardized protocols for long-term testing under industrially relevant conditions further complicates performance benchmarking. Addressing these challenges requires holistic strategies that simultaneously enhance stability, conductivity, active site accessibility, selectivity, and device compatibility.

5.3. Advances in science and technology to meet challenges

Recent research has introduced multiple strategies to overcome these limitations. Framework stabilization has been achieved through the incorporation of stronger covalent bonds, such as β-ketoenamine or C=C linkages, which significantly improve resis-

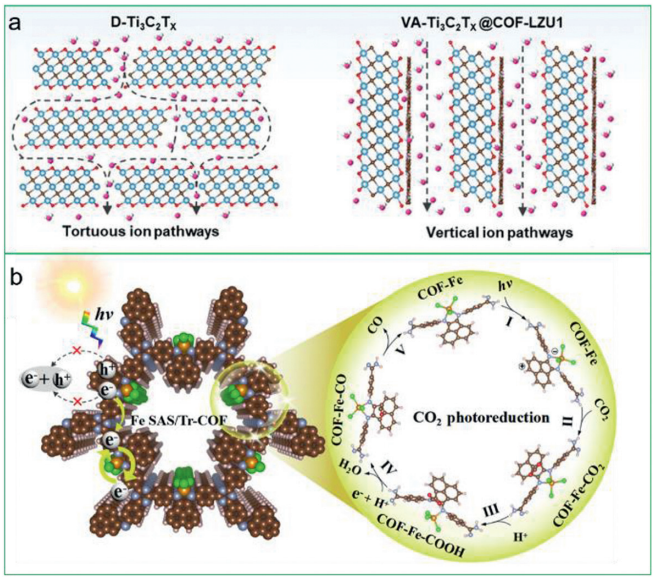


Fig. 8. (a) The ions transport in D-Ti_3C_2TX and VA-Ti_3C_2TX@COF-LZU1. Reproduced with permission [89]. Copyright 2023, Wiley. (b) Proposed reaction mechanism for photocatalytic CO_2-to-CO conversion over Fe SAS/Tr-COFs. Reproduced with permission [90]. Copyright 2022, American Chemical Society.

tance to hydrolysis and oxidative degradation [87]. Post-synthetic modifications, including cross-linking, surface hydrophobization, and embedding into protective matrices, have also been shown to maintain structural integrity under harsh electrochemical conditions [88].

Conductivity enhancement has been realized through several complementary strategies. Highly conjugated building blocks, such as thiophene, carbazole, or pyrene derivatives, extend π-electron delocalization, while hybridization with conductive carbonaceous materials (graphene, carbon nanotubes) or 2D materials (MXenes) further facilitates electron transport. For example, vertically aligned conductive $Ti_3C_2T_x$ MXene integrated with interlayer electroactive COFs has been used to fabricate hierarchically ordered hybrid electrodes. This hybrid nanosheet architecture maximizes the exposure of the intrinsic nanopores of COFs, improves ion transport pathways and accessibility, and enhances overall electrochemical activity (Fig. 8a) [89].

Single-atom catalysis within COFs represents a transformative approach. The well-defined coordination environment stabilizes isolated metal atoms (*e.g.*, Fe, Co, Ni, Zn, Cu, Mn, and Ru), ensuring maximal atomic utilization and tunable electronic structure. For instance, Fe SAS/Tr-COF achieves a CO generation rate as high as 980.3 μmol g^{-1} h^{-1} under visible light with a selectivity of 96.4%, approximately 26 times higher than that of the pristine COF (Fig. 8b) [90]. Beyond CO_2RR, constructing atomically dispersed Au sites within COFs and precisely tuning the microenvironment of the Au catalytic centers, by modulating the positions of proximal and distal porphyrin functional groups, have also been shown to effectively enhance catalytic activity for NRR [91].

Electrode integration strategies have evolved to enable practical application. *In situ* growth of COF films on conductive substrates ensures uniform coverage, low interfacial resistance, and strong adhesion, while ultrathin nanosheets maximize active surface area and promote mass transport [92]. Coupled with gas diffusion electrode (GDE) configurations, these materials demonstrate excellent performance under industrially relevant conditions, including high current densities and continuous operation.

Mechanistic insights have been greatly advanced by *operando* characterization and theoretical modeling. Techniques such as X-ray absorption spectroscopy (XAS), Raman spectroscopy, and electron paramagnetic resonance (EPR) enable real-time monitoring of active sites, intermediates, and electronic structures. DFT calculations and molecular dynamics simulations provide atomic-level understanding of heteroatom effects, metal coordination, pore environment, and reaction energetics, guiding rational design and optimization.

5.4. Concluding remarks and prospects

COFs, with their tunable frameworks, chemical versatility, and ordered porosity, have emerged as promising electrocatalysts for HER, OER, ORR, NRR and CO_2RR. Recent advances in framework stabilization, conductivity enhancement, single-atom integration, and nanoscale engineering have markedly improved both activity and selectivity, bridging the gap between molecular design and practical application. Future research should focus on three key directions: (1) Ultrastable framework design, capable of enduring extreme pH, high potentials, and oxidative conditions for long-term operation; (2) Atomic-level site engineering, including co-doping, ligand modulation, and controlled defect introduction, to precisely tune electronic and geometric properties for enhanced selectivity and activity; (3) Scalable electrode fabrication and device integration, ensuring mechanical robustness, high conductivity, and compatibility with standardized testing protocols, enabling industrial deployment.

The integration of predictive computational modeling, advanced *operando* characterization, and innovative synthetic strategies will be essential to accelerate rational catalyst design. By combining these approaches, COFs can be engineered into high-performance, selective, and durable electrocatalysts, contributing to sustainable hydrogen production, CO_2 conversion, and large-scale energy conversion technologies. These developments not only expand the fundamental understanding of structure–activity relationships but also lay the foundation for the translation of COFs from laboratory research to practical industrial applications.

6. COF for photocatalysis

Chunyang Dong*

6.1. Status

By harvesting energy directly from the renewable solar light to initiate chemical reactions, photocatalysis has demonstrated great potential in waste valorization, energy storage, chemical synthesis and environmental remediation [93–96]. The development and rational design of photo-responsive material are fundamental aspects in photocatalytic studies [97–99]. Solid semiconductors, especially metal oxides, nitrides and their derivatized composites are well studied photocatalysts [100]. However, the rigid and bulk-like structure of such materials has greatly restricted their surface properties and number of surface coordination sites for modification and enhancement, making the current photocatalytic processes less efficient and not yet economically viable.

As an emerging type of photocatalysts, COFs with superior structural advantages relative to traditional bulk semiconductors have received great attention in recent years. COFs are essentially crystalline porous polymers with periodically extended π units as building blocks and connected by strong covalent bonds [101,102]. The diversity of topologies and building blocks for synthesis endows COFs with tailorable structural and catalytic properties [103,104]. To date, advanced COFs with precise architectures have achieved remarkable performance in numerous photocatalytic reactions, including solar-driven water splitting, CO_2 re-

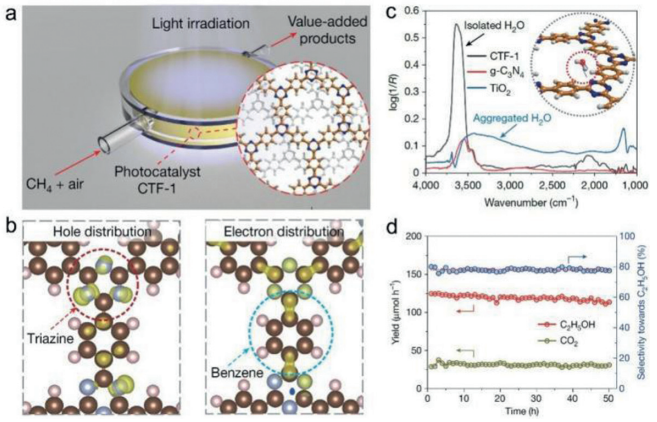


Fig. 9. (a) Schematic illustration of photocatalytic transformation of methane and air selectively to ethanol over COF. (b) The molecular structure of hole and electron distribution over the triazine and benzene unite within the COF photocatalyst respectively. (c) DRIFT spectra of different photocatalysts after a saturated H_2O adsorption. (d) Long-term stability test of COF photocatalyst in methane oxidation to synthesize ethanol. Reproduced with permission [105]. Copyright 2025, Springer Nature.

duction, pollutant degradation, and green chemical synthesis [105–116].

Compared with inorganic semiconductors, COFs offer long-range π-conjugation, molecular-level tunability, and well-defined donor-acceptor (D-A) frameworks that enable efficient light absorption and charge separation [117,118]. Their adjustable pore structures further promote mass transport and provide accessible catalytic sites for solar-driven reactions. The intrinsic porosity and modularity of COFs also make them ideal for selective adsorption and degradation of organic pollutants, representing a sustainable platform for environmental remediation.

6.2. Current and future challenges

The main targets of photocatalysis contain the following three sections. First, the photocatalytic processes should achieve higher efficiency, precision and long-term stability. Second, both the photocatalyst and reaction medium should be cost-effective and avoid involving harsh conditions and toxic reagents [119]. Finally, the photocatalytic systems should aim for industrial demand, pursuing scalable production/treatment possibility [120].

Taking methane oxidation as an example, since methane molecules are thermodynamically more stable than the desirable oxygenate products (*e.g.*, methanol, ethanol), achieving high methane conversion while preserving singular target product is extremely challenging [93]. Recently, Tang and coworkers addressed such challenges by utilizing a triazine-based COF photocatalyst, enabling selective ethanol synthesis alongside significantly improved methane conversion (Fig. 9a). Notably, the alternating triazine and benzene unit of COF not only modulated the distribution of photo-excited charge carriers (Fig. 9b) but also promoted the spillover and desorption of intermediates and products. Meanwhile, H_2O molecules, a key ingredient in ethanol synthesis were effectively adsorbed by the COF material due to its intrinsic porous structure (Fig. 9c). This resulted in moderately selective methane activation with excellent operational stability (Fig. 9d) [105].

Another representative application for COF-based photocatalysts is the synthesis of H_2O_2 directly from H_2O and oxygen. Recently, with the development of an unique doner-acceptor (D-A) type COF material, Jiang and coworkers showcased the sustainable synthesis of H_2O_2 without requiring a noble metal cocatalyst or sacrificial reagent, using air as the sole oxidant [108]. In this work, the non-conjugated hydrazone linkage separated the HOMO and LUMO of this D-A COF material, facilitating a rapid photo-excited electron transfer and charge separation. Besides, the intrinsic one-dimensional channel, with its hydrophilic nature, enhanced the delivery of H_2O and O_2 molecules for their oxidation and reduction, respectively.

Beyond energy conversion, COF photocatalysts have shown increasing potential in environmental remediation, such as photodegradation of dyes, antibiotics, and volatile organic compounds (VOCs). However, challenges persist: (1) The relatively narrow visible-light response window of many COFs, (2) inefficient charge separation and migration across π-stacked layers, (3) limited long-term stability under continuous irradiation, and (4) high synthetic cost and structural complexity that restrict large-scale deployment. In addition, the durability and recyclability of COFs during pollutant degradation, especially under realistic water or air environments, remain underexplored, requiring more systematic evaluation of stability, reusability, and potential secondary pollution from linker degradation. Moreover, the mechanistic understanding of photoinduced charge dynamics and active-site evolution is still incomplete, calling for advanced *in situ* characterization and theory-guided design to accelerate material discovery and ensure environmental safety in real applications.

