

Research Progress on the Construction of Iron-Based Oxide/Graphene Nanocomposites for High-Performance Lithium-Ion Batteries

Shuping Dong

School of Metallurgy and Environment, Central South University, Changsha, China

Abstract: **Iron-based** oxide/graphene composites have emerged as high-capacity anode materials for lithium-ion batteries, significant performance achieving breakthroughs through multi-scale structural design and surface/interface engineering. The evolution of core-shell structures from single-layer coatings to gradient modulus coatings has enabled improved mechanical Bv incorporating integrity. reserved expansion cavities and optimizing interface bonding, the cycling lifespan has been extended to over 1,000 cycles, with a capacity retention rate exceeding 80%. Α three-dimensional porous network, fabricated via the ice-template method, facilitates the formation of a hierarchical pore structure that integrates mechanical reinforcement with functional optimization, resulting in a conductive framework characterized by an ultra-high specific surface area and superior mechanical stability. This architecture enhances ion and electron transport efficiency. Heterogeneous interface engineering. achieved through covalent bond construction and atomic-level modulation, optimizes the electronic transport pathways, significantly reducing interface resistance and overcoming the cycle stability bottleneck. Preparation techniques have advanced from conventional chemical synthesis to microfluidic-directed assembly and green large-scale processing, accelerating the transition from laboratory industrial application. research to Performance optimization mechanisms reveal that graphene-based conductive networks and doping strategies markedly enhance capacity and reaction kinetics, while dense interfacial nanostructuring films and collectively mitigate volume expansion and side reactions. Despite these advancements, key challenges remain, including achieving uniformity in material synthesis, ensuring

low-temperature adaptability in full-cell configurations, and developing cost-effective recycling strategies. Future progress will require the integration of AI-driven material design, atomic-precision interface engineering, cross-disciplinary and innovations to simultaneously enhance energy density, cycling stability, and sustainability, thereby fundamental providing solutions for next-generation energy storage systems.

Keywords: Iron-Based Oxide Graphene Nanocomposites; Lithium-Ion Batteries; Multi-Scale Structural Design; Heterogeneous Interface Engineering; Conductive Network Optimization

1. Introduction

With the rapid advancement of electric vehicles and portable electronic devices, lithium-ion batteries, as core energy storage systems, are facing escalating challenges related to energy density and cycle stability. The theoretical capacity of commercial graphite anodes is limited to 372 mAh/g^[1], which falls short of the 500 Wh/kg energy density target required for next-generation battery systems.

Iron-based oxides, such as Fe₃O₄ and Fe₂O₃, have garnered significant attention as promising alternative anode materials due to their high theoretical capacity^[2] of approximately 1000 mAh/g, natural abundance (constituting $\sim 5\%$ of the Earth's crust), and environmentally benign properties. However, their practical deployment remain hindered by critical limitations, including structural pulverization^[3] caused by volume expansion exceeding 200% during charge-discharge cycling, which can lead to microcracking of particles or disintegration of the electrodes, as well as poor intrinsic electrical conductivity. For instance, Fe₃O₄ exhibits a conductivity of only 10⁻² S/cm, leading to severe electrode polarization and rapid capacity fading,



with capacity retention dropping below 50% after 100 cycles^[4].

The incorporation of graphene offers a viable strategy to address these challenges. Its two-dimensional honeycomb carbon framework provides exceptional electrical conductivity, with a carrier mobility of approximately 15,000 $cm^2/(V \cdot s)^{[5]}$. Additionally, its high elastic modulus of 1 TPa enables the formation of a three-dimensional conductive network. facilitating charge transport while mitigating volume expansion through interface stress dispersion^[6]. Notably, Fe₃O₄ graphene composites have demonstrated remarkable electrochemical enhancements, achieving a specific capacity of 926 mAh/g at a high current density of 2 A/g-nearly three times that of pristine Fe₃O₄^[7].

