

Review

Synergistic Innovations: Organometallic Frameworks on Graphene Oxide for Sustainable Eco-Energy Solutions

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Abstract: Combining organometallic frameworks with graphene oxide presents a fresh strategy to enhance the electrochemical capabilities of supercapacitors, contributing to the advancement of sustainable energy solutions. Continued refinement of materials and device design holds promise for broader applications across energy storage and conversion systems. This featured application underscores the inventive utilization of organometallic frameworks on graphene oxide, shedding light on the creation of superior energy storage devices for eco-friendly solutions. This review article delves into the synergistic advancements resulting from the fusion of organometallic frameworks with graphene oxide, offering a thorough exploration of their utility in sustainable eco-energy solutions. This review encompasses various facets, including synthesis methodologies, amplified catalytic performances, and structural elucidations. Through collaborative efforts, notable progressions in photocatalysis, photovoltaics, and energy storage are showcased, illustrating the transformative potential of these hybrids in reshaping solar energy conversion and storage technologies. Moreover, the environmentally conscious features of organometallic–graphene oxide hybrids are underscored through their contributions to environmental remediation, addressing challenges in pollutant elimination, water purification, and air quality enhancement. The intricate structural characteristics of these hybrids are expounded upon to highlight their role in tailoring material properties for specific eco-energy applications. Despite promising advancements, challenges such as scalability and stability are candidly addressed, offering a pragmatic view of the current research landscape. The manuscript concludes by providing insights into prospective research avenues, guiding the scientific community towards surmounting hurdles and fully leveraging the potential of organometallic–graphene oxide hybrids for a sustainable and energy-efficient future.

Keywords: sustainable eco-energy; photocatalysis; environmental remediation; solar energy conversion; eco-friendly materials; nanocomposites; carbon-based materials



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1. Introduction

Graphene oxide (GO) is a derivative of graphene, a single layer of carbon atoms arranged in a hexagonal lattice. GO is characterized by the presence of oxygen-containing functional groups, such as epoxides, hydroxyls, and carboxyls, which are introduced during its synthesis. Several methods are employed to produce GO [1,2]. Chemical exfoliation

involves oxidizing graphite to produce monolayer or few-layer GO sheets [3]. Other methods include the Hummers method, Staudenmaier method, Hofmann method, and modified versions involving oxidation and reduction steps [4–8]. GO retains the remarkable mechanical strength of graphene, albeit with a reduction in conductivity due to the presence of oxygen functional groups. This is why GO is typically reduced to reduced graphene oxide (rGO) to enhance its conductivity for practical applications [9]. The balance between strength and electrical properties makes it suitable for various applications. The functional groups on GO make it highly reactive. This reactivity allows for easy functionalization and interaction with different molecules, expanding its applicability in various fields. GO is used in electronic devices, such as transistors and sensors, owing to its electrical properties and compatibility with existing semiconductor technologies [10,11]. In materials science, GO is employed to enhance the mechanical and thermal properties of composites, making them suitable for applications in structural materials [12].

Organometallic frameworks (OMFs), also known as metal–organic frameworks (MOFs), are coordination polymers distinguished by metal ions linked together by organic ligands [13,14].

These intricate three-dimensional structures often manifest porosity and modifiable properties, rendering them highly versatile. OMF synthesis involves self-assembly processes, wherein metal ions and organic ligands spontaneously assemble into extended networks. The selection of metal ions and ligands enables tailored adjustments to the framework's characteristics [15]. Notably, OMFs exhibit notable porosity and surface area, rendering them well-suited for applications such as gas storage and separation [14].

The versatility of OMFs extends further through the capacity for property tuning via specific metal ion and ligand selection, allowing for customization to fulfil diverse application requirements [16]. Leveraging their porous nature and selective adsorption capabilities, OMFs find widespread utility in gas storage, separation, and water purification endeavors [17,18]. Moreover, these frameworks serve as efficient catalysts across various chemical reactions, capitalizing on the synergistic interplay between metal centers and organic ligands [19,20].

Organometallic frameworks inherently possess catalytic properties, and their amalgamation with GO amplifies catalytic activity through synergistic interactions between metal centers and graphene [21]. With its elevated surface area and distinctive electronic properties, GO serves as an exceptional support material for anchoring organometallic catalysts, facilitating efficient catalysis in eco-energy applications [22]. Notably, GO offers a stable platform for immobilizing organometallic frameworks, averting their agglomeration and ensuring sustained catalytic activity [23,24]. The robust nature of GO further enhances the overall stability and durability of the organometallic–graphene oxide hybrid, rendering it suitable for diverse eco-energy applications under challenging conditions [25,26].

Moreover, the high electrical conductivity of rGO expedites efficient electron transport within the hybrid structure, promoting rapid charge transfer during electrochemical processes in energy applications [27,28]. The integration of organometallic frameworks with GO engenders a synergistic effect, optimizing charge transfer pathways and enhancing overall performance in eco-energy devices [27,28]. Additionally, GO's functional groups offer ample opportunities for surface modifications, enabling precise tuning of the hybrid material's surface chemistry to meet specific requirements in eco-energy applications [29].

Furthermore, the selection of organic ligands in organometallic frameworks can be tailored to complement the functional groups on GO, enhancing compatibility and fostering targeted interactions. The resulting hybrid material exhibits improved light absorption and efficient charge separation, rendering it well suited for solar energy harvesting applications, including photovoltaics and solar fuel generation [30,31]. Moreover, organometallic–GO hybrids demonstrate promising performance in energy storage devices (e.g., batteries and supercapacitors) and conversion systems (e.g., fuel cells), thereby contributing to sustainable eco-energy solutions [32].

By aligning with the principles of environmental sustainability, the integration of an OMF onto GO provides eco-friendly alternatives for energy production and storage [33]. Furthermore, these hybrid materials can be designed for recyclability and reusability, minimizing waste and contributing to a circular economy in eco-energy technologies [34]. In elucidating the rationale for integrating an OMF with GO in eco-energy applications, this review furnishes a comprehensive understanding of the synergies and advantages that emerge from combining these materials for sustainable energy solutions.

The formation of OMF/GO composites is a complex process influenced by various factors, with the nucleation and growth of OMF structures being paramount among them. However, these crucial aspects have frequently been disregarded in previous studies [35]. Consequently, as depicted in Figure 1, it becomes imperative to delve deeper into the underlying mechanisms governing the formation of these composites to fully understand their properties and potential applications. Additionally, considerations such as precursor concentration, reaction conditions, solvent choice, and the presence of functional groups on GO can significantly impact the composite's structure and performance. Furthermore, the interplay between the OMF and GO at the interface, including interactions such as π - π stacking and hydrogen bonding, plays a crucial role in determining the final composite morphology and properties. Thus, a comprehensive examination of these factors is essential for advancing our knowledge and optimizing the synthesis of OMF/GO composites for diverse applications ranging from catalysis to energy storage.