6.3. Advances in science and technology to meet challenges

Recent research efforts have focused on addressing the above challenges through several key strategies:

(1) Bandgap and D-A structure engineering. Incorporating strong donor-acceptor pairs and conjugated linkers (*e.g.*, triazine, porphyrin, benzothiadiazole) can significantly narrow bandgaps and enhance visible-light harvesting. These D-A COFs exhibit directional charge separation, improving overall photocatalytic efficiency [121].
(2) Construction of COF-based heterostructures. Forming heterojunctions with semiconductors (g-C_3N_4, TiO_2, MXenes) or embedding metal nanoparticles can promote charge transfer at interfaces and improve structural stability [122]. Such hybrid systems combine the high surface area and porosity of COFs with the electrical conductivity of inorganic components.
(3) Mechanism-guided design using *operando* and computational tools. *In situ* spectroscopic methods (XPS, transient absorption, EPR) reveal charge-carrier dynamics and active-site evolution [123]. Combining these insights with DFT and machine learning enables predictive design of COFs with optimal band structure, charge separation efficiency, and catalytic activity.
(4) Scalable synthesis and device integration. Interfacial polymerization, microwave-assisted synthesis, and 3D printing enable gram-scale and thin-film COF fabrication. These approaches facilitate integration into photoelectrochemical cells, solar reactors, and wastewater treatment systems, expanding their utility from energy conversion to environmental purification.

To contextualize COF-based photocatalysts within the broader landscape of porous materials and highlight potential synergies, it is instructive to compare COFs with MOFs, MOF/COF-derived carbons, and carbon aerogels. Compared with MOFs, COFs often exhibit higher π-conjugation and chemical robustness, facilitating directional charge transport and long-term stability under operational conditions. MOF- and COF-derived carbons inherit the tunable architectures of their precursors while providing enhanced electrical conductivity and mechanical resilience, thereby bridging the gap between crystalline frameworks and practical functional electrodes or catalysts. Carbon aerogels and graphene-based scaffolds further complement these systems by improving mass transport, electron percolation, and flexibility.

Collectively, these insights underscore the importance of integrated design strategies that leverage the strengths of each material class: MOFs for structural and compositional tunability, COFs for electronic robustness and precision, derived carbons for conductivity and stability, and carbon aerogels for hierarchical transport and mechanical support. Such a cross-material perspective provides a continuous roadmap rather than isolated snapshots, emphasizing opportunities for hybrid architectures that achieve multifunctional performance in both energy conversion and environmental remediation.

6.4. Concluding remarks and prospects

With innovations in structural engineering, synthetic methods, and data-driven design, the applicability of COF-based photocatalysts is evolving rapidly. The highly tailorable porous architecture and modular building blocks endow COFs with superior advantages in charge separation and reactant activation. However, their precise structures often come with high production costs and limited scalability. To meet industrial and environmental demands, future research must develop cost-effective, durable, and recyclable COFs capable of operating under realistic conditions.

Looking toward 2030, the future development of COF-based photocatalysts should emphasize four constructive directions: (1) Scalable and low-cost synthesis while maintaining crystallinity and activity; (2) integration of conductive and flexible components to improve charge separation and durability; (3) mechanism-driven design through *operando* and AI-assisted approaches to achieve predictive control over active-site functionality; and (4) exploration of multifunctional COFs that couple solar energy conversion with pollutant degradation and environmental remediation.

Ultimately, the convergence of structural precision, electronic tunability, and multifunctionality will position COFs as next-generation programmable photocatalysts bridging solar energy conversion, green chemical synthesis, and sustainable environmental technologies by 2030.

7. Porous carbon materials derived from MOFs and COFs for energy and environmental applications

Lan Ding*

7.1. Status

Building upon the preceding discussion of COF-based photocatalysts, porous carbon materials derived from MOFs and COFs have emerged as critical bridging components that integrate structural tunability, electronic functionality, and mechanical robustness. These materials have become indispensable in energy storage, electrocatalysis, photocatalysis, and environmental remediation owing to their exceptional electrical conductivity, chemical stability, tunable porosity, and scalable synthesis. In particular, MOF- and COF-derived carbons represent a new generation of framework-templated porous architectures that inherit the long-range order, compositional versatility, and modular design of their crystalline precursors, while providing the conductivity and stability required for practical device integration [124,125].

Through carefully controlled pyrolysis or carbonization, these frameworks can be transformed into heteroatom-doped carbon matrices or metal–carbon composites that retain hierarchical pore structures, optimize electronic conductivity, and facilitate rapid ion and mass transport. This approach effectively bridges the gap between crystalline frameworks and functional carbon architectures, enabling a seamless transition from molecular-level design principles to high-performance electrochemical and photocatalytic applications.

Recent studies have demonstrated that MOF- and COF-derived carbons exhibit outstanding performance across a wide spectrum of applications, including supercapacitors, lithium- and sodium-ion batteries, and electrocatalysis (HER, OER, ORR), as well as photocatalysis and environmental pollutant removal. Compared with pristine MOFs and COFs, the derived carbons offer markedly enhanced conductivity, chemical robustness, and long-term stability, enabling reliable operation under harsh electrochemical or oxidative conditions [126,127]. Moreover, their hierarchical pore connectivity, tunable surface chemistry, and capacity for heteroatom or metal functionalization promote rapid charge, ion, and mass transport, key attributes for the design of next-generation electrochemical devices, hybrid photocatalysts, and integrated energy–environmental platforms.

By situating these derived carbons within the broader context of porous materials, it becomes evident that they not only complement the functionalities of MOFs, COFs, and carbon aerogels but also serve as a unifying platform for multi-material hybridization. Such integration underscores the potential of MOF/COF-derived carbons to contribute to the roadmap toward 2030, where scalable, multifunctional porous architectures will be essential for meeting energy conversion, storage, and environmental remediation targets.

7.2. Current and future challenges

Despite these significant advances, several critical challenges continue to hinder the rational design and large-scale utilization of MOF- and COF-derived carbons. The retention of structural integrity during high-temperature carbonization remains a fundamental difficulty, as phase collapse and pore coalescence often occur, weakening the structural hierarchy inherited from the precursor. Furthermore, precise control over heteroatom distribution and metal dispersion is essential to ensure homogeneous active site environments and reproducible catalytic activity.

Another major challenge lies in achieving an optimal balance between porosity and conductivity. While increased graphitization enhances charge transport, it often sacrifices accessible surface area and reduces the density of active sites. From a practical perspective, the scalability and sustainability of current synthesis protocols are limited by energy-intensive thermal treatments and the use of hazardous solvents or metal precursors. Moreover, although MOF- and COF-derived carbons share mechanistic similarities with carbon aerogels and graphene-based frameworks, these systems are often studied independently rather than within an integrated design continuum. This fragmentation restricts the ability to develop unified, multifunctional architectures that combine conductivity, reactivity, and environmental resilience.

7.3. Advances in science and technology to meet challenges

Recent scientific and technological developments have substantially advanced the controlled synthesis and functional integration of MOF- and COF-derived porous carbons, enabling hierarchical architectures with precise electronic, structural, and catalytic properties. One widely adopted approach is controlled pyrolysis under optimized atmospheres, which preserves the precursor's morphology and pore hierarchy. For instance, ZIF-8-derived carbons carbonized under nitrogen at 900 °C retain a high surface area of over 1200 m^2/g, predominantly microporous architecture, and uniformly distributed nitrogen dopants, leading to superior supercapacitor performance with capacitance exceeding 400 F/g at 1 A/g. Similarly, COF-derived carbons subjected to stepwise pyrolysis maintain their π-conjugated frameworks, resulting in improved electronic conductivity and structural stability that are critical for high-rate lithium-ion storage and long-term electrocatalytic operation.

Heteroatom engineering has become a cornerstone strategy to tailor the electronic structure and catalytic activity of these carbons. Co-doping with nitrogen and sulfur, or phosphorus and boron, modifies local charge densities, enhances spin polarization, and introduces abundant active sites for redox reactions. For example, MOFs have been employed as structural templates to fabricate hierarchically porous nanocarbon materials with abundant mesopores, such as NPS-C-MOF-5 obtained through N/P/S ternary heteroatom doping. The simultaneous incorporation of nitrogen, sulfur, and halogen dopants enhances the polarization of C–C bonds and modulates the local electronic environment, thereby creating a high density of active sites. As a result, the catalyst exhibits markedly improved ORR activity under both alkaline and acidic conditions, highlighting the effectiveness of MOF-derived heteroatom-doped carbons for electrocatalysis [128]. Similarly, N-doped COF-derived carbons enhance hydrogen evolution reaction kinetics by promoting proton adsorption and electron transfer efficiency [129].

At the interface level, metal–carbon coupling has been widely employed to stabilize single-atom catalysts or ultrafine nanoparticles within nitrogen-doped carbon matrices. Fe–N–C single-atom sites embedded in MOF-derived carbons exhibit remarkable ORR activity and durability, with negligible activity loss over 50,000 potential cycles. Hybridization with secondary carbon scaffolds, such as graphene, carbon nanotubes, or aerogel networks, further enhances electron percolation, ion diffusion, and mechanical flexibility. A recent study demonstrated that uniformly anchoring cobalt-containing catalysts onto COF-derived carbon enabled a Li-S battery to deliver a high specific capacity of 1288 mAh/g at 0.3 C, with 92 % capacity retention over 200 cycles and excellent long-term cycling stability [130]. Moreover, sustainable and scalable synthesis strategies have gained prominence. Biomass-MOF composites, solvent-free assembly, and low-temperature plasma carbonization allow production of hierarchically porous carbons while reducing energy consumption and environmental footprint.

Collectively, these strategies converge toward integrated carbon architectures that combine hierarchical porosity, tunable surface chemistry, and robust mechanical properties. Such architectures not only enhance conductivity and catalytic activity but also provide a flexible platform for the design of multifunctional systems that couple charge, ion, and mass transport.

7.4. Concluding remarks and prospects

Porous carbons derived from MOFs and COFs are emerging as a bridging class of materials that connect the structural precision of crystalline frameworks with the practical functionality of advanced carbons. Their intrinsic combination of high conductivity, chemical resilience, and tunable micro–meso–macroporosity positions them as indispensable components in future energy conversion and environmental purification technologies. Moving forward, several research directions will be critical to realize their full potential.

First, establishing quantitative structure–property relationships through *in situ* spectroscopy and multiscale modeling will enable predictive control over precursor-to-carbon transformation. Second, multi-heteroatom co-doping and single-atom coordination strategies should be pursued to fine-tune local electronic structures and enhance intrinsic catalytic activity. Third, integrative design across carbon-framework-aerogel systems will be essential to promote coupled charge, ion, and photon transport, leading to multifunctional energy-environment platforms. Finally, comprehensive life-cycle assessment and carbon footprint analysis are urgently needed to align porous carbon technologies with sustainable manufacturing and circular economy principles. In summary, MOF- and COF-derived carbons serve as the conductive backbone within the broader ecosystem of porous materials. Their design evolution not only enhances the performance of energy and environmental devices but also defines a new paradigm of cross-material integration that will underpin the next generation of sustainable porous architectures toward 2030 and beyond.

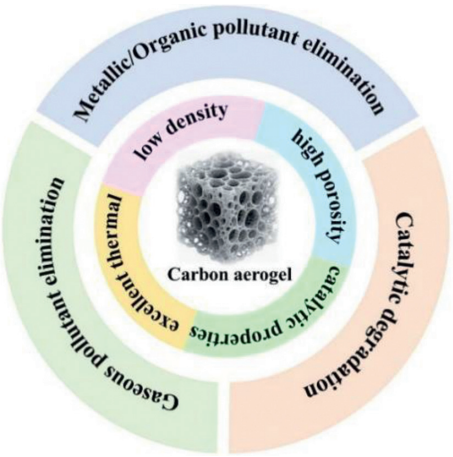


Fig. 10. Environmental application of carbon aerogels.