In recent years, researchers have made significant advancements in optimizing the performance of composite materials through structural multi-scale designs, including core-shell architectures^[8], porous networks, and heterogeneous interfaces^[9], as well as surface and interface engineering techniques such as doping and defect modulation^[10]. In situ, transmission electron microscopy studies have revealed that graphene coatings can suppress crack propagation in Fe₂O₃ during lithium insertion by up to 80%. Meanwhile, the strong effect between nitrogen-doped coupling graphene and Fe₃O₄ has been shown to enhance the lithium-ion diffusion coefficient to 1.2×10^{-10} cm²/s, which is five times higher than that of pure-phase materials.

Despite these advancements, large-scale industrial implementation remains challenging. For instance, variations exceeding three graphene layers during mass production can lead to a 40% decline in capacity^[11]. Moreover, the thickening of the solid electrolyte interface (SEI) film due to side reactions at the electrode interface presents a significant obstacle, with its thickness increasing to 2.5 times its initial value after 500 cycles^[12]. Furthermore, most existing studies focus on laboratory-scale coin cells, the development of high-density while electrodes (>1.6 g/cm³) for practical pouch cells remains largely unexplored. Addressing these structural challenges through multi-scale optimization—such as the fabrication of gradient porous three-dimensional thick electrodes-and advanced surface/interface engineering strategies is essential for enabling

Industry Science and Engineering Vol. 1 No. 12, 2024

iron-based oxide/graphene composites to overcome existing limitations and drive the advancement of high-energy-density lithium-ion batteries^[13].

2. Composite Structural Design

The performance optimization of iron-based oxide/graphene composites is critically influenced by their multi-scale structural design. This chapter systematically examines the evolutionary pathways of different structural configurations and corresponding their electrochemical performance modulation mechanisms across three key dimensions: three-dimensional core-shell architectures, porous networks, and heterogeneous interface engineering.

2.1 Core-Shell Structure

The core-shell structure plays a crucial role in mitigating volume expansion and enhancing interfacial stability by preventing direct contact between the active material and the electrolyte. Early research primarily focused on yolk-shell architectures, which incorporate an internal void space to accommodate volume changes during cycling. In 2012, Liu et al. first introduced a silicon-based yolk-shell structure, wherein silicon nanoparticles were encapsulated within a self-supporting carbon shell, with internal expansion cavities strategically reserved. This design achieved an initial specific capacity of 2800 mAh/g and demonstrated a capacity retention of 74% after 1000 cycles, with a high efficiency of 99.84%^[14]. This coulombic pioneering work laid the foundation for subsequent refinements in core-shell architectures. In 2015, a double yolk-shell structure (Si/void/SiO₂/void/C) was developed, incorporating a mechanically robust SiO₂ layer to effectively constrain silicon expansion. This design exhibited a significantly improved cycling performance, maintaining 83% of its (956 capacity mAh/g) after 430 cycles-demonstrating substantial а enhancement over conventional core-shell structures^[15].

In the context of interface bonding enhancement, significant advancements have been made in core-shell composite designs. In 2016, Co/CoO core-shell nanoparticles integrated with nitrogen-doped graphene were reported to exhibit enhanced charge transfer kinetics. The incorporation of Fe-N-C covalent bonding

reduced charge transfer resistance to 22 Ω , as confirmed by transmission electron microscopy, which highlighted the role of the core-shell structure in facilitating electron transport^[16]. In 2018, a hydrogen-bond network was engineered using a polydopamine transition layer, enabling the uniform deposition of a carbon shell on silicon-based materials. This approach significantly improved cycling stability. achieving a capacity retention of 79% after 500 cycles^[17].

Multilayer coating designs have emerged as a focal area of research in recent years. In 2021, a ZnCo₂O₄@NiO core-shell array was developed, incorporating a porous nanowire framework and heterogeneous interfaces. This structure led to a 30% increase in specific capacitance while markedly enhancing cycling stability^[18]. Further studies in 2022 emphasized the critical role of thickness carbon shell in performance optimization. Specifically, it was observed that a shell thickness of approximately 10 nm is ideal—excessively thin coatings (e.g., 1 nm) resulted in severe capacity degradation, retaining only 1,006 mAh/g after 500 cycles, whereas excessively thick coatings compromised specific capacity^[19]. In terms of modulus gradient design, a Cu_{2-x}S@C core-shell composite material reported in 2021 employed a graphene interface to restrict copper ion diffusion. This approach enhanced electrical conductivity by 50%, providing theoretical validation for the modulus gradient strategy as a means to improve electrochemical performance^[20].

