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Research Progress on Preparation of Reduced Graphene Oxide (rGO) Composites Based on Electrochemical Properties

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Abstract. Carbon electrode materials have emerged as pivotal components for electrochemical energy storage devices owing to their ability to meet stringent requirements for high energy density, prolonged cycle life, cost efficiency, and operational safety. Graphene, a two-dimensional single-atom-layer material, has been engineered into reduced graphene oxide (rGO), a modified derivative that integrates enhanced electrical conductivity with structural stability. rGO-based composites exhibit substantial potential for energy-related applications. This review comprehensively outlines recent advances in preparation strategies and detailed synthetic protocols for diverse rGO composites, specifically focusing on rGO/metal oxide hybrids and rGO/conducting polymer systems. The electrochemical performance enhancements of these fabricated materials, including elevated specific capacitance, improved rate capability, and reinforced cycling stability, are systematically demonstrated. Critical analysis reveals that future research priorities may center on three-dimensional multicomponent architectures and novel structural modifications of rGO composites. Furthermore, the development of innovative dispersion techniques and assembly strategies for rGO-based materials could serve as pivotal solutions to overcome current limitations in scalable synthesis, thereby advancing their practical implementation in next-generation energy storage technologies.

Keywords: rGO composites, electrochemical energy storage materials, nanostructured fabrication strategies, multicomponent synergistic effects

1. Introduction

The development and modification of novel electrode materials have become a central focus in energy storage research, driven by the imperative to meet stringent requirements for high energy/power density, prolonged cycling stability, cost efficiency, and operational safety in electrochemical energy storage systems. Carbon-based materials, owing to their exceptional chemical stability, structural versatility, superior electrical conductivity, abundant availability, and environmental compatibility, have been extensively utilized in advanced carbon-containing or carbon-modified electrode materials. These materials significantly enhance electrochemical performance, effectively addressing critical challenges in energy storage devices such as lithium-ion batteries and supercapacitors [1,2].

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The two-dimensional monolayer graphene, which is made up of sp^2 hybridized carbon atoms organized in a hexagonal lattice, has remarkable electron mobility, mechanical stiffness, and heat conductivity. When integrated with other components, electrons migrate freely and rapidly across graphene's π-conjugated network without scattering from extrinsic impurities or lattice distortions, endowing graphene-based composites with unparalleled electrical conductivity. The oxidation of graphene yields graphene oxide (GO), which has several functional groups that hold oxygen and improve its stability and electrochemical activity. However, excessive functionalization in GO inevitably disrupts the conjugated π-system, thereby deteriorating electrical conductivity [3]. In contrast, reduced graphene oxide (rGO), obtained through controlled reduction processes, achieves an optimal balance between conductivity and stability. This unique combination has positioned rGO-based composites as promising candidates for energy-related applications, spurring extensive research into their synthesis methodologies.

Recent years have witnessed remarkable progress in rGO composite fabrication techniques, yet significant challenges persist, particularly in scalable synthesis and performance optimization [4]. This review focuses on recent advances in rGO composite systems for electrochemical energy storage, with emphasis on two dominant categories: rGO/metal oxide hybrids and rGO/conducting polymer architectures. By analyzing key synthesis strategies and their corresponding electrochemical enhancements—including specific capacitance, rate capability, and cycle life—this review identifies critical research hotspots and technical bottlenecks. Furthermore, researcher explore potential future research directions to overcome current limitations and accelerate the practical deployment of rGO composites in next-generation energy storage technologies.

2. Architecture and synthesis approaches for rGO composites

2.1. Structural classifications

rGO composites can be categorized into six distinct structural configurations, as illustrated in Figure 1, which are (a) immobilized, (b) encapsulated, (c) capsule-like, (d) sandwich-like, (e) layered, and (f) hybrid architectures [5]. Each configuration exhibits unique advantages and limitations in terms of interfacial interactions, charge transport efficiency, and structural stability. Among these, the immobilized configuration remains the most prevalent structural paradigm in current composite fabrication.