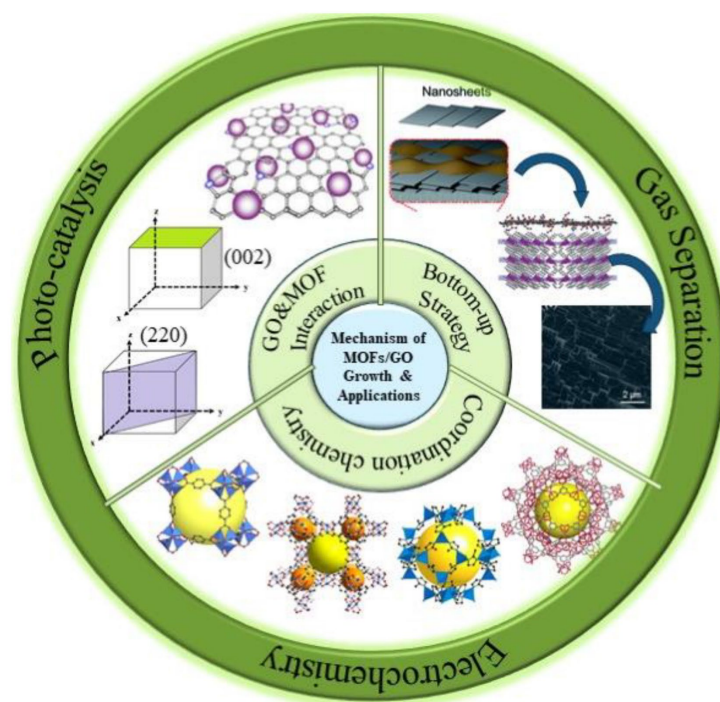


Figure 1. Schematic diagram of factors affecting the growth of a metal–organic framework (MOF) or organometallic framework (OMF) in the presence of graphene oxide (GO) and the applications of oriented OMF/GO nanocomposites. Reprinted from Ref. [35].

This review topic holds critical significance in addressing contemporary energy challenges. With the pressing need to transition towards sustainable energy sources due to environmental concerns and dwindling fossil fuel reserves, this research offers a promising avenue. By leveraging the remarkable properties of graphene, such as its exceptional electrical and thermal conductivity, and incorporating organometallic frameworks renowned for their tunable structures, researchers aim to create hybrid materials with enhanced functionality. These materials hold immense potential for improving energy storage and conversion devices like batteries, supercapacitors, and fuel cells, thus advancing renewable

energy technologies. By promoting the development of eco-friendly energy solutions, this research contributes to the global effort to combat climate change and foster sustainable development.

2. Synthesis Techniques

Figure 2 depicts various synthesis approaches for the preparation of MOF-grafted GO composites. One common method involves the chemical reduction of graphene oxide in the presence of organometallic precursors, leading to the simultaneous formation of organometallic frameworks on the graphene oxide surface [23,36,37]. This one-pot synthesis route ensures intimate contact between graphene oxide and organometallic components, promoting uniform distribution and enhanced interfacial interactions [37,38]. Table 1 provides a comparative overview of the characteristics associated with each synthesis method, highlighting their respective advantages and limitations in preparing graphene-based composites for eco-energy applications.

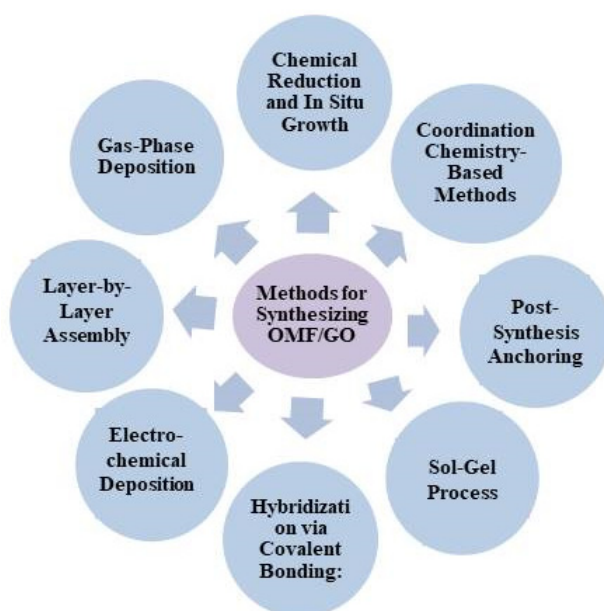


Figure 2. Various methods for synthesis of OMF/GO composite.

Table 1. Comparison of characteristics for graphene-based composites prepared by various main synthesis methods.

Synthesis Method	Characteristics	References
Chemical vapor deposition (CVD)	Uniform and continuous graphene layers grown on substrates; high scalability for industrial production; controlled thickness and morphology of graphene films; potential for precise control over dopants and functionalization	[39]
Hydrothermal synthesis	Facile and cost-effective synthesis in aqueous solutions; formation of well-defined GO sheets; incorporation of metal ions into GO structure; limited control over size and distribution of metal nanoparticles	[40]
Solvothermal synthesis	High-temperature synthesis in organic solvents; enhanced control over size and morphology of nanoparticles; formation of stable organometallic complexes with graphene oxide; potential for precise tuning of composite properties through solvent selection and reaction conditions	[41]
Electrodeposition	Direct deposition of metal ions onto graphene surfaces; fine control over deposition parameters and film thickness; high purity of deposited metal nanoparticles; possibility for in situ modification of graphene surface during deposition process	[42]

Table 2 provides a concise overview of how factors such as precursor concentration, reaction conditions, solvent choice, and the presence of functional groups impact the structure and performance of graphene oxide-based composites, supported by relevant citations [43–46].

Table 2. Summarizing the factors influencing the structure and performance of composites involving graphene oxide.

Factors	Influence on Composites	References
Precursor concentration	Determines density of active sites, impacting composite properties	[43]
Reaction conditions	Dictates kinetics, morphology, and crystallinity of the composite	[44]
Solvent choice	Affects dispersion and homogeneity of the composite	[45]
Presence of functional groups	Alters electronic and chemical properties, impacting performance	[46]

Coordination chemistry provides a versatile platform for synthesizing organometallic–graphene oxide composites. Metal ions coordinate with organic ligands and functional groups on GO, allowing for controlled attachment of organometallic entities through π – π interactions or covalent bonding. In the post-synthesis anchoring method, GO is first synthesized, and then organometallic species are anchored onto the surface through various grafting techniques or chemical modifications. This approach provides control over GO functionalization, enabling precise attachment of organometallic frameworks.

The sol–gel process converts metal-containing precursors into a sol or gel, which is then combined with GO and subjected to controlled gelation or drying processes to form the composite material, offering excellent control over composite composition and structure [47]. Covalent bonding strategies form strong chemical bonds between functional groups on GO and organometallic moieties, ensuring a stable and durable hybrid structure under various environmental conditions [48]. Electrochemical deposition allows for controlled growth of organometallic frameworks on graphene oxide surfaces by applying an electric potential, enabling precise control over layer thickness and morphology, which influences composite properties [23,49,50]. Layer-by-layer assembly involves stepwise deposition of alternating layers of graphene oxide and organometallic components, creating a well-defined multi-layered composite structure, allowing for tailoring of layer thickness and composition for fine control over composite properties [23,51–53].

Gas-phase deposition methods involve introducing organometallic precursors in vapor phase for deposition onto graphene oxide surfaces [54–58]. Gas-phase deposition ensures uniform coating of graphene oxide with organometallic species, ensuring composite structural homogeneity [59,60]. In considering the various synthesis methods for organometallic–graphene oxide composites, it is evident that a variety of techniques exist, each offering unique advantages in terms of control, scalability, and resulting composite properties. The choice of synthesis method depends on specific requirements of intended eco-energy applications and desired characteristics of the hybrid material.