8. Carbon aerogels for environmental applications

Zijun Huang, Yongming Luo, Dedong He*

8.1. Status

With the continuous advancement of research, the family of aerogel materials has expanded significantly and has been recognized as one of the top ten technological and scientific breakthroughs [131–133]. Over the past few years, carbon aerogels (CAs) have attracted significant attention owing to their unique 2D carbon nanomaterial propertie and 3D structure. Their unique combination of structural properties (low density and high porosity) and surface characteristics (large specific surface area and numerous active sites) renders them highly promising for environmental applications [134–137]. It is specifically manifested in the following three aspects, as shown in Fig. 10: (1) Metallic/organic pollutant removal. Their high surface area and hierarchical pore structure provide abundant adsorption sites, enabling highly efficient removal of metallic ions and other aquatic pollutants. Furthermore, owing to their low density and super-hydrophobicity, aerogels can float on the water surface and exhibit excellent adsorption performance toward organic contaminants. (2) Gaseous pollutant elimination. Their high surface area and porous structure enhance pollutant diffusion and exposure to active sites, significantly improving removal efficiency for volatile organic compounds in air purification applications. (3) Photocatalysis. Owing to their outstanding light absorption capability, high electrical conductivity, and porous architecture that facilitates photogenerated carrier migration, CAs have garnered an exponentially surging research interest in photocatalysis [138].

8.2. Current and future challenges

Recently, CAs show great potential in the field of materials science, however, their industrial production still faces several challenges. Firstly, the production cost remains high. The preparation process of CAs is complex, and the cost of raw materials is relatively high. Especially for high-performance carbon aerogel, the materials such as carbon nanotubes and graphene required are extremely expensive. Furthermore, steps like supercritical drying

need specialized equipment, which further increases production costs and limits large-scale application. Secondly, there are difficulties in synthesis efficiency and environmental protection. The preparation of carbon aerogel involves multiple steps and the production process is quite complex. During the production and processing of CAs, waste materials may be generated, which can have a negative impact on the environment. Therefore, targeted research is needed to overcome these challenges. Potential solutions include developing low-cost alternative raw materials, such as biomass-derived carbon sources (*e.g.*, cellulose, chitosan) to replace graphene or carbon nanotubes, thereby reducing material costs. It is also essential to optimize the sol-gel process parameters and utilize AI-assisted design to maximize production efficiency. Furthermore, efforts should be directed toward developing green synthesis methods, advancing the understanding of synthesis mechanisms, and improving the overall environmental friendliness.

8.3. Advances in science and technology to meet challenges

CAs, as sustainable functional materials with tremendous application potential, are accelerating their advancement toward industrialization. Their core advantage stems from the synergistic effect of their 3D porous structure and high density of active sites [139]. This structure-function integrated design is key to its ability to adsorb water pollutants and gaseous pollutants. Specifically, the 3D hierarchical pore system effectively addresses the issues of active site agglomeration and adsorption capacity decay common in traditional materials: the nanoscale micropores provide high specific surface area and a high density of adsorption sites, while mesopores and macropores serve as efficient mass transfer channels. This enables rapid diffusion of pollutant molecules to active sites while preventing agglomeration and deactivation of active components. In terms of surface functionalization, CAs can not only introduce specific functional groups *via* surface crosslinking, but also support metal nanoparticles to create bifunctional catalytic sites [140]. The abundant defect sites and functional groups within CAs create numerous anchoring positions for nanocatalysts. During catalyst preparation, these features enable the catalysts to be highly dispersed and immobilizes catalytic particles. Consequently, the efficiency and stability of the reaction centers are significantly improved [141]. In addition, their excellent conductivity and carrier mobility further enhance charge separation and redox reaction efficiency in photocatalytic processes. Currently, CAs demonstrate promising application potential in environmental remediation fields such as wastewater treatment and air purification [142]. Despite these advances, the complexity of precisely controlling pore architecture and surface functionalization during synthesis can lead to potentially affecting performance in practical applications.

8.4. Concluding remarks and prospects

CAs represent a class of highly promising functional materials, exhibiting exceptional structural and surface properties that enable high performance in environmental remediation, including metallic/organic pollutant adsorption, gaseous purification, and photocatalysis. Despite significant advances, their industrial scalability remains constrained by high costs, complex synthesis, and environmental concerns. Future efforts should focus on developing low-cost and green synthesis routes, optimizing process parameters with AI assistance, and achieving precise control over pore structures and surface properties. Overcoming these challenges will accelerate the industrialization of CAs and unlock their full potential in sustainable applications.

9. Recent advances in aerogel photocatalyst

Ruiyang Zhang*, Ying Zhou

9.1. Status

Indeed, the advancement of high-performance, yet economically viable catalysts is vital to the progression of photocatalytic applications. However, two primary challenges significantly hamper their real-world efficacy: the low efficiency of the photocatalyst and the practical difficulties associated with handling powdered catalysts, particularly regarding their recovery. These factors collectively impede the large-scale application of photocatalytic technologies [143]. To address these shortcomings, aerogel-based photocatalysts, which boast exceptional monolithic characteristics, emerge as a promising solution. Their unique structure not only enhances the photocatalytic activity but also facilitates easier usage and recycling, thereby potentially overcoming the limitations inherent in conventional powdered catalysts [132,136,144]. In our preceding reports, we have outlined the challenges encountered with aerogel catalysts, encompassing issues related to fabrication techniques, photocatalytic activity and the reaction processes [145]. Furthermore, we delineated the avenues for prospective research and development in this domain.

Recently, the methodology combining straightforward surface functional group crosslinking with freeze-drying techniques has gained extensive application in the fabrication of aerogel photocatalysts [146,147]. This approach has proven advantageous in facilitating the fabrication of aerogel materials from crystalline catalyst precursors, thereby enhancing their catalytic performance. The integration of 3D printing technology in the fabrication of aerogel materials has illustrated the influence of a micro scaffold structure on enhancing catalyst efficacy [148,149]. This methodology highlights the potential for tailored structural design in aerogels, offering new avenues to optimize catalyst performance through precise control of their architecture. In terms of catalytic activity, the attainment of quasi-homogeneous photocatalytic activity has been successfully demonstrated through the design of photoactive components within aerogels [150]. By integrating single atom active centers onto the aerogel's surface, a synergistic effect is harnessed that combines the advantageous porous nature of the aerogel architecture with the interactions occurring between single atoms [151]. This approach exhibits outstanding catalytic capabilities, underscoring the potential of such aerogel-based catalysts for enhanced reaction efficiency and selectivity in catalytic processes. The catalytic process employing aerogel-based photocatalysts exhibits immense promise for practical applications, particularly in photothermal coupled water splitting as a sustainable method for hydrogen generation [152]. Capitalizing on their inherent characteristics of low density, which enables exceptional floatability, these aerogels efficiently harness solar energy. This unique combination not only facilitates their easy exposure to sunlight in aqueous environments, but also enhances thermal management during the reaction, thereby optimizing the hydrogen production efficiency. Consequently, aerogel photocatalysts are poised to play a pivotal role in advancing green energy solutions through innovative photocatalytic processes.

9.2. Current and future challenges

Despite the notable strides in the advancement of aerogel photocatalysts in recent years, which have witnessed rapid development across their preparation methodologies, enhanced catalytic activities, and optimized reaction process, it is evident that sustained endeavors will be indispensable to further propel progress

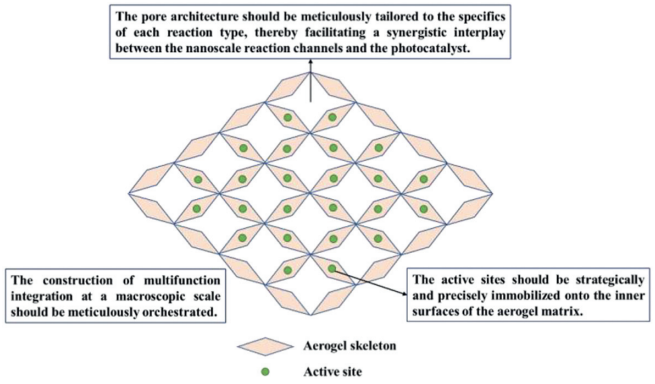


Fig. 11. Diagram of aerogel photocatalyst.

in these core issues. Moreover, as the comprehension of the underlying photocatalytic processes deepens and aerogel materials increasingly find applications, a fresh set of challenges has emerged, which is summarized in Fig. 11.

A particular hurdle lies in fully capitalizing on the unique micro-porous architecture of aerogels. Aerogel materials with porosities often reaching an astonishing 99 % boast a profusion of pores that have proven instrumental in accelerating photocatalytic reaction kinetics [133]. Nonetheless, precisely engineering and controlling this complex pore structure remains a formidable challenge. Consequently, while aerogel-based materials in theory possess the potential to surpass even molecular sieve in terms of catalytic activity from the point of view of its pore structure, realizing this superiority has proven difficult. This entails the precise design and fabrication of complex pore structures that are optimized for enhanced light harvesting, efficient charge carrier separation, effective gathering of reactants, and facilitated diffusion of reaction products. While 3D printing technology can exert control over the pore architecture, achieving high precision in pore structure and shape optimization still poses great challenges. Furthermore, the development of precise methods to immobilize active sites onto the profuse interior surfaces of aerogels remains an area warranting exploration.

Concurrently, apart from meeting the strength prerequisites of the aerogel's macrostructure, integrating the aerogel photocatalyst into practical applications presents an additional challenge. This entails resolving issues of application compatibility, which necessitates multifunctionality to accommodate the complex requirement of real-world environments. For example, in the case of photothermal coupling hydrogen generation [152], practical implementation necessitates that the aerogels possess hydrophilicity, which is crucial for maintaining the reactivity at the solid-liquid interface, and also must endure rigorous conditions, including variable pH levels and high salinity typically encountered in aqueous media. Conversely, a fraction of the aerogel must exhibit hydrophobic characteristics to prevent water refraction and reflection, thereby safeguarding light absorption efficiency. Furthermore, the aerogel's skeletal framework should demonstrate exceptional electrical conductivity to facilitate the efficient migration of photogenerated charge carriers. Therefore, adapting to real-world environments by achieving multifunctionality is another challenge for aerogel photocatalyst.

9.3. Advances in science and technology to meet challenges

Addressing these challenges necessitates advancements in fabrication techniques, alongside a multidimensional comprehension of the photocatalytic reaction mechanisms. It also calls for the integration of novel concepts and insights from interdisciplinary collaborations. The rapid progression of artificial intelligence and big data analytics now enables the efficient delineation of the intricate links between material composition, structure, and their catalytic activity [153,154]. This, in turn, paves a swift avenue for tailoring the composition and architecture of aerogel-based photocatalysts. Concurrently, the evolution of spectroscopic technologies has empowered researchers to monitor charge carrier dynamics across multiple temporal and spatial dimensions, thereby unraveling the intricate microscale events underlying photocatalytic processes [155]. These technological strides collectively contribute to a deeper understanding and accelerated optimization of aerogel materials for enhanced performance. Despite efforts by researchers to address real-world challenges using aerogel photocatalysts, much of the current research merely converges the porous structure of aerogels with photocatalytic properties, struggling to fully demonstrate the application process of aerogels. By advancing fabrication techniques, such as nanoscale-precise 3D printing technologies, aerogels can be engineered to attain an optimal fusion of nano reaction channel design and catalytic capability, which integration promises to enhance not only the catalytic efficiency but also the selectivity of reactions, thereby unlocking their latent potential in practical applications.