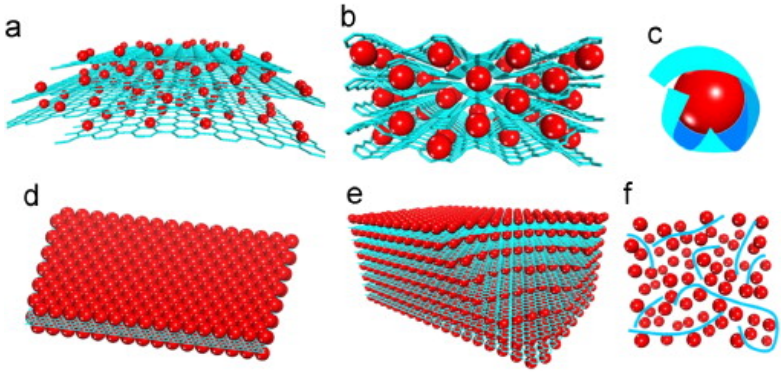

Figure 1: Structural configurations of graphene-metal oxide hybrid architectures. Red spheres represent metal oxide nanoparticles; blue planes denote graphene nanosheets

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2.2. Synthesis methodologies

The synthesis of rGO composites is primarily classified into two categories based on interfacial interaction mechanisms: in situ growth and ex situ assembly strategies.

In situ growth involves inducing the formation of micro/nano crystalline nuclei that directly nucleate and grow on the reactive sites of graphene nanosheets. Representative techniques include liquid-phase deposition, hydrothermal/solvothermal synthesis, chemical vapor deposition (CVD), sol-gel processes, and template-assisted methods. As exemplified in Figure 2, vertically aligned carbon nanotube (CNT) arrays were grown on both sides of GO/rGO substrates via CVD using acetonitrile as the carbon source and Ni nanoparticles as catalysts [6].

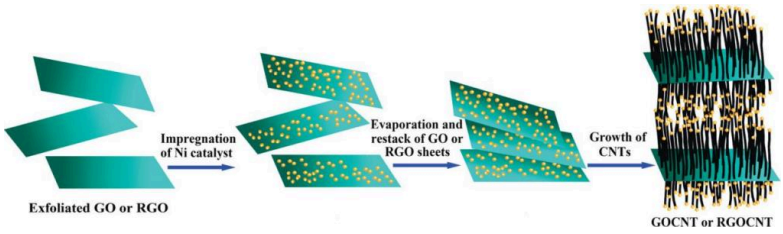

Figure 2: Synthesis protocol for carbon nanotube-pillared GO/rGO heterostructures

Ex situ assembly entails the pre-synthesis of functional nanomaterials, followed by their surface modification and subsequent integration onto functionalized graphene substrates. This approach is further divided into covalent interaction-based and non-covalent interaction-based routes, with electrostatic self-assembly and layer-by-layer assembly being widely adopted for rGO composites. Figure 3 demonstrates a sol-driven electrostatic self-assembly process, where negatively charged GO nanosheets are combined with positively charged Fe $(OH)_3$ colloidal nanoparticles [7].

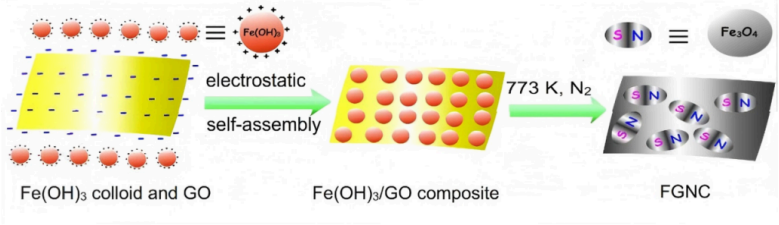

Figure 3: Schematic of the procedure for synthesized magnetic Fe_3O_4/rGO nanocomposite (FGNC) with electrostatic self-assembly

Owing to its inherent advantages—such as eliminating the need for tedious surface modification of metal oxides and avoiding the introduction of surfactants or auxiliary agents—in situ growth has become the predominant methodology for rGO composite fabrication in contemporary research [8].

3. Synthesis and electrochemical performance of rGO/metal oxide composites

3.1. Tin-based metal oxides

Yao et al. developed a carbon-coated SnO_2/rGO composite (C@SnO_2-rGO-SnO_2) with a sandwich-like structure through sequential growth, thermal treatment, and etching processes (Figure 4) [9]. Utilizing mesoporous SiO_2/graphene/SiO_2 (SiO_2-rGO-SiO_2) nanosheets as templates enable precise control over SnO_2 nanocrystal dimensions. The synergistic interplay between rGO and the carbon

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coating established a robust conductive network, mitigating the inherent low conductivity of SnO_2. The composite demonstrated exceptional electrochemical performance with high reversibility.