Traditional methods for synthesizing graphene oxide pose significant environmental concerns due to their reliance on harsh chemical reagents and energy-intensive processes [61]. The widely used Hummers method, for example, employs strong oxidizing agents like potassium permanganate and concentrated sulfuric acid, generating hazardous waste products such as sulfur dioxide and nitrogen oxides [62]. High temperatures and prolonged reaction times further contribute to elevated energy consumption and greenhouse gas emissions, while the disposal of waste byproducts exacerbates environmental degradation and can potentially contaminate soil and water sources [63]. To address these challenges, researchers are exploring alternative, more sustainable approaches to graphene oxide synthesis [64], including green synthesis methods using renewable resources and non-

toxic reagents, as well as innovative techniques such as microwave-assisted synthesis and electrochemical methods [40]. These eco-friendly approaches aim to mitigate the environmental impact of graphene oxide production by minimizing the use of hazardous chemicals and reducing energy consumption while maintaining high quality and performance [65]. Additionally, advancements in nanotechnology are enabling the development of scalable and environmentally sustainable synthesis techniques, such as using water-based solvents and biocompatible compounds derived from natural sources, promoting the widespread adoption of graphene oxide in various industries while minimizing adverse effects on the environment and human health [66].

Exploring alternative, environmentally friendly synthesis methods for graphene oxide and its hybrids is essential to mitigate the environmental impact of traditional routes [67]. Green chemistry principles offer promising avenues, emphasizing the use of renewable resources, biodegradable solvents, and natural catalysts to reduce ecological footprints [68,69]. Innovative techniques like microwave-assisted synthesis, ultrasonication, and electrochemical exfoliation optimize energy efficiency and resource utilization, minimizing reliance on hazardous reagents while maintaining high-quality graphene oxide production [23]. Additionally, bioinspired synthesis strategies emulate natural processes to guide the assembly and functionalization of graphene oxide hybrids, offering sustainable pathways for tailored material design across diverse applications, including energy storage and biomedical engineering [70]. Through interdisciplinary collaboration and innovation, researchers are advancing sustainable nanomaterial production by prioritizing environmental considerations and adopting green chemistry principles, aiming to develop eco-friendly processes that align with resource conservation and pollution prevention goals [71]. By embracing alternative synthesis methods and bioinspired approaches, graphene oxide synthesis is evolving towards a greener and more sustainable future, facilitating the widespread adoption of these advanced materials in various industries.

Figure 3 illustrates the multifaceted applications of organometallic–graphene oxide composite materials, emphasizing their importance in energy harvesting, energy storage, catalytic processes, and environmental applications. Detailed explanations are provided in the subsequent sections.

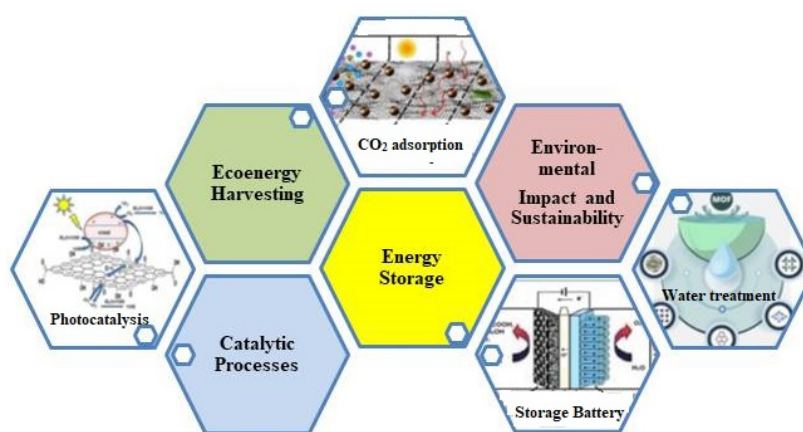


Figure 3. Highlights key applications of organometallic–graphene oxide composites.

3. Ecoenergy Harvesting

3.1. Photocatalytic Properties of Organometallic–Graphene Oxide Hybrids

Photocatalysis, utilizing light to initiate chemical reactions, is pivotal in eco-energy harvesting [68], offering a promising avenue for converting solar energy into chemical energy [72]. Integrating organometallic frameworks onto GO enhances the photocatalytic properties of the hybrid material by combining the catalytic activity of organometallic species with GO's excellent charge transport properties [69,70]. Particularly, rGO's high conductivity facilitates efficient charge separation, preventing electron–hole pair recombina-

tion [73,74], while organometallic species generate active sites for photocatalytic reactions, promoting efficient charge migration. The choice of metal complexes and ligands significantly influences photocatalytic performance [75], with coordination on GO surfaces creating active sites for light-induced reactions. GO's π -conjugated structure allows for efficient light absorption across a broad spectrum, complemented by the light-absorbing characteristics of organometallic moieties, resulting in enhanced overall absorption. OMF-GO hybrids show promise in solar water splitting for hydrogen production [76,77], providing a sustainable pathway for clean fuel generation. Additionally, these hybrids demonstrate photocatalytic activity in degrading pollutants like organic dyes and contaminants, with the OMF enhancing process efficiency for environmental remediation [78,79]. GO's functional groups allow for surface modifications, critical for tailoring the photocatalytic activity of OMF-GO hybrids to specific eco-energy applications. Assessing long-term stability and reusability is crucial for practical use, ensuring economic viability and environmental sustainability. Addressing challenges such as material stability, scalability, and cost-effective catalyst development remains imperative for advancing the field.

Exploration of new organometallic species, ligand designs, and GO modifications holds promise for achieving enhanced photocatalytic performance in future eco-energy applications [80,81]. Graphene oxide nanocomposites exhibit significant potential for eco-energy harvesting through their enhanced photocatalytic properties [82], synergizing GO's unique characteristics with OMF's catalytic capabilities, opening avenues for sustainable energy applications from hydrogen production to environmental pollutant remediation [23,83]. Continuing research and addressing challenges in this field will contribute to developing efficient and environmentally friendly eco-energy technologies.

The production of hydroxyl radicals ($\text{OH}\bullet$) through photoreactions can have various applications in both energy and environmental contexts in Figure 4. Hydroxyl radicals ($\text{OH}\bullet$) are highly reactive and play a crucial role in several processes [84]. Here are some of the uses and applications:

- i. Advanced oxidation processes (AOPs): Hydroxyl radicals are powerful oxidizing agents, and they are often involved in advanced oxidation processes for water and wastewater treatment [85]. AOPs, such as photocatalysis and ozonation, utilize hydroxyl radicals to break down and degrade organic pollutants in water, rendering them less harmful.
- ii. Air purification: Hydroxyl radicals can be used to purify air by oxidizing volatile organic compounds (VOCs) and other pollutants [86]. Photocatalytic air purifiers, for example, use materials that generate hydroxyl radicals when exposed to light, helping to remove pollutants from the air.
- iii. Water disinfection: Hydroxyl radicals are effective in disinfecting water by destroying microorganisms, including bacteria and viruses [87]. Photocatalytic disinfection processes can use light-induced reactions to generate hydroxyl radicals and ensure water safety [88].
- iv. Solar water splitting: In the field of renewable energy, hydroxyl radicals can be involved in solar-driven water splitting processes [89]. These processes use sunlight to split water into hydrogen and oxygen, which can then be used as clean fuel sources [90].
- v. Photocatalytic fuel production: Hydroxyl radicals can be part of photocatalytic reactions aimed at producing fuels from renewable resources [91]. For instance, the generation of hydrogen from water using sunlight and semiconductor materials involves hydroxyl radicals as intermediates [92,93].
- vi. Environmental remediation: Hydroxyl radicals can be utilized in environmental remediation to break down and degrade persistent organic pollutants in soil and water [94]. This is particularly important for cleaning up contaminated sites.
- vii. Dye degradation in the textile industry: The textile industry often generates wastewater containing dyes that can be harmful to the environment. Hydroxyl radicals can

be employed in photocatalytic processes to degrade these dyes, providing a more sustainable approach to wastewater treatment in the textile sector [95].

