

CRITICAL REVIEW

View Article Online
View Journal



Cite this: DOI: 10.1039/d5va00319a

Magnetic graphene nanocomposites: a new frontier in radioactive waste remediation

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The management of radioactive waste presents formidable environmental and health challenges, necessitating the development of effective remediation technologies. Magnetic nanocomposites (NCs) derived from iron oxide (Fe₃O₄) and graphene derivatives have emerged as highly promising materials for the sorptive removal of radionuclides from contaminated aqueous streams. This comprehensive review critically examines the synthesis, characterization, and application of these NCs. The key physicochemical properties—including structural, magnetic, and surface characteristics—that underpin their high sorption capacities have been explored. The discussion covers various synthesis methodologies and the analytical techniques used to validate the properties of the materials. A central focus is placed on the sorption mechanisms, performance efficiency, and the operational factors influencing the sequestration of radioactive ions. Despite their significant potential, several challenges related to scalability, long-term stability, selective sorption in complex matrices, and potential environmental impacts have been identified and discussed. Finally, future research directions to advance the practical application of Fe₃O₄@graphene NCs in radioactive waste management have been discussed. This review provides a foundational understanding of the capabilities and limitations of these materials, aiming to guide future research toward their practical implementation in mitigating the hazards of radioactive contamination.

Received 10th September 2025
Accepted 7th January 2026

DOI: 10.1039/d5va00319a

rsc.li/esadvances

Environmental significance

Radioactive contamination from nuclear activities poses a severe and persistent threat to ecosystems and human health. This work reviews the significant advancements in magnetic graphene nanocomposites as a superior alternative to conventional approaches for radionuclide removal from contaminated water. These nanomaterials exhibit high sorption capacity and can be easily separated from water using a magnetic field, allowing for efficient reuse. The technology offers a transformative approach to radioactive waste management, providing an effective and sustainable solution to mitigate the long-term environmental hazards of nuclear contamination, thereby protecting natural environments and safeguarding public health from the risks of radiation exposure.

1.0 Introduction

The proliferation of anthropogenic radionuclides in the environment represents a persistent and insidious global threat. Arising from historical nuclear weapons testing and both routine and accidental releases from the expanding global network of nuclear facilities, these contaminants pose a long-term risk to ecosystems and human health.^{1–6} Catastrophic

events, such as those at Kyshtym (1957), Chornobyl (1986), and Fukushima Daiichi (2011), serve as stark reminders of the potential for widespread and lasting environmental contamination.^{6–9} Compounding this issue is the vast accumulation of legacy radioactive waste in nations with established nuclear programs, including the United States, Russia, and the United Kingdom.^{2,3} Once released, radionuclides can enter the food chain, where they bioaccumulate and biomagnify, posing a severe radiological risk to human health even from initially trace concentrations.^{4,6,10–12}

Consequently, the safe and effective management of radioactive waste has become a paramount challenge at the intersection of nuclear technology, environmental science, and materials chemistry.^{13–17} The extreme longevity of many radionuclides, which can remain hazardous for millennia, necessitates containment strategies that are not only robust but also sustainable over geological timescales.^{11,18,19} Traditional

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Table 1 Comparison of key remediation techniques for radioactive waste management