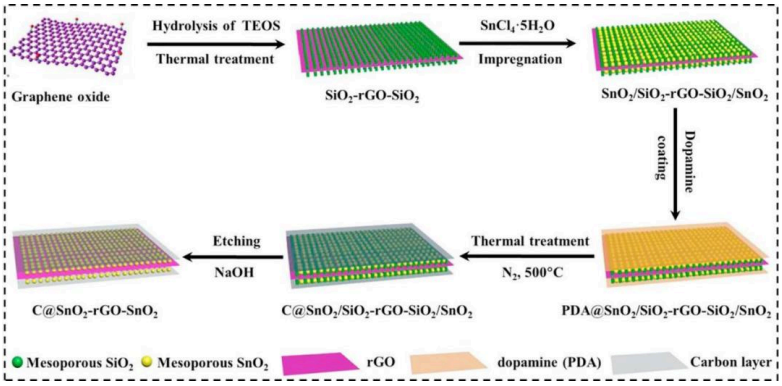

Figure 4: Schematic diagram of the synthesize procedure for the 2D porous C@SnO_2-rGO-SnO_2 nanosheets [9]

A breakthrough in electrode architecture was achieved by Wang M's team through CTAB-mediated hydrothermal synthesis of LTO-integrated rGO/SnO_2 nanocomposites (Figure 5) [10]. The strategic incorporation of cetyltrimethylammonium bromide enabled precise morphological control, yielding a full-cell configuration (LTO/rGO/SnO_2@LFP) that demonstrated exceptional dual performance metrics—130.56 Wh kg^{-1} energy density and 2868.5 W kg^{-1} power density. Notably, the system delivered an initial discharge capacity of 1059.7 mAh g^{-1} at 0.5C cycling rates, establishing new benchmarks for hybrid lithium-ion systems.

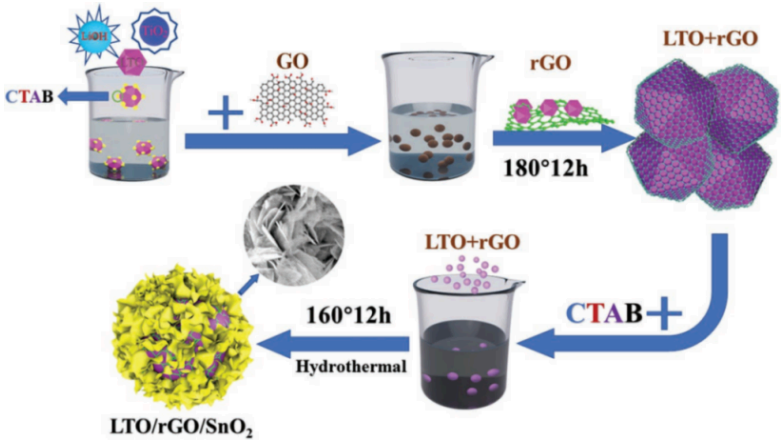

Figure 5: Mechanistic illustration of the triphasic hydrothermal synthesis protocol for constructing hierarchically ordered LTO-anchored rGO/SnO_2 nanocomposites [10]

3.2. Transition metal oxides

Sun et al. fabricated a sandwich-structured CoO-Co_3O_4-rGO composite via a solvothermal templating approach, where rGO sheets served as conductive scaffolds [11]. The material retained a reversible capacity of 994 mAh g^{-1} after 200 cycles at 100 mA g^{-1}, outperforming single-component CoO and Co_3O_4[12].

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Wu et al. embedded Fe_3O_4 nanoparticles between rGO and carbon layers via a solvothermal method, creating a sandwich-like C/Fe_3O_4/rGO composite [13]. The dual carbon architecture effectively suppressed volume expansion of Fe_3O_4 while enhancing electron transport, yielding superior rate capability and cycling stability.

3.3. Bimetallic oxides

Wang Y et al. constructed 3D hollow $ZnSnO_3$ cube/rGO aerogels (ZGAs) through electrostatic self-assembly and hydrothermal synthesis without inert gas protection [14]. The ZGAs' conductive network accommodated volume changes during lithiation/delithiation and facilitated rapid ion/electron transfer, achieving exceptional discharge capacity and rate performance.

Li et al. synthesized $NiGa_2O_4$/rGO composites via hydrothermal-calcination methods [15]. The rGO substrate prevented nanoparticle aggregation and exposed additional Li+ storage sites, delivering a stabilized capacity of 610.5 mAh g^{-1} at 0.1 A g^{-1}, significantly higher than Ga-based bimetallic oxides [16].

3.4. Other chalcogens metal compounds

Zhai et al. fabricated sea-urchin-inspired $CoSe_2$/rGO architectures through a single-step hydrothermal synthesis [17]. The carbon-anchored framework enhanced electroactive surface exposure while suppressing particle coalescence, sustaining a stable discharge capacity.