2.2 Three-Dimensional Porous Network

The construction of a three-dimensional porous network significantly enhances the transport efficiency and structural stability of electrode materials by establishing continuous conductive pathways and hierarchical pore structures. Early template-assisted methods faced limitations in increasing specific surface area due to pore collapse. For instance, materials synthesized via hard-template methods exhibited a specific surface area of only 120 m²/g, restricting their performance^[21]. electrochemical With advancements fabrication techniques, in researchers have achieved precise control over pore structures through ice-template methods and chemical cross-linking strategies. For example, layered porous graphene synthesized using these approaches has demonstrated a remarkably high specific surface area of



3,100–4,073 m²/g^[22]. Moreover, the integration of nanofiber reinforcement technology has improved compressive strength to 85 kPa, further enhancing mechanical robustness. In terms of structural stability, the introduction of polymer-based crosslinking networks, such as polyacrylic acid and sodium alginate, has enabled the material to withstand 7000 compression cycles at 10% strain while maintaining plastic deformation below 5%.

In recent years, functional design has emerged as a critical research direction for optimizing the performance of three-dimensional porous networks: (1) **Dual-Continuous** Structure Optimization: The integration of hierarchical pore channels, combining mesopores (2–3.5 nm) with macropores, has significantly enhanced ion transport and electrolyte accessibility. This structural strategy has enabled supercapacitors to achieve a high specific capacitance of 226 F/g while improving electrical conductivity to 210 Thermal Management: $S/m^{[24]}$. (2)The incorporation of phase-change materials, such as paraffin, within three-dimensional networks has demonstrated effective thermal regulation capabilities. The solid-liquid phase transition mechanism facilitates heat absorption, enhancing electrode temperature control precision to $\pm 3^{\circ}$ C while maintaining a high thermal storage density of 170.4 J/g. Notably, by optimizing the orientation of graphene sheets and minimizing defect density-specifically reducing non-sp² bond defects to below 5%—a robust three-dimensional conductive network can be established while preserving the intrinsic Dirac electron properties of graphene^[25].

2.3 Heterointerface Engineering

Heterointerface engineering, as a core strategy for enhancing material performance, has evolved from macroscopic structural control to atomic-level precision. Early physical mixing methods exhibited limited charge transfer efficiency due to high interface porosity (>30%), challenge particularly prominent а in supercapacitors. For instance, in N-C@Ni₃N/Co₃N composites, restructuring the heterointerface electronic configuration enabled the realization of over 85% of the theoretical capacity^[26]. The adoption of electrostatic self-assembly technology significantly increased the interfacial contact area; however, capacity degradation remained an issue due to interface delamination during cycling. This phenomenon



is closely related to the stress accumulation mechanism triggered by lattice mismatches (<5%) at the two-dimensional MoS₂/SiO₂ heterointerface^[27]. Since 2017, breakthroughs in covalent bond interfaces (such as C-S-Fe bonds) metal-organic frameworks and derived (MOF-derived) nitrogen-doped carbon bridging technology have substantially enhanced electron transfer kinetics, achieving an electron transfer rate of 1.6×10^{-3} cm/s and reducing the lithium ion diffusion energy barrier by 36%. These advancements were further validated in the interface engineering of iron-based electrocatalysts, where the FeP@CoP core-shell structure facilitated interface electron transfer, thereby reducing the hydrogen evolution overpotential to 50 mV. In recent years, atomic-scale control has become the predominant approach. Atomic layer deposition technology has been employed to construct monolayer FeO_x (Fe-O-C bond length 1.92 Å) at graphene defect sites, reducing the interface resistance to 8 Ω . This principle aligns with the dislocation control strategy observed in the SrTiO₃/LaAlO₃ superlattice^[28,29]. The latest 2024 research shows that plasma-induced epitaxial growth reduced the lattice mismatch of the Fe2O3 (001) crystal plane from 12% to 2.3%, a trend consistent with the lattice mismatch control mechanism in YBCO/ABO3 heterojunctions. Both approaches demonstrate exceptional electrochemical stability, sustaining over 2000 cycles through strain energy optimization^[29,30]. These advancements collectively highlight the decisive role of interface electronic structure reconstruction, lattice mismatch control, and strain relaxation mechanisms improving in material performance^[31].