| Technique | Action | Advantages | Disadvantages |
|------------------------|--|--|--|
| Chemical precipitation | Converts soluble radionuclides into insoluble precipitates <i>via</i> the addition of chemical reagents | <ul style="list-style-type: none"> • High efficiency for high-concentration waste • Cost-effective for treating large volumes | <ul style="list-style-type: none"> • Generates substantial volumes of secondary radioactive sludge • Low efficiency for removing trace-level or low-concentration radionuclides • High cost of resins |
| Ion exchange | Utilizes solid materials (<i>e.g.</i> , resins or zeolites) to reversibly exchange non-radioactive ions for radionuclide ions in the liquid phase | <ul style="list-style-type: none"> • High selectivity and removal efficiency • Effective for large volumes of low-salinity aqueous waste | <ul style="list-style-type: none"> • Regeneration produces concentrated liquid secondary waste • Performance is susceptible to competing non-radioactive ions • High energy consumption due to operating pressure requirements • Prone to membrane fouling, necessitating frequent maintenance • Generates concentrated retentate streams requiring further treatment • Challenges in maintaining selectivity in complex matrices containing competing ions • Finite sorption capacity requires periodic regeneration or replacement of the sorbent |
| Membrane separation | Employs semi-permeable barriers to separate contaminants based on size exclusion or ionic charge | <ul style="list-style-type: none"> • Versatile removal of particulates, colloids, and ions • Operates at ambient temperatures | <ul style="list-style-type: none"> • Performance is susceptible to competing non-radioactive ions • High energy consumption due to operating pressure requirements • Prone to membrane fouling, necessitating frequent maintenance • Generates concentrated retentate streams requiring further treatment • Challenges in maintaining selectivity in complex matrices containing competing ions • Finite sorption capacity requires periodic regeneration or replacement of the sorbent |
| Sorption | Uses porous solid materials (<i>e.g.</i> , graphene nanocomposites) to capture and bind radioactive ions from solution | <ul style="list-style-type: none"> • Minimal sludge generation compared to precipitation • Superior for “polishing” effluents with trace radionuclide concentrations (<1 ppm) • Rapid kinetics and energy-efficient separation • Amenable to regeneration or immobilization for long-term safe disposal | <ul style="list-style-type: none"> • Minimal sludge generation compared to precipitation • Superior for “polishing” effluents with trace radionuclide concentrations (<1 ppm) • Rapid kinetics and energy-efficient separation • Amenable to regeneration or immobilization for long-term safe disposal |

immobilization techniques, such as vitrification and cementation, are energy-intensive and designed primarily for high-level waste. They are often limited by high operational costs, process inefficiencies, and a lack of versatility in treating the large volumes of low-level, chemically diverse contaminated water generated by nuclear reactors, medical isotope production, and industrial applications.^{4,16,20,21}

Chemical precipitation, ion exchange, membrane separation, and sorption are considered key remediation techniques for radionuclide removal.^{22–24} A detailed comparison of these methods clearly articulates the scientific rationale for sorption's superiority, particularly its efficacy in polishing effluent streams and minimizing secondary waste (Table 1).

In response to these limitations, advanced materials science offers a promising paradigm shift, moving from passive containment to active treatment and removal.^{6,11,19,25–27} Among the various innovations, magnetic nanocomposites (NCs) have received significant attention for their unique multifunctional properties, including exceptionally high specific surface area, robust mechanical strength, and, critically, facile separation from solution *via* an external magnetic field.^{16,25,26,28} Specifically, NCs that integrate the magnetic responsiveness of iron oxide (Fe_3O_4) nanoparticles (NPs) with the unparalleled surface properties of graphene derivatives have demonstrated exceptional potential for the sorption and immobilization of radioactive contaminants. This efficacy stems from a powerful synergy: the graphene scaffold provides a vast, functionalizable

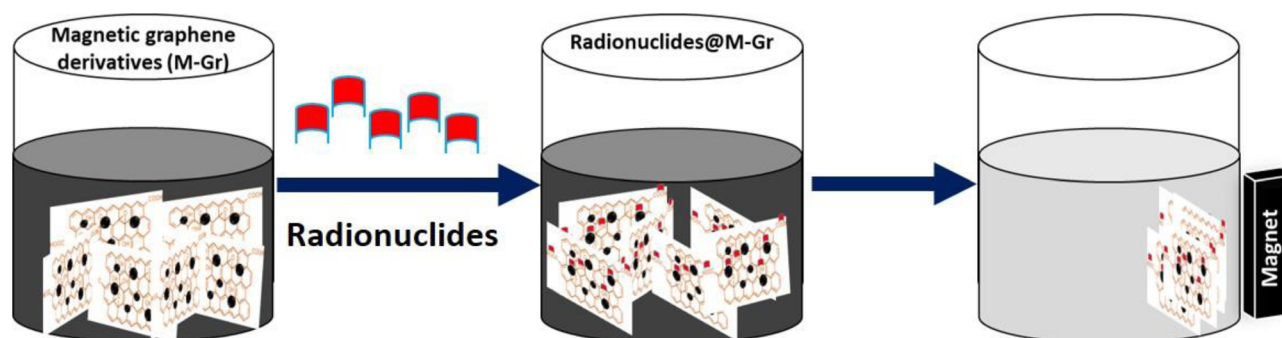


Fig. 1 Schematic diagram illustrating the process of radionuclide removal from contaminated water using magnetic graphene derivatives.