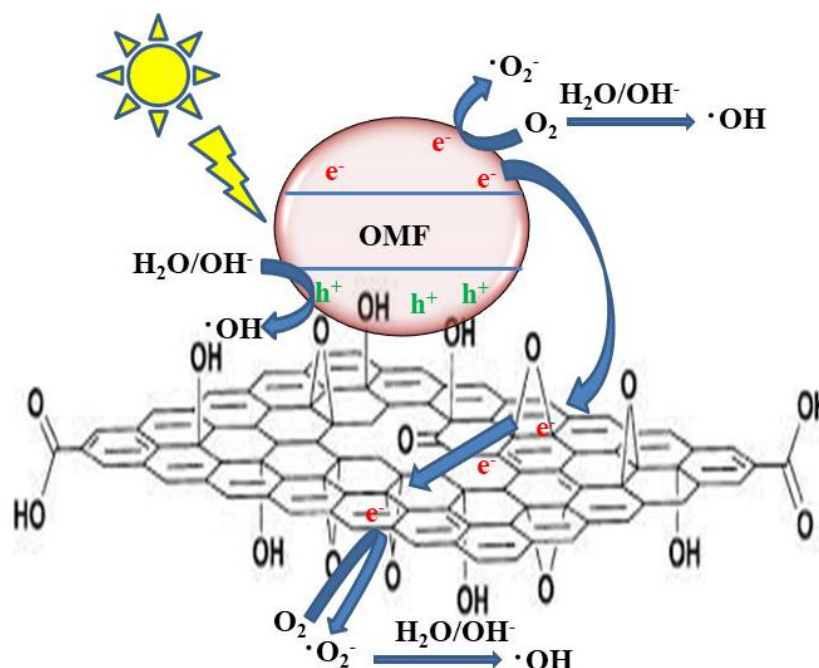


Figure 4. Illustrates the generation of hydroxyl radicals through sunlight irradiation using an organometallic framework–graphene oxide composite.

Overall, the use of hydroxyl radicals in photoreactions offers versatile solutions for addressing environmental pollution, water treatment, and the development of sustainable energy technologies [96]. These applications contribute to a cleaner environment and the efficient utilization of renewable resources.

In OMF-GO hybrids used for photocatalysis, each component plays a crucial role in enhancing overall efficiency. The diverse ligands and metals in the OMF serve as primary active sites, catalyzing reactions by providing surface sites for adsorption and facilitating charge carrier generation and separation. GO confers several advantages, acting as a support matrix to prevent OMF aggregation and ensuring effective use of active sites. Functional groups on GO, such as carboxyl ($-\text{COOH}$) and hydroxyl ($-\text{OH}$) groups, enhance electron transfer, promote light absorption due to GO's conductivity and surface area, and mitigate charge carrier recombination. These combined effects significantly boost the photocatalytic performance of the hybrid material. Understanding the distinct functions of each component allows for researchers to design and optimize OMF-GO hybrids for diverse photocatalytic applications, advancing sustainable energy technologies and offering innovative solutions to environmental challenges.

3.2. Harnessing Solar Energy for Sustainable Power Generation

Solar energy stands as an abundant and renewable resource with immense potential for sustainable power generation, crucial for reducing reliance on finite fossil fuels and mitigating environmental impacts. Photovoltaic (PV) technologies directly convert sunlight into electricity through the photovoltaic effect, with ongoing research focused on enhancing efficiency through GO-based material innovations, novel designs, and manufacturing processes [97,98]. Organic photovoltaics (OPVs), utilizing organic materials as the active layer, offer flexibility, lightweight, and cost-effectiveness [99], with integrating organic materials with graphene oxide and organometallic frameworks presenting opportunities for further improving OPV performance [100,101]. Concentrated solar power (CSP) systems concentrate sunlight onto a small area using mirrors or lenses, generating high

temperatures to drive thermal power cycles [102], with graphene oxide and organometallic frameworks potentially contributing to improved CSP designs. Tandem solar cells stack multiple layers of different materials to capture a broader spectrum of sunlight, enhancing overall efficiency, and the integration of graphene oxide and organometallic frameworks may further enhance tandem cell designs [103,104]. Solar-to-hydrogen conversion utilizes solar energy to split water into hydrogen and oxygen, with organometallic–graphene oxide hybrids exhibiting promising catalytic properties for enhancing efficiency [105–107]. Efficient energy storage is essential for solar energy harvesting, with advances in battery technologies and graphene-based supercapacitors contributing to sustainable energy storage solutions [108,109]. The integration of graphene oxide and organometallic frameworks enhances photocatalytic properties, making them suitable for solar water splitting and pollutant degradation [110,111], while graphene-based transparent conductive films improve electrical conductivity in solar cells while maintaining transparency [112].

Overcoming solar energy production intermittency and developing efficient storage solutions remain ongoing challenges, with hybrid materials like OMF-GO composites holding potential for addressing these challenges. Balancing cost-effectiveness and performance is critical for widespread solar technology adoption [113]. Perovskite solar cells represent a rapidly evolving technology with the potential to revolutionize solar energy harvesting [114], with ongoing research exploring integration with GO and OMF for improved stability and efficiency [115,116]. Integration with smart grids, advancements in energy management systems, and intelligent solar technologies contribute to optimizing solar energy utilization and grid integration [117]. Harnessing solar energy for sustainable power generation is pivotal for transitioning towards a cleaner and more sustainable energy future. The integration of graphene oxide, organometallic frameworks, and emerging materials in solar technologies presents exciting opportunities for improving efficiency, durability, and expanding the scope of solar applications in diverse eco-energy systems [118–120]. Continuous research and innovation are crucial for realizing the full potential of solar energy as a cornerstone of sustainable power generation.

4. Energy Storage Applications

4.1. Electrochemical Capacitance of Organometallic Frameworks on Graphene Oxide

Energy storage is pivotal for the transition to sustainable energy systems, offering flexibility to manage intermittent renewable energy sources. The exploration of novel materials for energy storage applications, particularly the integration of organometallic frameworks with graphene oxide, has garnered significant attention [121,122]. Organometallic frameworks, with their well-defined structures and tunable properties, offer unique opportunities in energy storage [123–125], introducing redox-active sites that influence electrochemical behavior and contribute to improved capacitance. Graphene oxide, with its high surface area, excellent electrical conductivity (for rGO), and mechanical strength, provides an ideal platform for hosting organometallic frameworks [126]. Its functional groups not only serve as anchoring sites but also enhance charge transfer kinetics, bolstering electrochemical performance. Various synthesis methods, including chemical reduction processes and coordination chemistry-based approaches, are employed to fabricate these hybrids [127,128], each method influencing the resulting material's structure, composition, and electrochemical properties [126].

Accurate characterization, crucial for understanding electrochemical behavior, employs techniques like cyclic voltammetry, galvanostatic charge/discharge, and impedance spectroscopy, providing insights into capacitance, charge storage mechanisms, and kinetics. Advanced methods like in situ spectroscopy enable real-time monitoring during electrochemical processes. Evaluation of electrochemical capacitance involves metrics such as specific capacitance, energy density, power density, and cycle stability, with organometallic framework–graphene oxide hybrids demonstrating notable improvements [127,128]. Their enhanced electrochemical capacitance positions them as promising candidates for supercapacitor applications [129], suitable for rapid charge and discharge cycles. Exploring their

potential in hybrid capacitive batteries offers avenues for bridging traditional capacitors and batteries. Despite progress, challenges remain in optimizing electrochemical performance and ensuring long-term stability, particularly regarding material volume expansion during cycling and degradation mechanisms. Future research may involve exploring new organometallic species, optimizing synthesis methods, and developing scalable production processes. The integration of organometallic frameworks on graphene oxide presents a compelling approach to enhance electrochemical capacitance for energy storage applications [130,131], opening possibilities for high-performance and sustainable energy storage devices [132]. As research progresses, these hybrid materials hold potential to play a pivotal role in shaping the future of energy storage technologies.