9.4. Concluding remarks and prospects

In recent times, the evolution of fabrication techniques for aerogel photocatalysts has progressed relentlessly, leading to unprecedented enhancements in catalytic performance and a diversification of application domains. At the same time, the challenges inherent to the practical implementation of aerogel photocatalysts have risen to prominence. Capitalizing fully on their distinctive attributes stands as a paramount challenge in the practical deployment of these aerogels, encompassing the engineering of microporous architectures, the exact placement of active sites on interior surfaces, and the integration of multifunctionality at a macroscopic scale. It is anticipated that, in the future, aerogel photocatalysts, with their distinctive properties and functionalities, will excel and illuminate the realm of practical applications.

10. Porous metal oxide nanomaterials for electrocatalysis and photocatalysis

Yunyun Ma, Zhuo Xing*

10.1. Status

Electrocatalysis has emerged as a promising strategy to address the global energy crisis and mitigate environmental pollution by enabling sustainable energy conversion and storage from renewable electricity. Central to these technologies are electrochemical reactions such as the HER, OER, and ORR, which are essential for processes ranging from water electrolysis to fuel cells [156–158]. Currently, benchmark electrocatalysts for these reactions are dominated by noble-metal-based materials, such as Pt, Pd, and Ir. Their exceptional activity arises from favorable electronic structures, including optimal D-band center positions, narrow band gaps, and diverse ligand coordination environments, which allow moderate adsorption of reactants and facile desorption of products. However, the scarcity and high cost of noble metals severely limit their large-scale application. This has motivated extensive efforts to explore earth-abundant alternatives. Among them, porous metal oxide nanomaterials have emerged over the past decades as attractive candidates due to their structural versatility, tunable electronic states, and relatively low cost [159].

Beyond electrocatalysis, similar structure-property relationships are also critical in photocatalysis, where photoexcited charge gen-

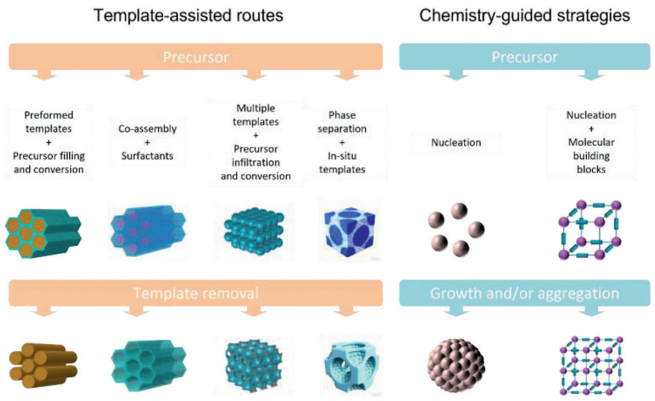


Fig. 12. The principal methods for synthesizing mesoporous materials: Template-assisted routes and chemistry-guided strategies.

eration, separation, and surface redox reactions are similarly governed by the band structures and surface states of metal oxides. Their tunable band positions and intrinsic stability make porous oxides highly promising for solar-driven hydrogen evolution and CO_2 reduction. Moreover, their redox flexibility and reactive oxygen species (ROS) generation capability endow them with additional potential for environmental remediation, such as degradation of organic pollutants and detoxification of wastewater streams.

10.2. Current and future challenges

Porous metal oxide nanomaterials, defined by pore sizes below 100 nm, provide large accessible surface areas and interconnected frameworks that facilitate charge transport. Their synthesis is most commonly achieved through template-assisted routes and chemistry-guided strategies (Fig. 12). Beyond their structural advantages, metal oxides offer diverse compositions and rich surface chemistry. Their lattice oxygen species and variable oxidation states enable adsorption, activation, and transformation of reactant molecules through redox-mediated pathways. Metal oxides are broadly categorized as non-reducible (*e.g.*, Al_2O_3, WO_3), which maintain stable oxidation states, and reducible (*e.g.*, NiO, Fe_2O_3), which can readily exchange lattice oxygen with the environment. The latter are especially appealing for catalysis, as oxygen vacancies and dynamic surface reconstructions often enhance activity. Indeed, porous oxides such as WO_3, NiO, Fe_2O_3, TiO_2, Co_3O_4, CeO_2 [160–163], and more complex high-entropy alloys oxides [164] have been synthesized with tailored morphologies, crystalline phases, and pore architectures for electrocatalytic applications.

Despite this progress, achieving a balance between high activity, stability, and durability under practical operating conditions remains a formidable challenge. Most reported oxide-based catalysts still fall short of the performance metrics required for commercial deployment, particularly under acidic conditions. Moreover, a comprehensive and predictive understanding of the relationships among structure, composition, and catalytic performance remains underdeveloped, hindering the rational design of next-generation catalysts. These challenges are not unique to electrocatalysis. In photocatalytic systems, similar issues such as inefficient charge separation, rapid recombination, and limited stability under illumination also restrict practical implementation. Overcoming these bottlenecks requires integrated design principles that simultaneously optimize electronic structures, surface chemistry, and porous architectures.

10.3. Advances in science and technology to meet challenges

In recent years, three representative strategies have been widely explored to improve the electrolysis efficiency of porous metal oxide nanomaterials, and these approaches are common across different energy conversion reactions. The first is the incorporation of additional metals to reconstruct the electronic structure of active sites, thereby optimizing adsorption and desorption energetics. The second involves defect and interface engineering to regulate oxygen vacancies and create coordinatively unsaturated sites that facilitate reaction pathways. The third strategy leverages perovskite oxides, in which compositional tuning and lattice modification enable fine control over electronic configurations and oxygen mobility. Collectively, these strategies converge to lower energy barriers, optimize intermediate adsorption, and accelerate charge and mass transport, thus addressing the critical limitations of conventional non-noble metal catalysts.

A typical example of incorporation of additional metals was reported by Tian *et al.*, who synthesized nitrogen-doped porous carbon–anchored Mo_2C/CeO_2 nanoparticles using an electronic fuel injection method [165]. The synergistic effects among Mo_2C, CeO_2, and the conductive nitrogen-doped carbon framework enhanced intrinsic catalytic activity and promoted electron/proton transport, while overcoming the agglomeration and poor conductivity issues of pristine Mo_2C. In particular, Ce oxides not only provided empty or half-filled d orbitals to facilitate hydrogen adsorption but also employed its unique f orbitals to stabilize adsorbed H intermediates, thereby boosting HER activity. As a result, the Mo_2C/CeO_2 catalyst delivered a low overpotential of 220 mV at 10 mA/cm^2, a Tafel slope of 123 mV/dec, and excellent durability after 20 h of continuous operation and 2000 CV cycles.

Defect and interface engineering is exemplified by the work of Xia *et al.*, who employed a template-assisted strategy to construct hollow porous $Co_3O_4/CoMoO_4$ hybrids [166]. The deliberate creation of heterointerfaces, coupled with a hollow porous structure, significantly increased the density of active sites and facilitated charge and mass transfer. The optimized $Co_3O_4/CoMoO_4$ catalyst demonstrated outstanding OER performance under alkaline conditions, requiring only 342 mV to reach 100 mA/cm^2 with a Tafel slope of 72 mV/dec. Theoretical calculations confirmed that interface engineering effectively tuned the adsorption energies of intermediates, lowered the reaction barriers, and thereby enhanced catalytic activity.

The potential of perovskite oxides was highlighted by Xian *et al.*, who developed a microwave-shock synthesis method to produce two-dimensional porous $GdFeO_3$ perovskites with controlled A-site Sr substitution [167]. This one-step approach simultaneously enabled precise Sr incorporation and construction of a porous nanosheet structure, preventing agglomeration while promoting accessibility of active sites. The partial replacement of Gd^{3+} with Sr^{2+} modified the electronic configuration of $GdFeO_3$, raised the O 2p band center, and lowered the formation energy of oxygen vacancies, all of which contributed to enhanced intrinsic catalytic activity. The optimized $Gd_{0.8}Sr_{0.2}FeO_3$ catalyst achieved an overpotential of 294 mV at 10 mA/cm^2 and a small Tafel slope of 55.9 mV/dec in alkaline electrolytes.

Together, these examples illustrate how electronic structure reconstruction, interface defect engineering, and perovskite modification can be strategically applied to porous metal oxide catalysts, offering viable pathways to achieve high activity, durability, and scalability in electrochemical energy conversion. Importantly, these design philosophies are also directly translatable to photocatalytic systems. By tailoring band structures, defect chemistry, and interfacial charge dynamics, porous metal oxides can couple light absorption with catalytic redox reactions, bridging the

fields of photo- and electrocatalysis through shared mechanistic principles.

10.4. Porous metal oxide nanomaterials for photocatalysis

Beyond electrocatalysis, porous metal oxide nanomaterials have also demonstrated great promise in photocatalytic applications such as water splitting, CO_2 reduction, and pollutant degradation. Their intrinsic semiconductor nature enables efficient utilization of solar energy, while their porous architectures facilitate light harvesting, reactant diffusion, and product desorption. Representative porous oxides including TiO_2, ZnO, Fe_2O_3, WO_3, and $BiVO_4$ have been widely explored as photocatalysts due to their excellent stability, earth abundance, and environmental friendliness [168–170]. The high specific surface area and interconnected pore networks provide abundant reaction sites and promote charge separation by shortening carrier migration distances. Meanwhile, the presence of oxygen vacancies and defect states within the lattice introduces shallow donor levels, extending visible-light absorption and enhancing charge carrier mobility.

Beyond energy-oriented reactions, porous metal oxides play a critical role in environmental remediation through photocatalytic degradation of dyes, antibiotics, and VOCs. The porous frameworks enhance pollutant diffusion to active sites, while photoinduced charge carriers generate reactive species such as $^{\bullet}OH$, $^{\bullet}O_2^-$, and $^1O_2$ that drive mineralization processes. For instance, mesoporous TiO_2 and WO_3 aerogels exhibit superior performance in decomposing persistent organics under visible light, attributed to their large surface area, defect-induced charge separation, and adjustable band alignment. Coupling oxide photocatalysts with carbonaceous or plasmonic components (graphene, Au, Ag) further broadens the light absorption range and improves redox selectivity, enabling synergistic CO_2 reduction and organic pollutant removal in complex aqueous environments [171].

Recent progress in nanostructure control and heterojunction construction has significantly advanced the performance of oxide-based photocatalysts. For instance, hierarchical TiO_2 aerogels with mesoporous–macroporous dual structures exhibit enhanced photon capture and reduced electron–hole recombination through multi-scattering effects [172]. Coupling porous metal oxides with carbonaceous materials (such as graphene or carbon nitride) or other semiconductors (such as ZnO/COF) creates Z-scheme or S-scheme heterojunctions that facilitate directional charge transfer [173,174]. In addition, surface plasmon coupling with metal nanoparticles (*e.g.*, Ag, Au) can locally intensify the electromagnetic field and extend photoresponse into the visible and near-infrared regions.

In parallel, defect and doping engineering have emerged as efficient strategies to tune electronic band structures. Introducing transition metal dopants (*e.g.*, Fe, Co, or Cu) or nonmetal dopants (*e.g.*, N, S) can narrow the band gap, improve visible-light absorption, and optimize the position of the conduction and valence bands for redox reactions. Moreover, the confined microenvironments within porous architectures can stabilize reaction intermediates, regulate local pH, and suppress back reactions, thereby improving quantum efficiency and selectivity.

Overall, the integration of light-harvesting design, defect modulation, and heterostructure engineering offers powerful routes to exploit the full photocatalytic potential of porous metal oxides. Future research should focus on elucidating the dynamic coupling between photogenerated charge carriers, surface states, and reaction intermediates under *operando* conditions. The combination of *in-situ* spectroscopy and theoretical modeling will be essential to reveal structure-function correlations, guiding the rational design of next-generation porous oxide photocatalysts for solar fuel and environmental remediation applications.