surface for radionuclide binding, while the embedded magnetic NPs enable rapid, low-energy recovery of the sorbent for reuse or safe disposal.^{4,17,20,21,25,26,29–31} The fundamental principle of this magnetic separation process is depicted in Fig. 1.

This review provides a comprehensive and critical overview of recent advancements in the synthesis, characterization, and application of Fe₃O₄@graphene-based NCs for radioactive waste management. We critically evaluate the key material properties, discuss the relative merits of various synthesis methods, and detail the suite of techniques used to characterize their structure and performance. Furthermore, this review examines the mechanisms governing radionuclide sorption, candidly addresses prevailing challenges, and outlines promising future research directions. By consolidating the current state of the field, this review aims to underscore the transformative potential of Fe₃O₄@graphene NCs to improve the safety, efficiency, and sustainability of radioactive waste management practices.

2.0 Key physicochemical properties of Fe₃O₄@graphene NCs

The exceptional performance of Fe₃O₄@graphene NCs in environmental remediation stems from a unique, synergistic combination of structural, magnetic, and surface properties. These characteristics function synergistically to facilitate both effective contaminant sorption and the subsequent recovery of the sorbent material.

2.1 Structural and magnetic properties

Fe₃O₄@graphene NCs are hybrid materials formed by the intimate integration of Fe₃O₄ NPs with a graphene-based matrix, typically graphene oxide (GO) or reduced graphene oxide (rGO). The synthesis method dictates the final architecture, with Fe₃O₄ NPs—typically ranging from a few to tens of nanometers in diameter—either decorating the surface of the graphene sheets or becoming intercalated between them.^{4,12,20,32–34} Within this composite structure, the Fe₃O₄ NPs critically retain their inverse spinel crystal structure, a crystallographic arrangement that is fundamental to the material's unique magnetic behavior.^{4,33,35}

A defining feature of these NCs is the superparamagnetism of the Fe₃O₄ component. This phenomenon arises in magnetic NPs below a critical size, where the thermal energy is sufficient to overcome the magnetic anisotropy, causing the particle's magnetic moment to fluctuate randomly. As a result, the material exhibits zero net magnetism in the absence of an external field but becomes strongly magnetized when a field is applied.^{19,29,32,33,35–39} This “on/off” magnetic behavior is essential for practical applications, as it allows for the rapid and complete recovery of the NC from aqueous solutions without the persistent particle–particle attraction (and subsequent irreversible aggregation) that would occur with permanently magnetic materials.^{12,21,35,40–42}

The magnetic saturation (Ms), a measure of the maximum induced magnetic moment, is a key performance metric. While the Ms value of the NCs is invariably lower than that of pure

Fe₃O₄ due to the mass contribution of the non-magnetic graphene matrix, it remains sufficiently high for efficient magnetic separation.^{13,19,32,33,35–38,42,43} This strong magnetic responsiveness is a significant advantage in radioactive waste management, where minimizing processing time and ensuring complete removal of the contaminant-laden sorbent are paramount.^{13,19,33,38,40} Furthermore, these composites generally exhibit high thermal and chemical stability, enabling them to function effectively across a wide range of environmental conditions, including highly acidic or alkaline environments often encountered in industrial and legacy radioactive waste streams.^{12,30,36,41}

2.2 Surface area and porosity

The efficacy of any sorbent is fundamentally linked to its surface area. Graphene and its derivatives are renowned for their exceptionally high theoretical specific surface area (up to 2630 m² g^{−1}), which provides an unparalleled platform for contaminant sorption.^{3,27,44} When integrated with Fe₃O₄ NPs, the resulting NCs retain a significant portion of this high surface area. Although the incorporation of the denser Fe₃O₄ phase may lead to a reduction in the gravimetric surface area, the composite's overall surface area remains substantially larger than that of conventional sorbent materials like clays or zeolites used in radioactive waste management.^{4,25} The NPs also act as spacers, preventing the graphene sheets from restacking and ensuring that the high surface area is accessible.