4.2. Advancements in Eco-Friendly Energy Storage Devices

Energy storage is critical for sustainable and eco-friendly energy solutions. Integrating organometallic frameworks with graphene oxide offers a cutting-edge approach to overcoming traditional limitations in energy storage devices [133,134]. Organometallic frameworks, with their well-defined structures and versatile metal centers, introduce unique properties, allowing for precise engineering of redox-active sites and enhancing device performance [135]. Combining them with graphene oxide creates a synergistic effect [136], with graphene oxide's high surface area and excellent electrical conductivity (for rGO) serving as an ideal support matrix [136,137]. This collaboration optimizes charge transfer, stability, and overall electrochemical performance. Various synthesis strategies, from chemical reduction to coordination chemistry, enable precise control of hybrid material composition for tailored properties in specific applications [50,138]. Accurate characterization techniques such as cyclic voltammetry, impedance spectroscopy, and advanced spectroscopic methods provide insights into electrochemical behavior, charge storage mechanisms, and long-term stability [139]. The integration of OMF-GO holds immense promise in eco-friendly devices like supercapacitors and hybrid capacitive batteries, where enhanced electrochemical capacitance enables rapid charge and discharge cycles [140]. These hybrids show advancements in specific energy and power densities, crucial for evaluating device efficiency and performance [141]. Their eco-friendly nature aligns with sustainability principles, offering alternatives to traditional materials with potential environmental impacts [48,142]. Despite significant progress, challenges such as scalability, cost-effectiveness, and long-term stability persist. Addressing these challenges presents opportunities for further innovation and optimization. Looking ahead, the integration of organometallic frameworks with GO is poised to advance eco-friendly energy storage devices, with continuous exploration and collaborative efforts holding the key to unlocking their full potential.

5. Catalytic Processes

5.1. Organometallic–Graphene Oxide Catalysts for Green Energy Production

Catalysis is crucial for efficient and selective chemical transformations in green energy production. Integrating OMF-GO as catalysts advances catalytic processes in this domain [77,143,144]. Organometallic frameworks grafted onto graphene oxide exhibit unique attributes that enhance catalytic performance by leveraging well-defined metal centers, tunable ligands, and the high surface area and conductivity of reduced graphene oxide to create synergistic effects [125,145,146]. These catalysts are applicable in green energy pathways such as hydrogen production through water splitting, carbon dioxide reduction for sustainable fuel synthesis, and other eco-friendly transformations [147,148]. The presence of GO enhances reaction kinetics by providing a conductive and high-surface-area platform, accelerating electron transfer and ensuring efficient catalytic site utilization, thus improving overall reaction rates.

Organometallic–graphene oxide catalysts demonstrate remarkable selectivity in catalyzing clean fuel synthesis reactions [149,150]. The controlled environment provided by the OMF allows for precise control over reaction pathways, minimizing unwanted

byproducts and enhancing sustainability [151]. The tunability of OMFs enables tailoring catalytic sites for high activity and selectivity, essential for green energy processes [152]. Various synthesis strategies, including chemical reduction, coordination chemistry-based methods, and post-synthesis grafting, are used to design these catalysts [54,153], with the choice of method affecting the hybrid material's structure, composition, and catalytic properties. Accurate characterization using techniques like in situ spectroscopy, X-ray absorption spectroscopy, and operando studies provides insights into active sites, reaction intermediates, and overall catalytic mechanisms.

The synthesis of palladium nanoparticles (Pd NPs) utilizing a molecular-level palladium complex supported onto reduced graphene oxide (2-rGO) has garnered significant attention in recent research endeavors (Figure 5). Understanding the formation and evolution of these active species is pivotal, prompting researchers to conduct a thorough analysis of the hybrid material rGO during recycling experiments [154]. Following catalytic runs, samples of the solid catalyst were extracted and subjected to detailed examination using high-resolution transmission electron microscopy (HRTEM) and X-ray photoelectron spectroscopy (XPS) (Figure 6).

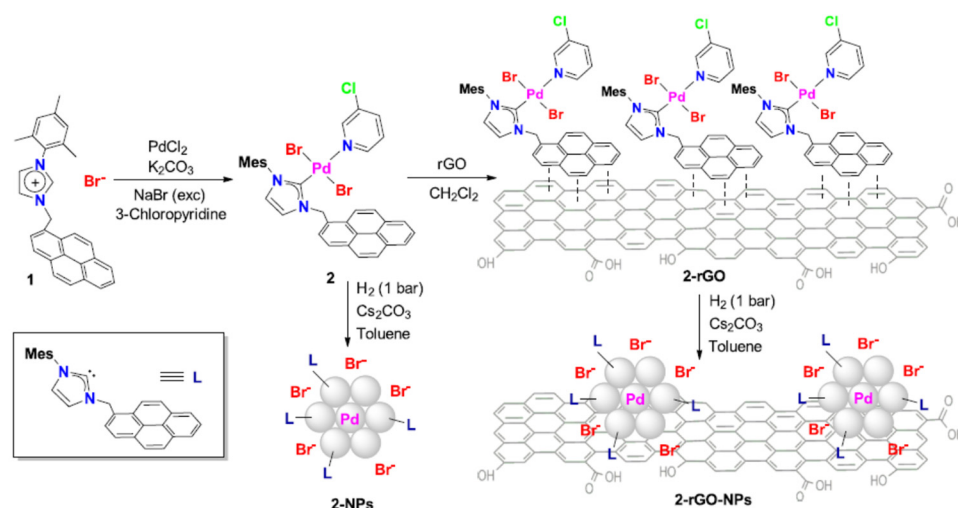


Figure 5. Synthesis of palladium complex 2, immobilization onto rGO (2-rGO), and synthesis of free palladium NPs (2-NPs) and supported onto rGO (2-rGO-NPs). Reprinted from Ref. [154].

HRTEM imaging consistently confirmed the presence of palladium NPs across all catalytic runs. Initially, a narrow distribution of particle sizes was observed, with an average size of 3.54 ± 1.03 nm ($n = 236$) in the first run (Figure 6d). However, after ten runs, the particle size distribution widened, accompanied by a slight increase in the average size of the Pd NPs to 6.62 ± 2.80 nm ($n = 282$) (Figure 6i). These findings indicate that Pd NPs undergo subtle changes with successive catalytic reactions.

XPS analysis further elucidated the composition of the palladium NPs, revealing the presence of both Pd(2+) and Pd(0) oxidation states. Additionally, surface decoration of these NPs with N-heterocyclic carbene (NHC) and bromide (Br^-) ligands was observed, as evidenced by the assignment of characteristic peaks corresponding to N (400.6 eV) and Br (68.6 eV). Intriguingly, the ratio of Pd(2+)/Pd(0) was found to vary throughout the recycling runs, indicating fluctuations in both the size and composition of the Pd NPs during catalytic experimentation. The observed decrease in the proportion of Pd(2+) and increase in Pd(0) from the first to the tenth run suggests a structural rearrangement within the Pd NPs. Specifically, it is proposed that Pd(2+) resides predominantly at the surface of the nanoparticles, while Pd(0) is concentrated in the core. The increase in NP size over successive runs results in reduced specific surface area, correlating with the decrease in Pd(2+) content. This dynamic exchange of size and composition of Pd NPs under catalytic

conditions underscores the intricate nature of these systems and highlights the necessity for a comprehensive understanding of their behavior.