10.5. Concluding remarks and prospects

Porous metal oxide nanomaterials have emerged as a versatile class of catalysts owing to their large surface areas, tunable electronic structures, and rich surface chemistry. Through strategies such as compositional modulation, interface defect engineering, and perovskite lattice tuning, their electrocatalytic and photocatalytic activities and stabilities have been markedly improved, enabling performance that begins to approach noble-metal benchmarks in key reactions including HER, OER, ORR, and solar-driven hydrogen evolution or CO_2 reduction. Looking forward, the confinement effect within porous architectures should be taken into consideration, as nanoscale confinement can reshape the local reaction environment, influence ion/charge transport, and stabilize short-lived intermediates, thereby impacting both electrochemical and photocatalytic kinetics. At the same time, compositional tuning, defect engineering, and heterojunction construction at oxide interfaces provide powerful levers to exquisitely regulate the behavior of reactants and photogenerated charge carriers within the active sites or the electrical double layer (EDL).

Since these electrochemical and photocatalytic processes share the fundamental principles of reactant adsorption, charge transfer, and intermediate stabilization, such interfacial modulation offers a pathway not only to improve individual reactions but also to reveal the common mechanistic principles underlying HER, OER, ORR, and light-driven redox transformations. Importantly, this mechanistic framework can be extended to emerging photocatalytic and photoelectrochemical transformations, such as CO_2 reduction, N_2RR, chlorine evolution (CER), urea oxidation (UOR), alcohol and biomass electrooxidation (AOR/BOR), H_2O_2 electrosynthesis *via* the $2e^-$ ORR, and solar-to-fuel conversions. Integrating insights from both electrocatalysis and photocatalysis will enable rational design of multifunctional porous oxide catalysts, offering pathways to high-efficiency energy conversion, environmental remediation, and decentralized chemical production. Such progress will play a pivotal role in enabling the broader energy transition and storage landscape, from green hydrogen to multiple high-value products and solar-driven chemical manufacturing.

11. Heterojunction material

Ramin Hassandoost, Alireza Khataee*

11.1. Status

The design and application of heterojunctions have become a foundational strategy to overcome the intrinsic limitations of single-component photocatalysts, such as rapid charge carrier recombination and poor light absorption [175,176]. The core principle involves creating an interface between two distinct materials, where energy band alignment generates an internal electric field that separates photo-generated electrons and holes, thereby enhancing photocatalytic efficiency [177–179]. This approach has evolved significantly, from bulk semiconductors to advanced nanoscale and 2D architectures, with a key advancement being the use of porous materials like MOFs and COFs [180]. With their high surface areas and tunable structures, these materials enable the precision engineering of charge transfer pathways [181]. The recent researches have also moved beyond simple Type II heterojunctions to more complex S-scheme and Z-scheme mechanisms designed to preserve the most energetic charge carriers for demanding catalytic reactions [182,183].

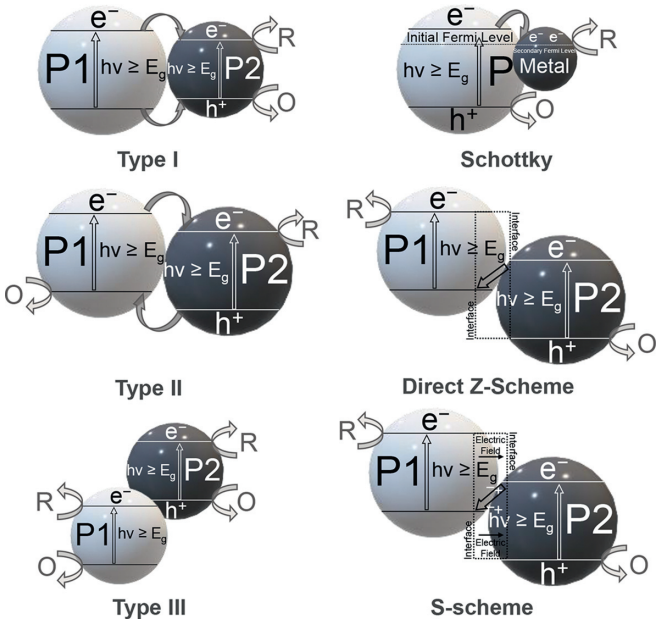


Fig. 13. Classification of heterojunctions and charge transfer mechanism (E_g: band gap, e^-: electron, h^+: hole, R: reduction, O: oxidation, and P: photocatalyst).

11.2. Current and future challenges

Despite having developed foundational concepts, the field faces critical challenges hindering widespread application. A primary weak point is low photocatalytic efficiency due to rapid electron-hole pair recombination and the limitations of specific mechanisms, such as the diminished redox potential of Type II heterojunctions for reactions requiring a larger driving force [184,185]. Synthesis remains difficult, as achieving "intimate and well-defined interfacial contact" is challenging, and current methods are often "lab-scale, low-yield" and lack precise control over morphology and defects [186,187]. Furthermore, long-term stability and resistance to photo-corrosion are key issues for porous materials, which can be susceptible to moisture and metal leaching [188,189]. A significant knowledge gap persists, as many advanced mechanisms like the Z-scheme and S-scheme are "largely theoretical" due to a lack of direct, time-resolved experimental evidence [190,191].

11.3. Advances to solve challenges

Researchers have developed advanced heterojunction types, refined synthesis methods, and new characterization techniques to address these challenges. The energy band gap is the most critical factor in heterojunction design (Fig. 13), which is used to classify them into three primary types and more advanced mechanisms. In Type I (Straddling Gap), the band gap of the smaller semiconductor is fully contained within the larger one. This alignment confines both electrons and holes, promoting recombination, and its primary purpose is carrier confinement. While this is disadvantageous for photocatalysis, it is useful for light-emitting devices like lasers. Type II (Staggered Gap) is the most common type for photocatalysis, with offset conduction and valence bands that promote charge separation by directing electrons and holes to different materials. The purpose in this mechanism is charge separation, which inhibits recombination but can reduce the system's overall redox potential [192]. A core/shell MIL-167/MIL-125-NH_2 composite, for example, demonstrated enhanced photocatalytic HER through charge migration from the conduction band of MIL-167 to the conduction band of MIL-125-NH_2 [193]. A 2D−2D SnS_2/TpPa-1-COF heterojunction also achieved a high HER rate due to its intimate interface contact and synergistic effect of the 2D layered structure and well-matched band positions [194]. In Type III (Broken Gap), the band gaps do not overlap, with the conduction band of one below the valence band of the other. This allows electrons to tunnel directly across the interface, with its primary purpose being interband tunneling. This property is engineered for devices like tunnel junctions in tandem solar cells [177].

Beyond these primary types, advanced junction concepts have been developed to describe charge transfer in more complex photocatalytic systems. A Schottky junction is a rectifying metal-semiconductor interface that enables a unilateral flow of majority carriers. The internal electric field at this interface enhances charge separation, with its primary purpose being charge separation and fast switching [195]. A Pt-loaded PY-DHBD-COF system, for example, achieved an excellent photocatalytic HER rate by exploiting this mechanism [196]. Inspired by photosynthesis, the direct Z-scheme is a charge transfer model that separates charges while preserving the strong redox potential of both photocatalysts [197]. This ensures that the most energetic electrons and holes are retained for reactions. For instance, the CdS nanorods/NMOF-Ni heterojunction operates *via* this mechanism and achieves a high HER rate [198]. The S-scheme is a more refined Z-scheme model that explicitly accounts for the internal electric field and band bending at the junction, which drives charge transfer and retains the most potent electrons and holes to maximize efficiency [199]. For instance, the N–COF/BiOBr S-scheme efficiently degraded tetracycline antibiotics using this mechanism [200]. A central principle of modern photocatalysis is that the unique properties of porous materials are critical enablers for next-generation heterojunctions. The "designability" of materials like COFs allows for precision engineering of the electronic and physical interface, directly controlling charge transfer efficiency [201]. A compelling case study is the UiO-66(Zr)-NH_2@MIL-88B (Fe) MOF-on-MOF heterostructure, which operates *via* a Z-scheme mechanism [202]. Synthesized through epitaxial face-selective growth, this design ensures intimate contact between the two MOF components, resulting in a synergistic effect that significantly boosts photocatalytic efficiency. This composite achieved a remarkable apparent quantum yield of approximately 0.9 % for water splitting under simulated sunlight. Another cutting-edge strategy involves time-resolved synthetic control of heterojunction formation, as demonstrated with a Pt@MUV-10 composite [203]. Pt nanoparticles were injected at different stages of the MOF's crystal growth, resulting in tunable heterojunctions with varying photocatalytic performance. Early injection of the nanoparticles led to their incorporation into the crystal's core, while a later injection restricted them to the outer surface. This precise control over the spatial distribution and electronic hybridization of the nanoparticles allowed researchers to boost photocatalytic H_2 production by up to 2.5 times, highlighting a new approach for tailoring catalyst performance.

Computational modeling, integrating force field molecular dynamics (MD), time-dependent density functional theory (TD-DFT), and non-equilibrium green function DFT (NEGF-DFT), was used to explore MOF/carbon nitride heterojunctions for dual CO_2RR and OER catalysis [204,205]. The DFT analysis on heterojunction cluster models offers a precise charge transfer mechanism. For a MOF/poly-heptazine imides (PHI) heterojunction, the projected density of states (PDOS) analysis initially showed a Type-II alignment. However, the study revealed that strategically incorporating Cu species in the MOF significantly destabilized the valence band edge, which shifted the band alignment to a Z-scheme. Similarly, doping the PHI with cations like K^+ was shown to boost charge transfer by confining holes on the PHI cores and delocalizing electrons on the MOF surface and demonstrated that the performance of photoreactors could be optimized not only by chemically and electronically tuning the components, but also by applying a neg-

ative bias to boost interfacial charge transfer further. A significant advancement is the shift from relying on indirect evidence to using direct, time-resolved evidence of charge transfer. Advanced techniques like Kelvin probe force microscopy and *in-situ* XPS can suggest direct proof of internal electric fields and electron migration, allowing researchers to move beyond speculation and gain a fundamental understanding of dynamic processes at the heterojunction interface [206,207].

11.4. Concluding remarks and prospects

In summary, the path of heterojunction photocatalysis has moved decisively from simple, bulk materials to sophisticated, multifunctional systems based on porous frameworks during the move from empirical methods to more deliberate and rational design using data-driven approaches. Looking ahead, the primary plan for the field must address the urgent need for scalability by moving beyond current batch processes to develop scalable, continuous-flow synthesis methods. The fundamental challenge of long-term stability and photo-corrosion must be addressed by engineering robust frameworks with irreversible bonds and intrinsic resistance to moisture. Ultimately, a deeper understanding of reaction mechanisms requires a unified approach integrating *in-situ* characterization with advanced computational modeling.

12. Design of advanced electrocatalysts based on aerogels

Claudio Imparato*, Aurelio Bifulco

12.1. Status

Electrocatalysis is flourishing in several sustainable energy conversion technologies, such as water splitting, fuels cells, and metal-air batteries, green production processes that exploit abundant resources, *e.g.*, CO_2 reduction to fuels and nitrogen reduction to ammonia, as well as the degradation of water pollutants [208–210]. While noble metals (chiefly platinum, palladium, iridium, rhodium) remain the main characters on the electrocatalysis stage, advances in the synthesis and engineering of less precious transition metals are boosting their wide-range applicability. Carbon-based (nano)materials, inorganic semiconductors (mainly metal oxides and metal chalcogenides), and emerging nanomaterials (*e.g.*, MOFs, COFs, MXenes) also play central roles in electrocatalytic processes. In the last years, research efforts have focused on the structure and morphology of electrocatalysts, identifying aerogels as an excellent basis for developments. First introduced in the 1930s, aerogels are materials characterized by ultra-low density (even below 1 mg/cm^3), due to the high void fraction (typically >90 %), very large specific surface areas, multi-scale porosity (from micro- to macropores) and tunable micro-/nanostructure [211].