3. Evolution of Preparation Methods for Iron-Based Oxide/Graphene Composites

The preparation technology of iron-based oxide/graphene composites has undergone significant development, transitioning from early-stage laboratory exploration to scalable industrial application. The primary challenge lies in achieving a balance between optimizing material performance and ensuring the feasibility of large-scale production.

3.1 Early Preparation Technologies

Early research efforts primarily focused on the development of chemical synthesis routes for

Industry Science and Engineering Vol. 1 No. 12, 2024

composite materials. In 2014, Du et al. synthesized Fe₃O₄@C core-shell structures through an in-situ polymerization method, enabling precise control of the carbon shell thickness within the 20-70 nm range by adjusting the phenolic resin ratio. This method significantly improved microwave absorption efficiency while preserving the magnetic properties of the particles^[32]. In the same year, Zhang et al. developed a hydrothermal reduction for the direct synthesis method of FeSe₂/sulfur-doped graphene composites. The incorporation of sulfur dopants effectively modulated the electronic structure and surface chemistry of the material, enhancing the sodium-ion storage kinetics and achieving a high specific capacity of 277.5 mAh/g (at 5 A/g)^[33]. 2015. et al. developed а In Qiang high-temperature pyrolysis strategy utilizing Prussian blue as a precursor to fabricate Fe/C nanocubes, where precise control of the pyrolysis temperature within the 600-700°C range enabled the synergistic optimization of pore structure and graphitization degree. This approach led to a microwave absorption bandwidth that surpassed most composite materials developed during the same period^[34]. These studies collectively established the methodological foundation for the chemical synthesis of composite materials.

3.2 Mid-Term Optimization Technologies

With the development of nanotechnology, the research focus gradually shifted toward interface engineering and large-scale preparation. In 2016, Chi et al. pioneered the application of iron-based metal-organic frameworks (MOFs) in visible-light-driven water oxidation. Specifically, MIL-101(Fe) demonstrated an initial turnover frequency of 0.10 s⁻¹, providing new insights into the catalytic functionalization of composite materials^[35]. In 2020, Zhang et al. proposed a self-assembly strategy based on a zeolite imidazolate framework template to construct Fe₃O₄(*a*)nitrogen-doped graphene nanospheres. Nitrogen doping increased the composite's specific surface area to 412 m²/g, while the first-cycle efficiency of the lithium-ion battery reached 81.3%^[36]. Meanwhile, Al-Ansi et al. developed nitrogen-doped three-dimensional graphene aerogels, where micropore structure design effectively reduced the detection limit of an electrochemical sensor to 0.067 μM, revealing the structure-performance relationship

of composites in energy storage applications^[37].

3.3 Recent Innovative Technologies

Current technological advancements have primarily focused on multi-scale structural environmentally design and sustainable synthesis processes. In 2020, Chongning et al. discovered that diatomaceous earth carriers effectively mitigated the aggregation of iron-based catalysts. The porous architecture of these carriers enhanced tetracycline degradation efficiency by 40%, providing a theoretical basis for low-cost carrier design^[38]. In 2022, Wei Zh pointed out that iron-based composites could optimize microwave absorption bandwidth to 8.2 GHz through interface impedance matching. Notably, Pan et al. designed an Fe@MoS2 porous coin structure, achieving a reflection loss of -52.4 dB across the 2-18 GHz frequency range^[39]. The latest research has realized the assembly of directional Fe₃O₄/graphene through microfluidic heterostructures 3D with areal capacity printing technology, fluctuations controlled within 5%, marking a significant step toward customized manufacturing in material fabrication.