Tian et al. developed $MoSe_2$-decorated rGO composites as advanced sulfur matrices for lithium-sulfur batteries via a straightforward hydrothermal approach [18]. The optimized $MoSe_2$@rGO/S cathode delivered an initial capacity of 1608 mAh g^{-1} (0.1C), exhibited minimal capacity decay (0.042% per cycle at 0.25C), and retained 870 mAh g^{-1} under high sulfur mass loading (4.2 mg cm^{-2}, 0.3C).

4. Synthesis and electrochemical performance of rGO/conducting polymer composites

The unique electrochemical properties of rGO/conducting polymer composites render them highly promising for supercapacitor applications [19].

Adam Moyseowicz et al. synthesized a porous polypyrrole/rGO (PPy/rGO-HT) composite electrode via a hydrothermal approach, achieving a high specific surface area of 597 m^2 g^{-1} [20]. This composite demonstrated superior specific capacity and cycling stability compared to pristine rGO. When configured into a hybrid supercapacitor, the device delivered an energy density of 6.5 Wh kg^{-1} alongside exceptional long-term stability.

Zahra Karami et al. developed a textile-based Ag nanoparticles/rGO/cotton fiber (Ag/rGO/CF) composite electrode for wearable supercapacitors using a scalable electroless deposition method (Figure 6) [21]. This strategy involved decorating GO-coated cotton fibers with Ag nanoparticles, resulting in simultaneous enhancements in electrochemical performance and mechanical robustness. The electrode exhibited a elevated specific capacitance of 426 ± 10 F g^{-1} in cyclic voltammetry tests, while the corresponding symmetric supercapacitor displayed extended cycle life.

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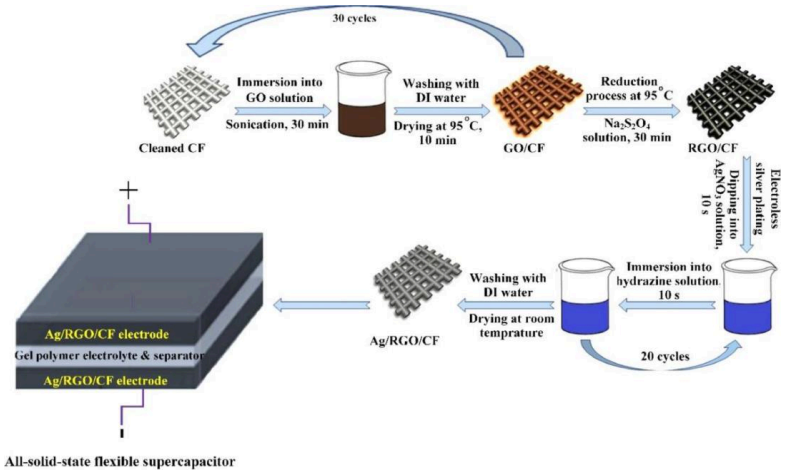

Figure 6: Fabrication process flow for sulfur-rich carbon fiber (SRCF) electrodes and corresponding flexible all-solid-state supercapacitor assembly [21]

A comparative analysis of electrochemical performance metrics for various rGO/conducting polymer composites in supercapacitors is summarized in Table 1. The data conclusively demonstrate that these composites exhibit elevated specific capacitance and cycling durability, underscoring their potential for advanced energy storage applications.

Table 1: Comparative electrochemical performance of rGO/Polymer composites in supercapacitors

Composities	Electrolyte	Current density	Specific capacitance/(F g^{-1})	Cycling performance		Ref.
				Cycle number	Capacity retention/%	
MB@3D-rGO	1 mol L^{-1} H_2SO_4	1.0 A g^{-1}	311	10000	96.0	[22]
DASNCs-rGO (1:2)	2 mol L^{-1} KCl	5.0 mV s^{-1}	214	5000	81.7	[23]
DASNCs-rGO (2:1)	2 mol L^{-1} KCl	5.0 mV s^{-1}	316	5000	76.0	[23]
rGO/PEDOT	1 mol L^{-1} H_2SO_4	0.053 A cm^{-3}	202.7	9000	90.0	[24]
ZnS/rGO/PANI	6 mol L^{-1}KOH	1.0 A g^{-1}	1045	1000	160.0	[25]
PANI/rcGO	1 mol L^{-1} H_2SO_4	0.5 A g^{-1}	299	2000	88.0	[26]