Aerogels are employed either as a scaffold to support catalytic species or as the catalyst itself. Aerogel supports offer vast surface areas where isolated active sites can be dispersed, while monolithic aerogel catalysts take advantage of their porosity to maximize the diffusion of reactants and products and their surface interactions, avoiding the issues related to the dispersion and anchoring of the active species on the support [212]. Among the latter, metallic aerogels, introduced in 2009, are a steadily expanding family that bypass the problems related to the porous support, acting as self-standing catalytic layers in direct contact with the conductive electrode substrate. The hierarchical architecture and open pore system of metal aerogels increase the electrochemical active surface area compared to compact layers, facilitating mass transport within the bulk structure and providing plenty of available reaction sites. Also, abundant macropores accelerate the discharge of gas products, limiting the formation of bubbles that can clog active sites and impair the turnover frequency [213].

Conventional methods for the production of aerogels are based on the sol-gel technique. After gel formation and ageing (useful to advance polycondensation reactions, strengthening the solid structure), solvent extraction is the crucial step. Supercritical drying is usually regarded as the most effective route, freeze-drying avoids the issues due to liquid surface tension but involves high energy consumption, while ambient pressure drying requires less energy and simple equipment, but needs careful control on solvent exchange and additional operations, depending on the material, to prevent the collapse of pores. In addition to sol-gel, other strategies are gaining attention for the controlled fabrication of aerogels, including template-assisted, self-assembly and emulsion methods, 3D printing, electrospinning and hydrothermal process [213,214].

12.2. Current and future challenges

Electrocatalysts have several requirements concerning surface area, interconnected pore structures, electrical conductivity, stability, and durability in the operating environment (including wide ranges of potential and current density). The unique features of aerogels make them an effective way to meet many of these requirements. Carbon-based supports are widely employed, as they are inexpensive and versatile, but suffer from durability issues, particularly corrosion, also caused by the temporary high potentials reached during operation, for example when fuel cells start up or shutdown. Also, the binding of the active phase to the support is crucial because the detachment of nanoparticles during the process can cause loss of activity and other undesired effects [215].

Due to the variability in the colloidal chemistry of metals, innovative synthesis methodologies are set up to afford the desired compositions and structures, especially in alloys that aim to leverage the synergy of different metals. Semiconductor and metal nanocrystals (low-dimensional nanomaterials exhibiting intrinsic quantum confinement effects) have been unveiled as promising building blocks for versatile catalytic aerogels. Although the synthesis of nanoparticles is developing fast, the manipulation of the dimensionality, linkage, and arrangement of nano-building blocks, which determine the aerogels' functionality, remains challenging [216].

As alternatives to metallic aerogels, three-dimensional conductive aerogels have been constructed based on graphene, carbon nanotubes, conductive polymers or MXenes. Harnessing gelation routes for metals and emerging nanomaterials requires a combination of critical factors to be considered, *i.e.*, initiators, precursors, reductants, ligands, solvents, and external fields (*e.g.*, temperature, forces). The fabrication of new materials in the form of aerogels urge researchers to go beyond the well-established chemistry of sol-gel metal precursors and explore original, convenient strategies. Indeed, non-gelation pathways are also available for the processing of metal aerogels [211]. Nowadays, several complex synthesis methodologies are still in their infancy and do not ensure proper control on composition, microstructure, pore sizes or mechanical resistance of the products. Most published works adopt a "trial and error" approach, report only the outcomes of successful experiments, and often miss a scrutiny of the factors that led to valuable results. It appears clear that deeper interpretation of the links between experimental protocols and materials properties are essential.

Sustainability criteria must be taken into account in the development of novel materials. The platinum group metals (Ru, Os, Rh, Ir, Pd and Pt) feature among the 34 critical raw materials and the 16 strategic raw materials identified by the European Union in 2023 and their high cost pushes the search for effective alternatives [217]. Aerogels, with their low densities and high spe-

cific surface areas, can help reducing the mass of precious metal used in electrocatalytic systems. Nonetheless, the exploitation of more available and less costly transition metals (*e.g.*, Fe, Cu, Ni, W) is a current objective, especially for devices that necessitate large amounts of catalyst. Also, circular-by-design principles should be followed to allow an easy recovery and recycling of the elements at the end-of-life. Finally, once the researchers master the synthesis of optimal aerogel electrocatalysts, the scale-up of the procedures must be addressed to enable their transfer to market.

12.3. Advances in science and technology to meet challenges

Traditional sol-gel chemistry is employed to synthesize tailored porous metal oxides as supports for active metals. In such systems, the size and dispersion of metal species as well as their linkage and interface with the substrate play a crucial role. For example, undoped and F-doped SnO_2 aerogels were prepared by epoxide-initiated method, adding propylene oxide to $SnCl_4/SnF_2$ mixtures to control gelation and drying with supercritical CO_2 after solvent exchange [218]. Sputtering Pt on the SnO_2 matrix yielded a corrosion-resistant electrocatalyst which showed robust HER performance, with low overpotential (42 mV at 10 mA/cm^2) and high turnover frequency, attributed to the electronic metal-support interaction that boosted H atom adsorption and the activity of the dispersed Pt nanoclusters. An interesting case of oxide/metal composite that leverages the redox properties of ruthenium was presented by Yan *et al*., who tested a simple *in situ* reduction process using $NaBH_4$, followed by freeze drying, to obtain a partially oxidized 3D porous aerogel with a peculiar Ru/RuO_2 interface [219]. The optimized catalyst proved outstanding performance simultaneously in HER (surpassing the benchmark Pt/C and OER, better than bare RuO_2), with a low working voltage of about 1.47 V in both alkaline and acidic environment. Carbon nanomaterials have acquired large popularity also as aerogel supports. A 3D N-doped graphene aerogel, produced by a simple freeze-drying and annealing procedure, was decorated with Pt-Ni alloy particles and the resulting catalyst was tested in methanol and ethanol fuel oxidation [214]. The enhanced performances and stability compared to control samples were related to the surface area, favoured mass transfer and defects on the graphene support combined with oxophilic Ni species.

A variety of methodologies have been proposed in the last years to produce metal-based aerogels with modulated nanostructure and porosity, starting from low-dimensional nanomaterials. Tuning nano-building blocks, either 0D (nanoparticles), 1D (nanowires/nanotubes) or 2D (nanosheets) and then the interaction with the initiator and ligands, allows diverse compositions, ligament size and architectures to be realized. The use of colloidal nanoparticles as building blocks was deeply investigated by Niederberger's group, which demonstrated effective strategies to turn nanoparticles into aerogels, founded on the finely controlled destabilization of the dispersion by varying the concentration and driving the kinetics of cluster aggregation [220]. Salt-mediated solution synthesis is a versatile tool to prepare noble metal aerogels: detailed protocols have recently been published, including the assembly of metal nanostructures into hydrogels through interactions controlled by salts, and successive freeze-drying [221]. Expanding the scope of freeze-mediated assembly strategies, Du's research group demonstrated a universal ice-mediated confining assembly method, based on ice chemistry, to produce various metal aerogels with tailored microdimensions (0D, 1D, and 2D) [216]. In the one-pot procedures, assembly is driven by varying temperature and precursor concentration and thus controlling ice crystals growth rate (Figs. 14a and b). Among the mono- and multimetallic aerogels, 1D samples were found to be the most advantageous in both H_2 evolution and ethanol oxidation electrocatalysis, owing to the enhanced mass transfer deriving from their broad pores. Another recent work focused on the effect of multimetallic interactions in the sol-gel processing of aerogels, revealing how auxiliary metals influence the sedimentation rate and ligament size of the product, in relation with the bulk density and their atomic radius [222]. Hence, Au-Pt gel-based electrocatalyst films were prepared, which afforded manyfold increased performances in alcohol oxidation reactions compared to Pt/C benchmark.

Multimetallic electrocatalysts offers a series of advantages over single metal ones: (1) The synergistic effects include the modulation of electronic structure, tuning the D-band center and adapting the system to oxidation or reduction reactions with enhanced selectivity; (2) More active sites are generated and the adsorption energies of reactants or intermediates can be lowered; (3) Catalyst stability is often improved (*e.g.*, preventing CO poisoning in fuel cells); (4) Moreover, noble metals can be partly replaced with less precious ones, with cost benefits. In this context, effective synthesis routes are needed to obtain homogeneous alloys with the desired features. A stamping method involving phase–boundary gelation, without elaborated drying methods, was developed by Eychmüller and colleagues, and used to grow a 2D Pt-Ni bimetallic aerogel at the interface of a biphasic liquid mixture [223]. They compared its structure and catalytic properties with those of a 3D counterpart and found that the 2D version offers superior performance in methanol oxidation reaction (MOR). Specifically, the 2D aerogel exhibited a high electrochemically active surface area ($\sim$50.6 m^2/g) and a mass activity of $\sim$1.8 A/mg for MOR, both better than those of the 3D Pt-Ni aerogel, and a lower Tafel slope (110 *vs.* 145 mV/dec for the 3D version), indicating more favourable reaction kinetics. The improved performance was partly attributed to a more oxidized Ni surface in the 2D network, which may aid methanol oxidation by supplying active oxygenated species. These results showed that structurally engineered 2D aerogels can outperform their 3D analogues in electrocatalysis, offering an efficient route to lower–cost and high–activity catalyst designs. Moving to all-noble-metal aerogels, a rapid ultrasound-assisted method was applied to combine ultrasonic cavitation with $NaBH_4$ reduction, achieving gelation in few seconds and across a wide precursor concentration range (*i.e.*, 0.02–62.5 mmol/L) [224]. The method enabled the formation of single-metal (Au, Ag, Ru, Rh, Pt, Pd) and alloy (bi-/tri-metal) aerogels with ligament sizes $\leq$10 nm. These aerogels exhibit a porous, network-like structure that enhances surface area, charge transport, and gas diffusion. Electrocatalytic tests showed excellent performance in HER and ethanol oxidation reaction, demonstrating that this fast, versatile synthesis process offers highly efficient electrocatalysts with practical scalability.