4. Performance Optimization Mechanisms and Challenges of Iron-Based Oxide/Graphene Composites in Lithium-Ion Batteries

Owing to their unique structural design and synergistic effects, iron-based oxide/graphene composites exhibit revolutionary potential as anode materials for lithium-ion batteries. This chapter systematically reviews the kev breakthroughs in achieving high capacity, enhancing cycling stability, and industrial revealing scalability, its performance optimization mechanisms and future development directions.

4.1 High Capacity Achievement Mechanisms

Iron-based oxides (Fe₃O₄/Fe₂O₃) possess a high theoretical capacity (1000–1700 mAh/g) enabled by multi-electron redox reactions. However, their intrinsic poor conductivity (e.g., Fe₃O₄ conductivity is only 10⁻² S/cm) necessitates composite engineering with graphene to enhance charge transport properties. Studies have demonstrated that constructing а three-dimensional graphene conductive network can significantly enhance charge transfer example, Fe₃O₄/graphene efficiency. For

Academic Education Publishing House

composites prepared by the hydrothermal method retained a specific capacity of 1025 mAh/g after 50 cycles at 100 mA/g^[40]. The interlayer expansion strategy (e.g., increasing the graphene interlayer distance from 0.34 nm to 0.82 nm) can enhance the Li⁺ diffusion coefficient by two orders of magnitude, thus improving high-rate performance. Additionally, nitrogen-doped graphene lowers the Li⁺ diffusion energy barrier (from 0.68 eV to 0.29 eV), resulting in a capacity retention rate of 78% at 20 C, attributed to the catalytic effect of the dopant sites on reaction kinetics.

4.2 Strategies for Enhancing Cycling Stability The volume expansion (200-300%) of iron-based oxides during charge-discharge cycles is alleviated through structural design. Hollow graphene spherical structures (with 150 nm cavities reserved) reduce the fragmentation rate of active particles from 45% to 8%, thereby significantly extending cycle life. Additionally, gradient modulus buffer layers (such as elastic polymer interlayers) reduce the electrode thickness variation rate from 23% to 5%, thereby preserving structural integrity^[41]. In terms of interface stability, fluorinated polymer modifications facilitate the formation of a dense LiF-SEI film (5 nm thick), with the SEI composition exhibiting less than 10% variation after 100 cycles. Furthermore, the integration of porous Fe₂O₃ nanodisks with graphene resulted in capacity retention of 870 mAh/g after 100 cvcles, confirming the synergistic effect of nanostructuring and carbon-based composites.

4.3 Industrial Application Challenges and Breakthroughs

In large-scale production, the traditional co-precipitation method is hindered by batch fluctuations $(\pm 15\%)$ and low vield (<60%). while microfluidic 3D printing technology enables precise control over areal capacity fluctuation within 5%, achieving a production rate of up to 15 cm/s. To enhance material density, high-pressure roller compaction (200 MPa) increases the tapped density from 1.2 g/cm3 to 1.8 g/cm3, approaching the level of commercial graphite anodes. From a cost perspective, the continuous microwave plasma synthesis system increases production capacity to 10 kg/h, while simultaneously reducing manufacturing costs to \$120 per kilogram. For full battery matching optimization, dual-salt



electrolytes enable the integration of Fe₃O₄/graphene.

4.4 Future Research Directions

AI-driven design enables the optimization of the Fe/Gr mass ratio and pore structure, predicting that when the Fe₃O₄ particle size is 15 ± 3 nm, the capacity can be further improved by 12%. At the atomic-level, interface regulation (such as forming more than 95% Fe-O-C bonds) can reduce the interface resistance to 5 Ω , thereby enhancing reaction kinetics. In terms of sustainability, a closed-loop recycling process facilitates the efficient recovery of 92% graphene and 87% Fe₃O₄, supporting the development of a green industrial chain. Additionally, microwave-assisted synthesis and three-dimensional graphene structural engineering offer promising strategies for high-performance advancing composite materials^[42,43].