MB: methylene blue; DASNCs: dialdehyde starch nanocrystals; PEDOT: poly(3,4-ethylenedioxythiophene); PANI: polyaniline; rcGO: reduced and crushed graphene oxide

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5. Synthesis and performance applications of other rGO composites

5.1. Pyramid photothermal architecture based on CuS/rGO for high-performance thermoelectric generation

Zeng et al. developed a hierarchical pyramid photothermal system by integrating CuS-rGO composites with carbon fiber (CF) through rGO modification (Figure 7)[27]. The architecture harnessed the combined benefits of rGO's wide-spectrum light harvesting and CuS's favorable bandgap characteristics with efficient photon utilization. This configuration demonstrated a record evaporation rate of 2.29 kg m^{-2} h^{-1} and peak power output of 1.32 W m^{-2}, achieving top-tier efficiency in integrated solar-thermal evaporation and thermoelectric energy conversion.

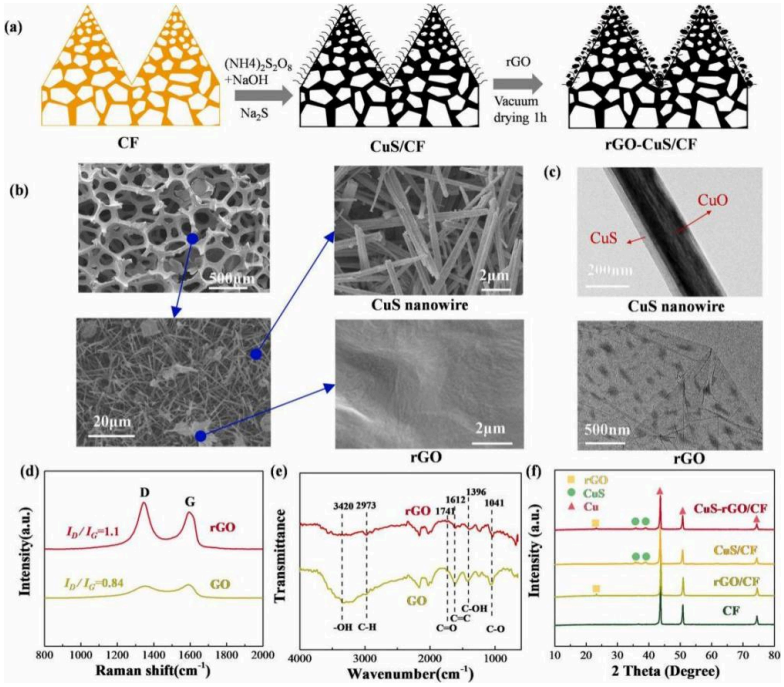

Figure 7: Construction of hierarchically porous architectures using CuS-rGO composite photothermal materials [27]

5.2. Zinc-induced 3D MXene/rGO composites for supercapacitor electrodes

Yang et al. fabricated a three-dimensional porous MXene/rGO architecture (PMG) with enhanced oxidation resistance, utilizing zinc powder as both a sacrificial template and reducing agent, where Zn^{2+} ions served as structural linkers to enable MXene-rGO nanosheet interlocking [28]. This design suppressed layer stacking between MXene and rGO while outperforming Zn-MXene counterparts in oxidative stability. The interconnected porous framework provided enhanced electrical conductivity, abundant active sites, and reduced ion diffusion distances. Configured as an asymmetric supercapacitor (PMG-5//NHRGO ASC) with N-doped porous RGO (NHRGO) cathodes, the device achieved a high energy density and robust cycling stability.

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6. Conclusion

Recent advancements in rGO composite research have established three-dimensional and porous architectures as primary structural forms, replacing outdated two-dimensional configurations. Traditional immobilized loading modes face limitations due to structural rigidity and insufficient active sites, while encapsulated, sandwich-like, and hybrid configurations dominate current studies owing to their adaptability and enhanced performance. Exploring bimetallic and multimetallic oxides alongside related compounds such as sulfides and selenides for rGO composites remains crucial to unlock their full potential.

rGO's structural diversity and processing flexibility make it indispensable in multicomponent composite systems, yet its limited solvent dispersibility restricts synthesis methods and applications. Addressing this requires focused efforts to improve rGO's dispersibility in diverse solvents at higher concentrations and to innovate assembly strategies that bypass current fabrication bottlenecks.

Innovative structural designs combining rGO with advanced functional materials have proven highly valuable. This approach will undoubtedly drive future research, particularly in developing next-generation composites with tailored properties for energy storage and conversion technologies.

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