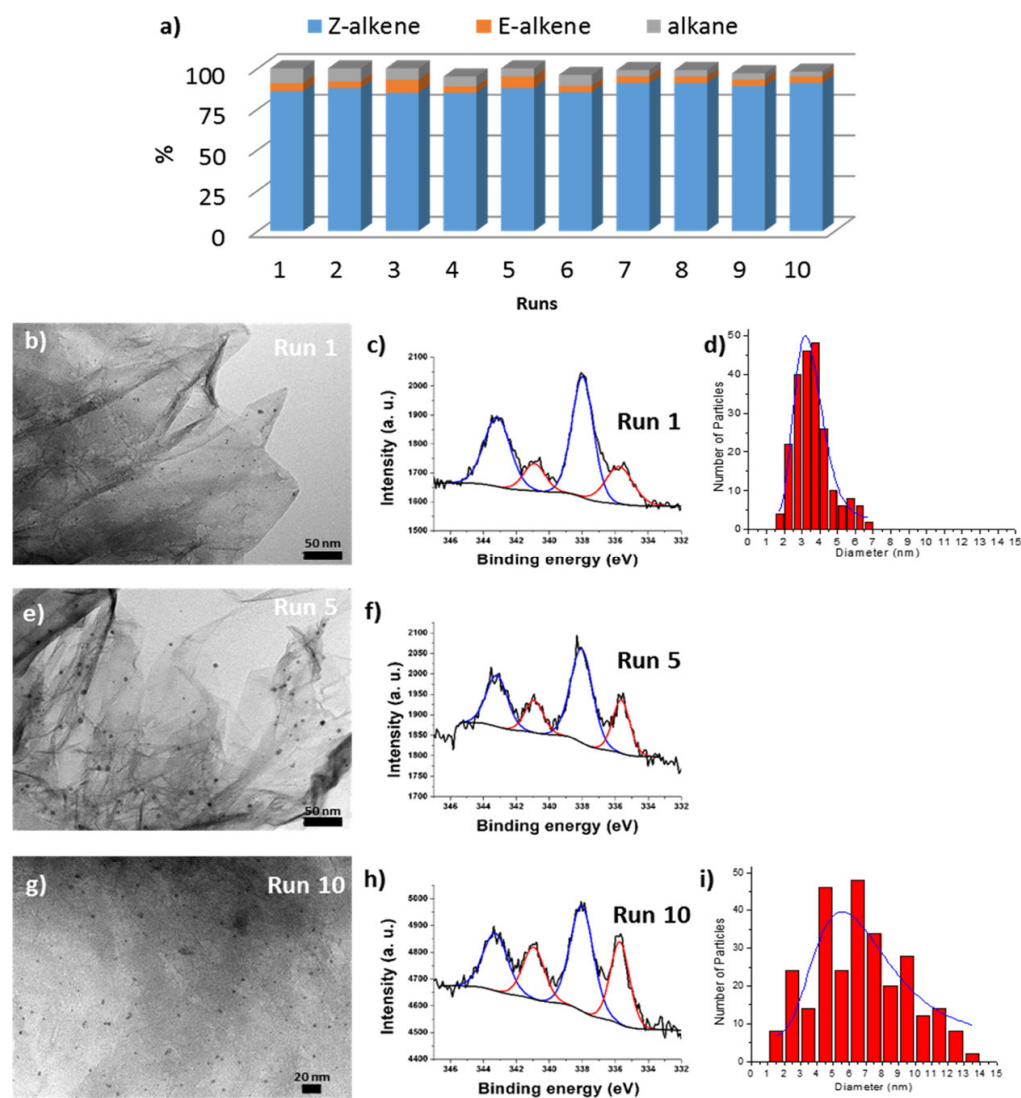


Figure 6. (a) Recycling of 2-rGO (0.5 mol %) in the hydrogenation of 1-phenyl-1-butyne with molecular H_2 . First run at 1.5 h because of the induction time and next runs at 1 h. HRTEM image after run 1 (b), run 5 (e), and run 10 (g) and the corresponding XPS spectra of runs 1 (c), 5 (f), and 10 (h). Blue line corresponds to the Pd(II) core-level peaks 3d and red line corresponds to the core-level peaks 3d of Pd(0). Particle size distribution histograms for run 1 (d) and run 10 (i). Reprinted from Ref. [154].

The dynamic exchange of metal NPs observed during catalytic transformations aligns with prior research findings, underscoring the importance of considering the dynamic nature of nanomaterials in catalytic processes. These insights significantly advance our understanding of Pd NP-based catalysts and promise to optimize their performance in various catalytic applications. Evaluating the stability and recyclability of organometallic-graphene oxide catalysts is crucial for practical applications, ensuring sustained catalytic activity over multiple cycles while minimizing leaching of active species into the reaction mixture [154]. Challenges in scalability, cost-effectiveness, and potential toxicity of certain metals in organometallic frameworks require careful consideration, with future prospects focused on addressing these challenges, exploring new catalytic reactions, and tailoring catalysts for specific green energy applications. Organometallic-graphene oxide catalysts represent a frontier in catalysis for green energy production [155,156], with their unique attributes, versatility, and efficiency positioning them as catalysts of choice for advancing

sustainable and environmentally friendly processes. As research progresses, the integration of these hybrid catalysts holds great promise for shaping the landscape of green energy production.

5.2. Role in Fuel Cells and Hydrogen Evolution Reactions

Fuel cells stand as a clean and efficient energy conversion technology, with advanced catalyst development being pivotal for their performance enhancement. The integration of organometallic frameworks with GO as catalysts has emerged as a promising strategy, particularly in the context of hydrogen evolution reactions (HERs) crucial for fuel cell applications [157,158]. The synergy between organometallic frameworks and graphene oxide imparts unique attributes to these composites [159,160], with organometallic entities introducing catalytic activity and graphene oxide providing high surface area and excellent electron conductivity, collectively enhancing the efficiency of fuel cell reactions, especially HERs [161]. Organometallic–graphene oxide composites find applications as catalysts in various types of fuel cells, including proton exchange membrane fuel cells (PEMFCs) and alkaline fuel cells (AFCs) [162,163], playing a pronounced role in the electrocatalytic processes involved in fuel cell reactions. The HER is a key component of fuel cell reactions, involving the electrochemical reduction of protons to produce hydrogen [164]. Organometallic–graphene oxide composites act as efficient catalysts in driving this reaction, offering advantages such as improved kinetics, reduced overpotential, and enhanced durability [165,166].

Illustrated in Figure 7, the fabrication pathway of the ICZTS/GO electrocatalytic system delineates the integration of metal precursors alongside the pivotal role of GO as a supporting material. GO not only serves as a scaffold but also enhances the HER activity, a detailed synthesis of which is expounded upon in Reference [165]. Notably, the comparative assessment of CZTS and GO electrodes reveals their subpar HER activities in contrast to I-CZTS/GO composites. The augmented electrochemical performance observed in CZTS/GO composites is attributed to the synergistic electronic and chemical interactions facilitated by the CZTS-GO interface (refer to the schematic depiction in Figure 8).