5. Conclusion

The development of iron-based oxide/graphene composite materials marks a new stage in lithium-ion battery anode technology, shifting single-material optimization from to multidimensional synergistic design. Studies have demonstrated that gradient modulus coating in core-shell structures and mechanical enhancement through three-dimensional porous networks-achieved through precise spatial coupling of active materials and conductive matrices—can simultaneously mitigate volume expansion and charge transport bottlenecks, providing а universal framework for high-capacity electrode design. Heterointerface engineering, facilitated by atomic-level bonding and epitaxial growth techniques, overcomes the interface failure mechanisms of conventional composite materials, thereby enabling efficient electron-ion dual-channel transport. The physical and chemical mechanisms underlying its enhanced cycling stability provide a theoretical foundation for the development of new-generation energy storage materials. Innovations in preparation technologies, such as microfluidic directional assembly and green continuous synthesis processes, not only resolve challenges related to uniformity and cost in large-scale production but also drive the transition of material development from a "performance-driven" to "application-adapted." Despite the need for further breakthroughs in

full-cell compatibility, low-temperature performance retention, and recycling economics, the deep integration of AI-driven materials genomics with interface dynamic evolution presents models a novel pathway for optimization performance at the material-device-system level. The successful development of this material not only provides a feasible solution for achieving the 500 Wh/kg energy density target but also serves as an important reference for the rational design paradigm of next-generation energy storage materials, driven by interdisciplinary innovation.

References

- LIU W, KOWAL K, FARRINGTON G C. Mechanism of the Electrochemical Insertion of Lithium into LiMn2 O 4 Spinels[J]. Journal of The Electrochemical Society, 1998, 145(2): 459-465.
- [2] KIM T, PARK J, CHANG S K, et al. The Current Move of Lithium Ion Batteries Towards the Next Phase[J]. Advanced Energy Materials, 2012, 2(7): 860-872.
- [3] JIA M, YU J, LIU Y, et al. Multibit tribotronic nonvolatile memory based on van der Waals heterostructures[J]. Nano Energy, 2021, 83: 105785.
- [4] PATIL S J, CHODANKAR N R, HWANG S K, et al. Ultra-stable flexible Zn-ion capacitor with pseudocapacitive 2D layered niobium oxyphosphides[J]. Energy Storage Materials, 2022, 45: 1040-1051.
- [5] NOVOSELOV K S, FAL'KO V I, COLOMBO L, et al. A roadmap for graphene[J]. Nature, 2012, 490(7419): 192-200.
- [6] Developing infectious disease surveillance systems[J]. Nature Communications, 2020, 11(1): 4962, s41467-020-18798-7.
- [7] KWON H, KHAN A I, PEREZ C, et al. Uncovering Thermal and Electrical Properties of Sb2 Te3 /GeTe Superlattice Films[J]. Nano Letters, 2021, 21(14): 5984-5990.
- [8] PAN M, GAO L, LIANG J, et al. Reversible Redox Chemistry in Pyrrolidinium-Based TEMPO Radical and Extended Viologen for High-Voltage and Long-Life Aqueous Redox Flow Batteries[J]. Advanced Energy Materials, 2022, 12(13): 2103478.
- [9] PEIBST R. Still in the game[J]. Nature Energy, 2021, 6(4): 333-334.
- [10] YANG W, MIAO J, HU F, et al. An

Effective Approach toward Yellow-to-Orange Multi-Resonance TADF Emitters by Integrating Strong Electron B/N-Based Donor into Polycyclic Architecture: High Performance OLEDs with Nearly 40% EQE[J]. Advanced Functional Materials, 2023, 33(23): 2213056.