Strain engineering is recognized as an efficient strategy for promoting the activity of electrocatalysts. Wei *et al*. reported that Ru-Au bimetallic aerogels, synthesized by a one–step *in situ* reduction–gelation method, exhibit superior HER performance in alkaline medium, due to built–in strain effects and charge redistribution [225]. The optimized composition ($RuAu_3$) achieved a very low overpotential of 24.1 mV at 10 mA/cm^2 and a Tafel slope of 36.7 mV/dec, with respect to commercial Pt/C under the same conditions. The strain arises from lattice mismatch (tensile strain on Ru, compressive on Au), which shifts the d–band center downward and tunes the adsorption of key intermediates (water, OH*, H*) to enhance kinetics. The catalyst also showed good stability of structure and performance (only $\sim$9 mV degradation in overpotential over 12 h) after prolonged cycling. Studies on other applications, combining environmental and sustainable production purposes, were recently reported. For instance, Pd-Ag aerogels, synthesized *via* two-step salting-out and cross-linking route, were tested in the selective electrocatalytic upcycling of PET [226]. The subsequent $NaBH_4$ reduction, NH_4F addition, flash freezing in liquid nitrogen and freeze drying (Fig. 14c) yielded ultrathin nanowires

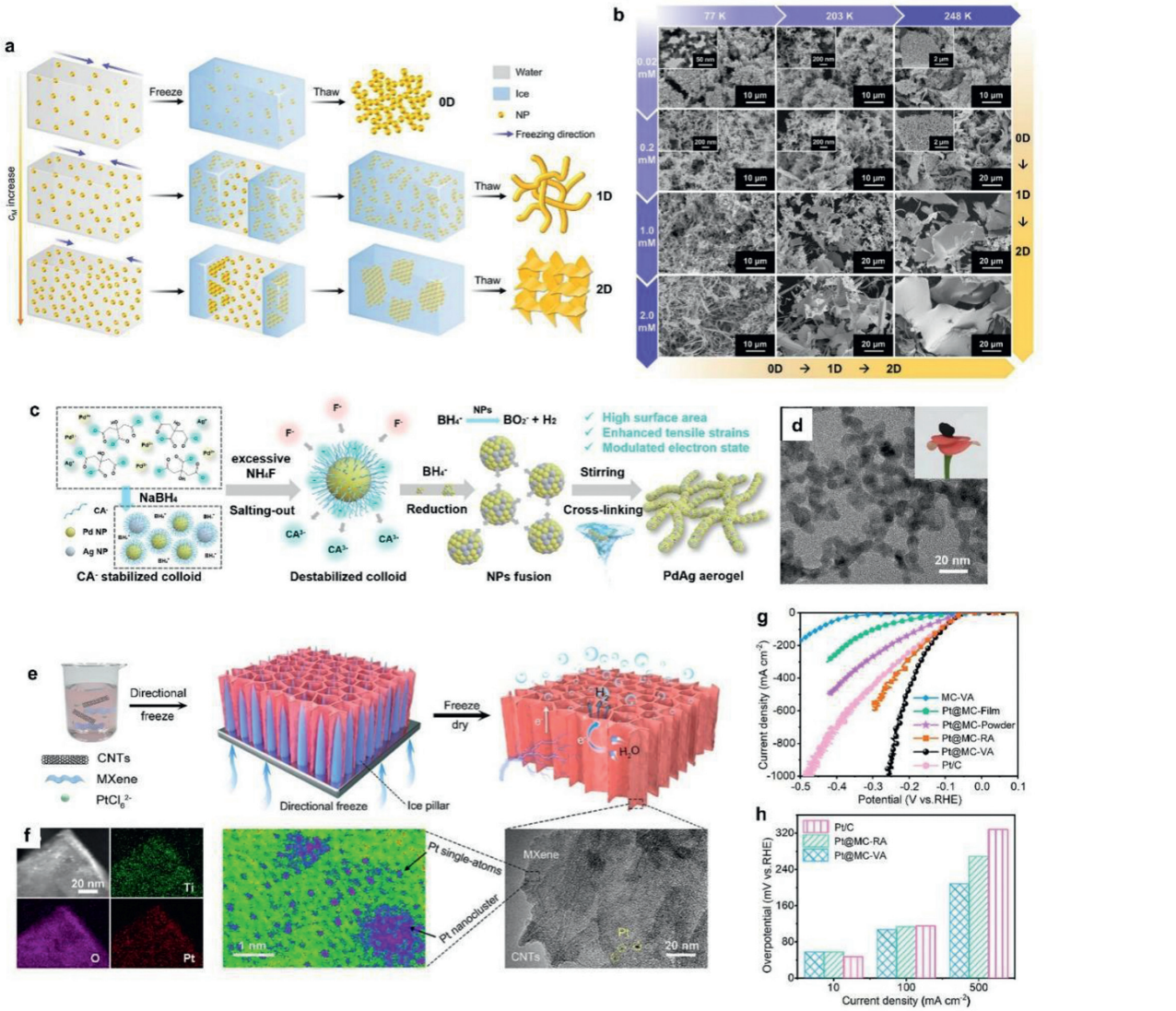


Fig. 14. (a) Mechanism proposed for the ice-mediated fabrication of Au aerogels with microdimension control. (b) Electron microscopy images of Au aerogels produced at different colloid concentration and freeze temperature. Reproduced with permission [216]. Copyright 2025, Wiley. (c) Schematic illustration of the preparation of $Pd_{67}Ag_{33}$ aerogel. (d) TEM images (inset: photograph) of $Pd_{67}Ag_{33}$ aerogel. Reproduced with permission [226]. Copyright 2024, National Academy of Sciences. (e) Schematic diagram of the fabrication by oriented temperature gradient of vertically aligned Pt/MXene/carbon nanotubes (Pt@MC-VA) aerogels. (f) Elemental mapping and electron microscopy images of Pt@MC-VA. (g) HER performances and (h) HER overpotential of the Pt@MC-VA heterostructure compared to reference samples. Reproduced with permission [231]. Copyright 2024, Elsevier.

with significant specific surface area (89.8 m^2/g) and pore volume (0.285 g/cm^3) for the $Pd_{67}Ag_{33}$ sample (Fig. 14d). This latter showed the best activity in the oxidation of ethylene glycol (derived from PET pretreatment) to glycolic acid, also coupled with CO_2 reduction, achieving Faradaic efficiency (FE) >80% for both electrodes. The outcomes were ascribed to the contribution of strain and ligand effects.

Multimetallic alloys offer advantages in terms of synergism and flexibility of properties and functions, however as the number of components increases so does the number of variables, complicating a thorough assessment of the influence of composition. Among metal alloys, medium-entropy and high-entropy metallic structures are gaining interest. Han *et al.* presented a general synthetic strategy to fabricate medium–entropy alloy aerogels (MEAAs), with ultralight porous 3D structures, able to overcome the usual difficulty of achieving homogeneous single–phase multimetallic alloys due to the immiscibility of elements [227]. By mixing metal precursors at the ionic level, they enable short–range diffusion of different metal atoms during a combined auto–combustion and low–temperature reduction process, driving the formation of a uniform alloy phase within the aerogel matrix. The resulting MEAAs achieve a very low density (39.3 mg/cm^3) and high porosity. As a proof-of-concept, a $Ni_{50}Co_{15}Fe_{30}Cu_5$ MEAA showed exceptional electrocatalytic activity for the MOR, with a mass activity of ~1.62 A/mg and a specific activity of ~132 mA/cm^2, outperforming simpler (lower–entropy) counterparts. In addition, the MEAA was integrated into a methanol-oxidation-assisted water electrolyser, achieving a low cell voltage of 1.476 V at 10 mA/cm^2 (~173 mV lower than conventional alkaline water electrolysers) and producing formate at the anode. Thus, the control of diffusion distances at the atomic level is a viable route to synthesize lightweight, highly active, multimetallic alloy aerogels. High-entropy metallic structures are valuable materials for electrocatalytic carbon dioxide reduction, as demonstrated by Li and coworkers, who developed a class of robust multi-metal high entropy alloy aerogels (HEAAs) using a freeze-thaw method [228]. Among the results, the PdCuAuAgBiIn HEAA achieved nearly 100% FE for C_1 products (*e.g.*, HCOOH) in the potential range from −0.7 V to −1.1 V *vs.* RHE. In a flow cell,

they obtained high current densities (~200 mA/cm^2) with ~87 % FE for formic acid. The enhanced performance is attributed to strong synergistic interactions among the multiple metal elements, which modify electronic structures, create abundant unsaturated surface sites, and optimize adsorption/desorption energies for the key HCOO* intermediate. HEAAs are promising new catalytic platforms for CO_2 reduction, combining the benefits of high-entropy alloys and 3D aerogel architectures.

Bifunctional electrocatalysts can be employed for a range of applications, including rechargeable metal-air batteries. In this scope, Zhang *et al.* integrated vertically aligned cobalt–manganese layered double hydroxide (CoMn-LDH) nanosheets onto a N,P co-doped graphene aerogel (NPGA) [229]. This 3D porous, free-standing monolith was synthesized through cross-linking gelation, hydrothermal self-assembly, and freeze-drying. The resulting CoMn-LDH/NPGA composite exhibited excellent catalytic activity for both ORR and OER, achieving a small potential gap of ~0.72 V, comparable to commercial Pt/C + IrO_2 catalysts. Its high performance arises from a large electrochemical active surface area, fast mass and charge transport, full exposure of active sites, and synergistic effects between doped graphene and LDH nanosheets. When used as cathode in a rechargeable liquid zinc-air battery, the catalyst showed strong cycling stability and superior bifunctional activity compared to many transition metal alternatives. This study demonstrates a promising, low-cost approach to replacing noble metals in advanced metal–air battery technologies.

Heterostructures conjugating noble metals with conductive nanomaterials, in synergy with engineered strain and ligand effects at heterointerfaces, can boost the performances of precious metal catalysts. For instance, 3D porous aerogels (Pt@$Ti_3C_2T_x$-rGO) were obtained by γ–radiolysis induced self–assembly of a 2D-2D heterostructure combining MXene ($Ti_3C_2T_x$) and reduced graphene oxide (rGO), with low loadings of anchored Pt nanoparticles [230]. This architecture helps avoid restacking of $Ti_3C_2T_x$ sheets, improves metal-support interaction, and enables enhanced exposure to electrocatalytic reactants. The resulting catalyst exhibits excellent electrocatalytic performance: HER overpotential of 43 mV at 10 mA/cm^2, OER overpotential of 490 mV at 10 mA/cm^2, and strong ORR activity (onset potential 0.957 V), along with outstanding stability under acidic conditions. Spectroscopic and electronic analysis show that due to the strong metal–support interaction, charge transfer from support to Pt and compressive strain on the Pt lattice shift its d–band center downward, improving catalytic activity. Another interesting case is the aerogel proposed by Zhang *et al.*, based on a 3D structure comprising vertically aligned MXene-carbon nanotube (CNT) hybrid scaffold decorated with tiny Pt nanoclusters [231]. Such architecture was fabricated through directional freezing on a copper substrate of a mixture of $Ti_3C_2T_x$ MXene, CNTs and H_2PtCl_6, followed by freeze-drying (Figs. 14e and f). The resulting electrode exhibited excellent HER performance, including low overpotentials (Figs. 14g and h), favourable kinetics (low Tafel slopes), and high durability. Specifically, it could reach low overpotentials of 208 and 249 mV to give current densities of 500 and 1000 mA/cm^2, respectively, keeping a satisfactory stability for at least 24 h. The vertical alignment and intimate contact among Pt, MXene, and CNTs contribute to maximizing active site exposure and accessibility, improving electronic and ionic conductivity, and facilitating mass transfer. These outcomes demonstrate a promising approach to achieving high–efficiency HER electrocatalysts with reduced Pt usage by leveraging hierarchical conductive supports.

Among the latest trends in the field, the integration of single-atom catalysts into aerogel has led to the novel class of "atomic aerogel materials" (or single-atom aerogels), which combine the porosity of aerogels with the high catalytic efficiency of single-atom catalysts (near 100 % atom utilization). According to their basic units, two categories can be distinguished, namely carrier-level (single atoms dispersed on aerogel networks with micro- or nanopore structures) and atomic-level aerogels (composed of atomically dispersed units with sub-nanopore structures) [151]. Such materials can be synthesized by different methods, including sol–gel processing and freeze-drying, for achieving uniform atomic dispersion and structural integrity, addressing challenges like atom aggregation and thermal instability. Atomic aerogel systems promise superior performances in electrocatalysis (*e.g.*, ORR, OER, CO_2 reduction), energy storage, and sensing, due to enhanced active site accessibility, rapid mass transport, and tailored electron pathways [151].

12.4. Concluding remarks and prospects

Aerogel electrocatalysts (particularly metal aerogels) are bringing about significant progress in hydrogen generation by water splitting, oxygen reduction and alcohol oxidation in fuel cells, CO_2 conversion and other relevant processes. Along with metal aerogels, especially multimetallic ones, those based on 2D nanomaterials, in particular graphene and MXenes, show excellent performances and hold great promise for further improvements. Lately devised synthetic strategies, such as ice-template, ultrasound-assisted or phase boundary gelation, allow the construction of advanced structures, with excellent dispersion of active species and strong metal-support interaction, or optimal mixing and interfacial contact between different components.