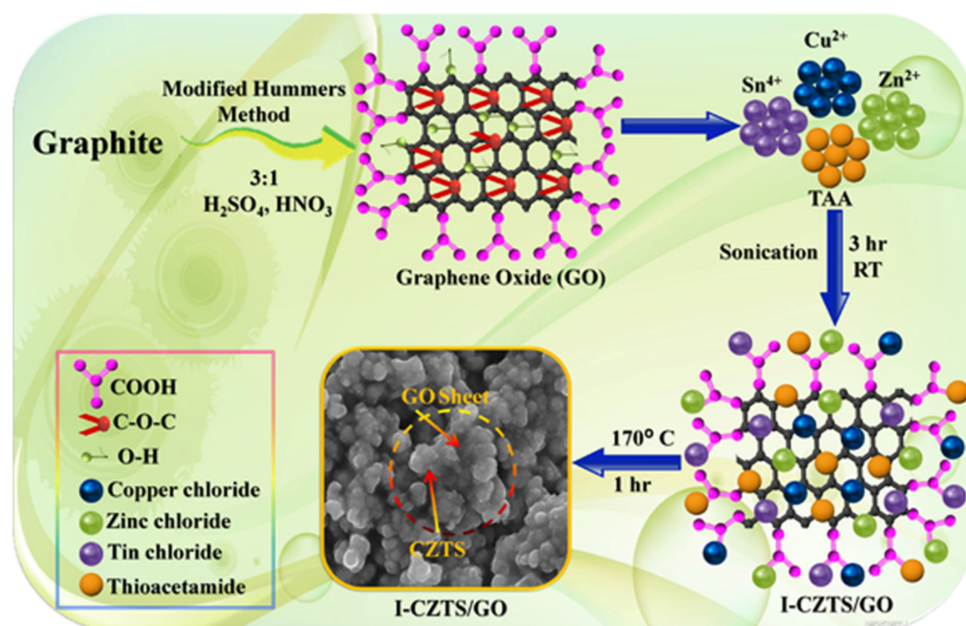


Figure 7. Schematic illustration of different steps for synthesis of in situ synthesized $\text{Cu}_2\text{ZnSnS}_4$ (I-CZTS)/GO composites. Reprinted from Ref. [165].

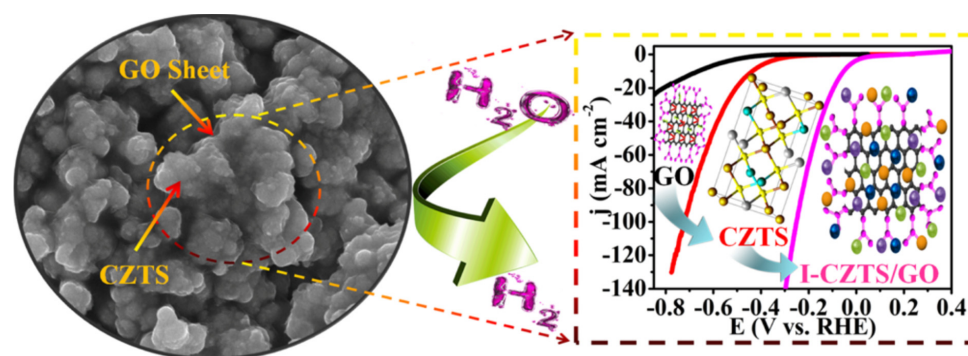


Figure 8. Schematic of HER on the I-CZTS/GO composite. Reprinted from Ref. [165].

Understanding the catalytic mechanisms of organometallic–graphene oxide composites in the hydrogen evolution reaction (HER) is crucial, with the active sites provided by the organometallic entities facilitating initial proton reduction steps, while graphene oxide enhances charge transfer and electron transport [166,167]. Various synthesis strategies, including wet chemical methods, electrochemical deposition, and chemical vapor deposition, are employed to fabricate these composites [54,168], with tailored synthesis parameters optimizing composition and structure for enhanced catalytic performance. Rigorous characterization, including cyclic voltammetry, chronoamperometry, and in situ spectroscopy, provides insights into electrochemical behavior, active sites, and overall efficiency. Organometallic–graphene oxide composites exhibit durability, with stable attachment of organometallic species ensuring long-term stability during extended fuel cell operation.

Efficient HER catalysis by these composites contributes to sustainable hydrogen production, critical for advancing fuel cell technologies [169,170], aligning with eco-friendly energy conversion principles. Despite advancements, challenges such as optimizing catalyst loading, scalability, and cost-effectiveness need addressing [171]. Future directions involve exploring new organometallic species, modifying graphene oxide, and innovating synthesis methods to further improve catalytic efficiency. These composites play a pivotal role in fuel cells and the HER, driving advancements in clean and efficient energy conversion [172], underscoring their importance in shaping the future of fuel cell technologies.

The integration of organometallic frameworks onto graphene oxide surfaces holds promise for enhancing the efficiency and durability of fuel cells [173]. However, fuel cell technology, despite its environmental advantages over conventional energy sources, faces challenges [174]. Degradation mechanisms observed in metal–air fuel cells, attributed to byproduct accumulation like peroxide, hinder optimal performance and longevity, necessitating innovative approaches to mitigate peroxide formation and enhance system stability. Managing heat release during fuel cell operation is also a significant concern for environmental impact and energy efficiency, driving research into advanced thermal management and waste heat recovery strategies [175]. Ongoing research focuses on several fronts to tackle these challenges. Novel catalyst materials are being developed to improve selectivity, durability, and resistance to byproduct formation [176]. Electrode structures and interfaces are being optimized to enhance mass transport, charge transfer, and electrochemical reactions [177]. Additionally, innovations in electrolyte formulations and management strategies aim to minimize side reactions and enhance ion conductivity. Efforts in thermal management and waste heat recovery seek to optimize heat dissipation and harness excess thermal energy for supplementary power generation or heating applications. Through multidisciplinary endeavors, researchers aim to advance the environmental sustainability and efficiency of fuel cell systems enhanced with organometallic frameworks on graphene oxide.

6. Current Challenges in the Development and Implementation of Ecoenergy Solutions

The landscape of eco-energy solutions presents dynamic challenges demanding innovative approaches. Organometallic–graphene oxide hybrids offer promising solutions for overcoming hurdles in sustainable eco-energy technologies [178]. Identifying prevailing challenges in energy storage, catalysis, renewable energy conversion, and overall system sustainability is crucial [179]. These hybrids possess unique properties, including catalytic activity, conductivity, and tunability, poised to address obstacles in eco-energy applications [23]. Efforts to enhance sustainable fuel production require close examination of the catalytic properties of organometallic–graphene oxide composites [48]. Exploring applications in hydrogen evolution, carbon dioxide reduction, and other key processes holds promise for leveraging these hybrids' capabilities. Through targeted research and development, they can play a pivotal role in advancing eco-energy technologies towards a greener future.

Discussing the potential of organometallic–graphene oxide systems in overcoming energy storage challenges is paramount [180]. Evaluating their role in enhancing device performance and compatibility with renewable energy sources [181] is essential for advancing sustainable energy solutions. Addressing challenges related to large-scale production, scalable synthesis methods, and cost-effectiveness is vital [182,183]. Moreover, assessing their environmental impact and contribution to improving system efficiency and longevity is paramount [184,185]. Understanding how these hybrids minimize energy losses and enhance overall system reliability provides valuable insights into their transformative potential.

Considering the economic viability and affordability of incorporating organometallic–graphene oxide materials in eco-energy solutions [186], as well as their compliance with environmental regulations and standards [187], ensures responsible manufacturing and deployment. Exploring the future prospects of organometallic–graphene oxide hybrids in addressing challenges in eco-energy solutions and suggesting potential research directions, innovations, and collaborations to further harness the capabilities of these materials for sustainable energy development is essential. By exploring their unique properties and applications, it contributes to the advancement of sustainable energy technologies.