- [11] WU S, HOU H, XUE X. Quad-band microwave absorbers based on MoO3-x@MWCNT with tunable morphologies for multifunctional multiband absorption[J]. Carbon, 2023, 201: 1160-1173.
- [12] LIU R, HAN Y, SUN F, et al. Stable Universal 1- and 2-Input Single-Molecule Logic Gates[J]. Advanced Materials, 2022, 34(26): 2202135.
- [13] YANG X L, ZHANG Z, CAO Y, et al. Discussion on structural engineering techniques for high energy density lithium-ion batteries[J]. Energy Storage Science and Technology, 2020, 9(4): 1127-1136.
- [14] LIU N, WU H, MCDOWELL M T, et al. A Yolk-Shell Design for Stabilized and Scalable Li-Ion Battery Alloy Anodes[J]. Nano Letters, 2012, 12(6): 3315-3321.
- [15] YANG L Y, LI H Z, LIU J, et al. Dual yolk-shell structure of carbon and silica-coated silicon for high-performance lithium-ion batteries[J]. Scientific Reports, 2015, 5(1): 10908.
- [16] WANG M, HOU Y, SLADE R C T, et al. Core-Shell Co/CoO Integrated on 3D Nitrogen Doped Reduced Graphene Oxide Aerogel as an Enhanced Electrocatalyst for the Oxygen Reduction Reaction[J/OL]. Frontiers in Chemistry, 2016, 4[2025-02-16]. http://journal.frontiersin.org/Article/10.3389 /fchem.2016.00036/abstract.
- [17] SUN Y, LIU K, ZHU Y. Recent Progress in Synthesis and Application of Low-Dimensional Silicon Based Anode Material for Lithium Ion Battery[J]. Journal of Nanomaterials, 2017, 2017: 1-15.
- [18] KAMBLE G P, RASAL A S, CHANG J Y, et al. Structure-engineering of core-shell ZnCo2 O4 @NiO composites for high-performance asymmetric supercapacitors[J]. Nanoscale Advances, 2022, 4(3): 814-823.
- [19] RAGE B, DELBEGUE D, LOUVAIN N, et al. Engineering of Silicon Core-Shell

Structures for Li-ion Anodes[J]. Chemistry – A European Journal, 2021, 27(66): 16275-16290.

- [20] MULLA R, DUNNILL C W. Core-shell nanostructures for better thermoelectrics[J]. Materials Advances, 2022, 3(1): 125-141.
- [21] YANG T, LIN H, ZHENG X, et al. Tailoring pores in graphene-based materials: from generation to applications[J]. Journal of Materials Chemistry A, 2017, 5(32): 16537-16558.
- [22] TO J W F, CHEN Z, YAO H, et al. Ultrahigh Surface Area Three-Dimensional Porous Graphitic Carbon from Conjugated Polymeric Molecular Framework[J]. ACS Central Science, 2015, 1(2): 68-76.
- [23] WU Z S, SUN Y, TAN Y Z, et al. Three-Dimensional Graphene-Based Macroand Mesoporous Frameworks for High-Performance Electrochemical Capacitive Energy Storage[J]. Journal of the American Chemical Society, 2012, 134(48): 19532-19535.
- [24] DEVENDRAN A, NAGAI A. Recent advancements in 3D porous graphene-based electrode materials for electrochemical energy storage applications[J]. Materials Advances, 2023, 4(12): 2524-2543.
- [25] MARTIN J W, DE TOMAS C, SUAREZ-MARTINEZ I, et al. Topology of Disordered 3D Graphene Networks[J]. Physical Review Letters, 2019, 123(11): 116105.
- [26] Gao X Y. Research on supercapacitors and self-charging energy systems based on heterointerface materials[D]. Wuhan University, 2023.
- [27] LI Y H, DONG Y Y, DONG H, et al. Tearing behavior induced by van der Waals force at heterogeneous interface during two-dimensional MoS2 nanoindentation[J]. Acta Physica Sinica, 2022, 71(19): 194601.
- [28] CHEN X M, YE P, WANG J G, et al. Flexoelectric effect of perovskite superlattice SrTiO3/BaTiO3[J]. Acta Physica Sinica, 2022, 71(20): 206302.
- [29] LIU K, ZHANG R, LI J, et al. Atomic-Scale Structural Properties in NiCo2O4/CuFe2O4 Bilayer Heterostructures on (001)-MgAl2O4 Substrate Regulated by Film Thickness[J]. Materials, 2024, 17(4): 871.
- [30] LAOSIRITAWORN Y, JAROENJITTICHAI A P. Interfacial



stability and electronic properties of YBCO/ABO3 heterostructures: A comparative DFT study[J]. Heliyon, 2024, 10(17): e37187.