Notwithstanding many recent encouraging results, the field of aerogel electrocatalysts appears to be still largely unexplored and rich in opportunities for practical advancements. The combination of fundamental understanding and technological insight is necessary to promote the production of aerogels and their application in efficient and stable electrocatalytic systems. An optimized aerogel product needs a strict control over crucial characteristics, like well-defined structures across different length scales, dispersion of components at the nanoscale, tailored crystal facets and mechanical resistance. Original synthetic approaches require insight into the involved mechanisms in order to generalize the findings and establish reliable and reproducible protocols. The rational design of novel materials should benefit from the previous research efforts, therefore the significance of unsuccessful experiments should be reevaluated, and the critical interpretation of successful approaches encouraged, so to help other researchers endeavouring to develop advanced aerogel systems.

Different electrocatalyzed reactions need specific materials' properties, thus precisely recognizing structure-properties-activity relationships is a key for attaining optimized catalysts. On the other hand, as described above, some aerogels, particularly multicomponent ones, can be adaptable and well-performing in various processes, expanding their applicability [231]. Artificial intelligence tool can be very effective in supporting scientists during the development of new aerogel-based electrocatalysts, especially for the prediction of their features or operational conditions. To give an example, Ram *et al.* recently presented a computational strategy combining DFT and ML to design and screen advanced electrocatalysts for OER, HER and ORR [232]. Integrating DFT-calculated descriptors (such as adsorption energies, electronic structure features, binding energies) with regression or classification ML models, efficiently predicted catalyst performance across multiple reactions. The approach enables high-throughput screening of candidate materials and helps identify key descriptors (features) that correlate strongly with catalytic activity for each reaction. The study highlights that multifunctional catalysts can be rationally designed through descriptor–based ML filtering, reducing the computational cost compared to brute–force DFT of all candidates. DFT and ML can synergically work to accelerate the discov-

ery of high–performance electrocatalysts for energy conversion. To properly function, ML tools need huge input datasets that are often incomplete, due to missing materials' parameters available in the literature. These limitations can be overcome by data augmentation strategies or generative ML tools, which allow, for example, the accelerated design of conductive MXene aerogels with programmable properties [154].

As efficient synthetic routes are discovered and validated, the pathway towards industrialization should be foreseen. This implies the consideration of the technical and economic aspects that may hinder the transition from lab-scale to industrial scale production, which are frequently overlooked in research articles, although the attention to high-throughput procedures is growing. It is expected that cooperative efforts by chemists, physicists, materials scientists and chemical engineers will pave the way to new efficient and sustainable aerogel-based electrocatalytic processes.

13. Integrated porous materials roadmap for energy and environmental applications toward 2030

Kezhen Qi, Lan Ding*

13.1. System-level integration of porous materials

Porous materials, including MOFs, COFs, aerogels, carbon frameworks, and metal oxides, represent a diverse yet interconnected family of functional solids that share key design principles such as controllable porosity, tunable electronic structures, and high surface reactivity. Although each category has been independently optimized for specific functions such as supercapacitors, photocatalysis, or pollutant removal, a unified vision integrating these materials within a coherent design framework for multifunctional applications has been limited.

Recent advances demonstrate that synergistic integration across porous systems can unlock functionalities inaccessible to single-component materials. For example, MOF-derived carbons or metal phosphides provide high conductivity for electrocatalysis; COFs contribute robust π-conjugated backbones for photoactive charge transfer; aerogels supply hierarchical macro–mesoporous scaffolds for mass diffusion and mechanical stability. When rationally combined, these materials can simultaneously achieve fast electron/ion transport, efficient charge separation, and mechanically stable catalytic interfaces, bridging the gap between energy conversion, storage, and environmental remediation.

Moreover, different energy storage devices impose distinct performance requirements on porous materials. High-energy-density devices, such as lithium-ion or sodium-ion batteries, require dense redox-active sites, robust frameworks, and ion-accessible architectures to ensure high capacity and long cycle life. High-power-density devices, such as supercapacitors, prioritize ultrafast ion transport, high electronic conductivity, and hierarchical porosity to support rapid charge-discharge cycles. These considerations highlight the need for a materials-by-design strategy across MOFs, COFs, carbon frameworks, and metal oxides, enabling rational tuning of pore structures, surface chemistry, heteroatom doping, and hybridization with conductive matrices to meet specific device demands, coordinating energy and power density while maintaining multifunctionality. Hence, the roadmap toward 2030 calls for a paradigm shift from material-specific optimization to system-level integration and multifunctional design, aiming to realize sustainable and versatile porous materials platforms.

13.2. Challenges and emerging strategies

Despite the shared design logic, several barriers impede effective integration and scalability:

(1) Incompatible synthesis windows. MOFs and COFs typically require mild solvothermal conditions, while aerogels and metal oxides often demand high-temperature treatment. Achieving structural coherence in hybrid systems remains a synthetic challenge.
(2) Interface mismatching. Distinct surface chemistries and electronic properties can cause poor interfacial contact, leading to charge recombination and mechanical instability.
(3) Limited scalability and environmental sustainability. Solvent-intensive synthesis and costly organic linkers hinder large-scale deployment, contradicting the goal of sustainable chemistry.
(4) Insufficient cross-disciplinary modeling. Most theoretical models focus on isolated systems; data-driven approaches capable of correlating structure, stability, and catalytic/energy-storage performance across materials classes are still underdeveloped.

Addressing these challenges requires a system-level strategy encompassing hierarchical assembly, interface engineering, data-driven design, green synthesis, and functional coupling. Hierarchical assembly and compositional hybridization, such as MOF–COF heterostructures or MOF-derived oxides embedded in carbon aerogels, enable simultaneous electrocatalytic, photocatalytic, and energy storage functionalities. Interface engineering guided by *operando* characterization ensures optimized energy-level alignment and chemical bonding, improving charge separation, stability, and device performance. Data-driven discovery using machine learning and high-throughput computation can unify structural and functional descriptors across materials, enabling predictive optimization for targeted applications. Green and scalable syntheses, including mechanochemistry, supercritical drying, and biomimetic gelation, reduce environmental impact and improve manufacturability. Functional coupling strategies allow simultaneous pollutant degradation and energy generation, such as photocatalytic hydrogen evolution from wastewater or CO_2 conversion under visible light. These strategies also enable rational tailoring of porous materials for diverse energy storage devices, ensuring integrated systems meet both high-energy and high-power demands without compromising multifunctionality.

A series of emerging strategies are paving the way toward integrated porous materials design for 2030:

(1) Hierarchical assembly and compositional hybridization: Combining multiple porous frameworks yields synergistic properties. MOF-COF heterostructures integrate metal coordination sites with π-conjugated backbones, promoting both redox and photoactive reactions. Embedding MOF-derived metal oxides within carbon aerogels results in mechanically robust and conductive composites for bifunctional electrocatalysis and energy storage.
(2) Interface engineering guided by *in-situ* characterization: *Operando* X-ray absorption spectroscopy, TEM, and time-resolved photoluminescence allow real-time tracking of charge dynamics at heterointerfaces. These insights enable precise tuning of energy-level alignment and chemical bonding across MOF/COF-aerogel-oxide junctions, optimizing electron pathways, reaction selectivity, and multifunctional performance.
(3) Data-driven materials discovery: High-throughput computational screening and machine learning models are beginning to integrate the structural databases of MOFs, COFs, and porous oxides to predict the optimal combinations for specific catalytic, adsorption, or energy storage functions.
(4) Green and scalable synthesis: Solvent-free mechanochemistry, supercritical drying, and biomimetic gelation reduce synthesis costs and environmental impact, aligning porous materials production with green chemistry principles.
(5) Functional coupling for energy and environmental convergence: Hybrid materials capable of coupling pollutant degradation

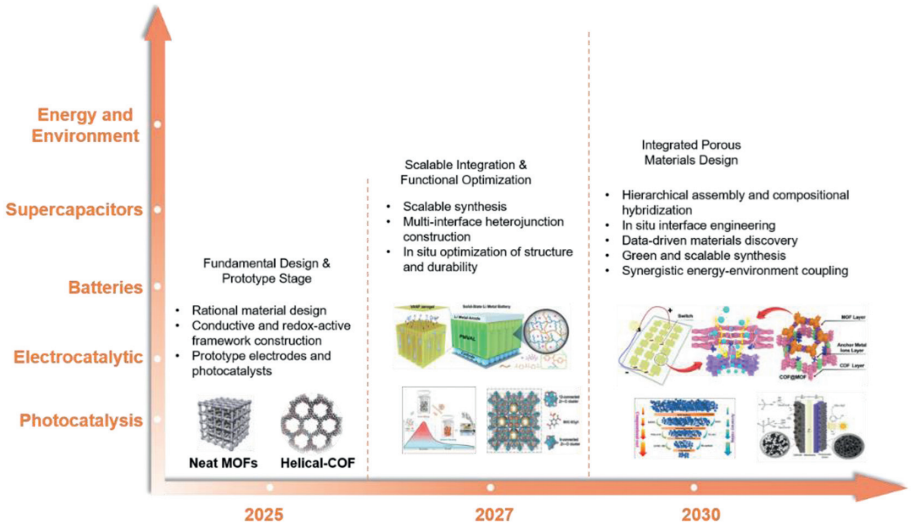


Fig. 15. 2030 Roadmap on pore materials for energy and environmental applications.

with energy generation, such as photocatalytic hydrogen evolution from wastewater or CO_2 conversion under visible light, illustrate the potential of unified porous platforms for circular carbon and water cycles.

13.3. Roadmap and outlook toward 2030

To provide a more intuitive overview of the development trends of various porous materials in energy and environmental applications, we present a schematic roadmap in Fig. 15. It covers MOFs, COFs, aerogels, carbon-based frameworks, and porous metal oxides, with key technological targets and research priorities projected for 2025, 2027, and 2030. The roadmap highlights critical milestones in energy storage, electrocatalysis, photocatalysis, and environmental remediation, while revealing the integrated evolution and synergistic potential of these materials, providing a strategic framework to guide future research directions.

The roadmap toward 2030 envisions porous materials not as isolated entities but as interconnected elements within a unified scientific ecosystem. Through data-driven design, hierarchical integration, and sustainable synthesis, the collective strengths of MOFs, COFs, aerogels, carbon frameworks, and metal oxides can be synergistically harnessed. By converging energy conversion, storage, and environmental remediation, and by rationally balancing device-specific energy and power requirements, the next decade will see the rise of integrated porous materials platforms that not only enhance performance but also embody the principles of green chemistry and sustainable development. Such a roadmap provides a concrete, strategic trajectory toward 2030, guiding both academic research and industrial implementation of multifunctional porous materials.

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CRediT authorship contribution statement

Kezhen Qi: Writing – review & editing. **Lan Ding:** Writing – original draft. **Pitcheri Rosaiah:** Writing – original draft. **Zhipeng Yu:** Writing – original draft. **Sofia Tikhanova:** Writing – original draft. **Vadim Popkov:** Writing – original draft. **Ahmed Ismail:** Writing – original draft. **Hui Dou:** Writing – original draft. **Derong Luo:** Writing – original draft. **Feng Liu:** Writing – original draft. **Yixue Xu:** Writing – original draft. **Shun-Qi Xu:** Writing – original draft. **Chunyang Dong:** Writing – original draft. **Ramin Hassandoost:** Writing – original draft. **Alireza Khataee:** Writing – original draft. **Ruiyang Zhang:** Writing – original draft. **Ying Zhou:** Writing – original draft. **Zijun Huang:** Writing – original draft. **Yongming Luo:** Writing – original draft. **Dedong He:** Writing – original draft. **Yunyun Ma:** Writing – original draft. **Zhuo Xing:** Writing – original draft. **Claudio Imparato:** Writing – original draft. **Aurelio Bifulco:** Writing – original draft.

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