The integration of organometallic compounds with graphene oxide nanocomposites represents a promising frontier in the quest for sustainable and eco-friendly energy solutions [137]. This review provides a comprehensive exploration of the achievements and potential of these innovative materials in advancing eco-energy applications. Notably, these hybrids have played a pivotal role in energy conversion, storage, and environmental remediation [188]. The discussion on energy conversion processes, such as photovoltaics and photocatalysis, is particularly enlightening [26]. The synergistic combination of organometallic species and graphene oxide contributes to enhanced efficiency and performance in these applications. Real-world examples and case studies effectively illustrate the successful translation of laboratory advancements into tangible eco-energy solutions.

The integration of carbon materials into MOFs induces significant changes in their coordination chemistry, leading to enhanced conductivity within the composite (Figure 9). This augmentation in conductivity establishes novel pathways for charge transport, thereby amplifying the composite's electrochemical performance. Pioneering investigations by Yaghi et al. have demonstrated the feasibility of doping nanocrystalline MOFs (nMOFs) with graphene sheets, rendering them viable as electrodes for supercapacitors [189]. The resultant composite thin-film electrodes, soaked in a 1 M tetraethylammonium tetrafluoroborate electrolyte, exhibited remarkable capacities for ion storage during charge and discharge cycles. While various nMOFs exhibited commendable capacitance, a zirconium-based MOF, specifically a composite of MOF 867 and graphene, emerged as the standout performer. This composite showcased the highest stack and areal capacitance, coupled with remarkable durability over 10,000 charge/discharge cycles. Such outstanding performance

was attributed to the composite's exceptional porosity and open structure, ensuring efficient ion diffusion and charge transfer kinetics.

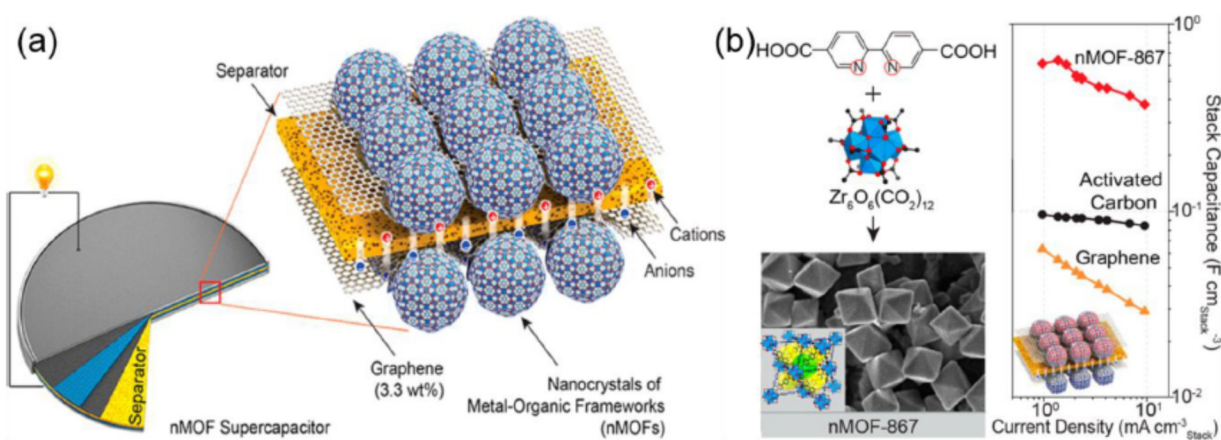


Figure 9. (a) Schematic depiction of nMOF/graphene supercapacitors, and (b) nMOF 867 along with the stack capacitance of nMOF-867/graphene, activated carbon, and pristine nMOF-867. Reprinted from Ref. [189].

The capacitance achieved by this composite notably surpassed that of supercapacitors using commercial activated carbon, demonstrating its potential to revolutionize energy storage technologies [180]. This enhancement underscores the superior performance and durability offered by carbon-doped MOF composites compared to traditional electrode materials. Additionally, the exploration of energy storage applications, including supercapacitors and batteries, provides insight into the impact of organometallic–graphene oxide nanocomposites on energy storage technology. This review also highlights environmental remediation applications, showcasing the versatility of these materials in pollutant removal and water purification [190]. Inclusion of challenges and future prospects enhances this review's value, serving as a guide for future research endeavors [190,191].

While the primary application of our research targets environmental remediation, the core of our work is grounded in chemical engineering. The synthesis and functionalization of the OMF-GO nanocomposite involve intricate chemical reactions and processes fundamental to chemical engineering. Our research focuses on designing, developing, and optimizing nanocomposite materials using advanced chemical engineering techniques. This includes precise control of reaction conditions, characterization of chemical properties, and understanding molecular-level interactions. Moreover, the environmental application of these nanocomposites demonstrates the practical implementation of chemical engineering principles to address environmental challenges. By leveraging our expertise in chemical engineering, we develop innovative solutions applicable to environmental systems, bridging the gap between these fields.

Overall, this review provides a comprehensive analysis of the tangible impact of these materials on advancing eco-energy solutions, making it a valuable resource for researchers, policymakers, and industry professionals invested in sustainable energy futures. Various synthesis methods, such as chemical vapor deposition (CVD) and hydrothermal synthesis, show promise for sustainable eco-energy solutions, with applications spanning energy storage devices, catalysis for energy conversion, and environmental remediation. Through innovative synthesis methods, researchers have demonstrated improved performance metrics, validating the effectiveness of these materials in practical eco-energy applications.

7. Conclusions

This review offers a comprehensive exploration of various synthesis methods for integrating organometallic frameworks with graphene oxide. These methods are highlighted for their efficacy in achieving controlled structures and desired properties, laying a strong

foundation for successful applications in eco-energy. The integration of organometallic frameworks with graphene oxide demonstrates notable enhancements in catalytic activities. The synergistic effects between these materials lead to improved reaction rates, selectivity, and overall efficiency, positioning these hybrids as promising candidates for catalytic applications in sustainable energy solutions. Significantly, the review underscores the remarkable contributions of organometallic–graphene oxide hybrids in photocatalytic and photovoltaic applications. Leveraging the unique properties of the combined materials facilitates enhanced light absorption, charge separation, and electron transport, resulting in superior performance in solar energy conversion processes. Furthermore, the collaborative approach of incorporating organometallic frameworks onto graphene oxide proves advantageous in energy storage applications. These hybrids exhibit superior electrochemical properties, contributing to the development of more efficient and sustainable energy storage devices, including batteries and supercapacitors. Importantly, this review emphasizes the eco-friendly aspect of organometallic–graphene oxide hybrids in environmental applications. These materials hold promise in pollutant removal, water purification, and air quality improvement, highlighting their potential contributions to addressing pressing environmental challenges. Structural insights into the hybrids elucidate the importance of the arrangement of organometallic species on the graphene oxide surface. This understanding is pivotal for tailoring the properties of the materials to meet specific eco-energy requirements. This review candidly addresses challenges encountered in practical implementations, such as scalability and stability issues. By acknowledging these challenges, it provides a realistic foundation for future research directions, offering insights that can guide researchers towards overcoming obstacles in the development and application of these materials. In summary, this review significantly contributes to understanding the collaborative potential between organometallic frameworks and graphene oxide in advancing sustainable eco-energy solutions. Its comprehensive coverage, from synthesis strategies to practical applications and challenges, renders it a valuable resource for researchers, engineers, and policymakers striving towards a more sustainable and energy-efficient future.

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