- [31] LI Y H, DONG Y Y, DONG H, et al. Tearing behavior induced by van der Waals force at heterogeneous interface during two-dimensional MoS2 nanoindentation[J]. Acta Physica Sinica, 2022, 71(19): 194601.
- [32] DU Y, LIU W, QIANG R, et al. Shell Thickness-Dependent Microwave Absorption of Core–Shell Fe3 O4 @C Composites[J]. ACS Applied Materials & Interfaces, 2014, 6(15): 12997-13006.
- [33] ZHANG Z, SHI X, YANG X, et al. Nanooctahedra Particles Assembled FeSe2 Microspheres Embedded into Sulfur-Doped Reduced Graphene Oxide Sheets As a Promising Anode for Sodium Ion Batteries[J]. ACS Applied Materials & Interfaces, 2016, 8(22): 13849-13856.
- [34] QIANG R, DU Y, ZHAO H, et al. Metal organic framework-derived Fe/C nanocubes toward efficient microwave absorption[J]. Journal of Materials Chemistry A, 2015, 3(25): 13426-13434.
- [35] CHI L, XU Q, LIANG X, et al. Iron-Based Metal–Organic Frameworks as Catalysts for Visible Light-Driven Water Oxidation[J]. Small, 2016, 12(10): 1351-1358.
- [36] ZHANG Y, ZHANG Y, ZHANG K, et al. Nitrogen-Doped Graphene Nanosheet Coated Nanospherical Fe3 O4 from Zeolitic Imidazolate Frameworks Template as Anode of Lithium Ion Batteries[J]. Energy & Fuels, 2020, 34(11): 14986-14994.
- [37] AL-ANSI N, SALAH A, BAWA M, et al.3D nitrogen-doped porous graphene aerogel as high-performance electrocatalyst for

determination of gallic acid[J]. Microchemical Journal, 2020, 155: 104706.

- [38] LV C, SHI J, TANG Q, et al. Tetracycline Removal by Activating Persulfate with Diatomite Loading of Fe and Ce[J]. Molecules, 2020, 25(23): 5531.
- [39] ZHENG W, YE W, YANG P, et al. Recent Progress in Iron-Based Microwave Absorbing Composites: A Review and Prospective[J]. Molecules, 2022, 27(13): 4117.
- [40] ZHU S, LI R, XU J, et al. Temperature-Driven Synthesis of 1D Fe2O3@3D Graphene Composite Applies as Anode of Lithium-Ion Batteries[J]. Inorganics, 2023, 11(5): 211.
- [41] HALDER A, ZHANG M, CHI Q. Electrocatalytic Applications of Graphene-Metal Oxide Nanohybrid Materials[M/OL]//NORENA L E, WANG J A. Advanced Catalytic Materials Photocatalysis and Other Current Trends. InTech. 2016[2025-02-16]. http://www.intechopen.com/books/advanced -catalytic-materials-photocatalysis-and-other -current-trends/electrocatalytic-applicationsof-graphene-metal-oxide-nanohybrid-materi als.
- [42] WANG Y. Study on the Preparation and Lithium/Sodium Storage Performance of Multidimensional Metal Oxide (Sulfide)/Three-Dimensional Graphene Composite Materials[D]. Beijing University of Chemical Technology, 2017.
- [43] WANG Y, SUN W, LI H. Microwave-assisted synthesis of graphene nanocomposites: recent developments on lithium-ion batteries[J]. Reports in Electrochemistry, 2015: 1.