Experimentally, the specific surface area of Fe₃O₄@graphene NCs is commonly determined using the Brunauer–Emmett–Teller (BET) method. Measured values typically range from approximately 30 to over 400 m² g^{−1}. This wide variation is a direct consequence of the synthesis method, the Fe₃O₄-to-graphene mass ratio, and the degree of graphene exfoliation and aggregation achieved during processing.^{12,16,25,43,45,46} Beyond sheer area, the porous structure of these NCs, often featuring a hierarchical combination of micropores (<2 nm) and mesopores (2–50 nm), further enhances their sorption capacity. This hierarchical porosity is ideal for environmental applications: the larger mesopores serve as transport channels, enabling efficient diffusion of hydrated ions from the bulk solution into the material's interior, while the smaller micropores provide high-energy binding sites that contribute to strong sorption, creating a synergistic effect that improves overall performance.

2.3 Regeneration and reusability

For any sorption technology to be economically viable and environmentally sustainable, the sorbent material must be capable of regeneration and reuse. The robust covalent and non-covalent framework of Fe₃O₄@graphene NCs allows them to withstand multiple cycles of sorption and desorption without significant mechanical or chemical degradation. Adsorbed contaminants can typically be eluted by shifting the chemical equilibrium, for example, by washing with acidic solutions that protonate surface functional groups and displace bound metal cations. This process restores the active sites, enabling the material to be used in subsequent treatment cycles.^{4,12,17,19–21,29,34,38,40,42,47–49}



The literature provides compelling evidence of this reusability. For instance, Zhao *et al.*²⁰ demonstrated that their Fe₃O₄/GO NCs retained nearly 70% of their initial U(vi) removal efficiency after five complete cycles. In another study, a similar composite maintained over 87% efficiency after five cycles using a dilute 0.1 M HCl solution for regeneration.¹² Zong *et al.*⁴ reported an even more stable magnetic graphene/iron oxide composite that exhibited only a 4% decline in U(vi) sorption over seven cycles. Similarly, Yang *et al.*⁴⁹ found that their advanced GO/Fe₃O₄/GC composite retained over 85% removal efficacy for U(vi) after five cycles with regeneration using 3 M HNO₃. This high degree of reusability, combined with the low-energy, highly efficient magnetic separation process, firmly positions Fe₃O₄@graphene NCs as a promising, sustainable technological solution to the complex challenges of radioactive waste treatment.

3.0 Synthesis of Fe₃O₄@graphene NCs

The synthesis method employed to produce Fe₃O₄@graphene NCs is a critical determinant of their final physicochemical properties, including particle size distribution, morphology, surface chemistry, and magnetic responsiveness. The choice of a specific method is therefore a strategic decision based on the desired material characteristics and the intended application.

This section provides a more detailed review of the most common strategies for synthesizing these advanced materials.

3.1 Simple mixing

The most direct approach to preparing these composites is to mix pre-synthesized Fe₃O₄ NPs with graphene derivatives physically. In this method, the individually prepared components are dispersed in a suitable solvent and then combined, typically under high-energy conditions such as ultrasonication or vigorous mechanical stirring, to promote the physical sorption of Fe₃O₄ NPs onto the expansive surface of the graphene sheets.^{25,36,37,50,51} For example, Yang *et al.*²⁵ prepared their Fe₃O₄/GO NCs by simply sonicating aqueous dispersions of GO and magnetite. To overcome the limitations of weak physisorption, chemical cross-linking is often employed to create a more robust and stable composite. Fatemeh *et al.*⁵⁰ used this strategy by first functionalizing the Fe₃O₄ NPs with amine groups, which could then form strong covalent amide bonds with the carboxyl groups present on the GO surface, resulting in a highly stable final product (Fig. 2).

While this method is valued for its simplicity, inherent scalability, and versatility, it often suffers from weak van der Waals interactions between the components (in the absence of cross-linking), which can lead to NP detachment or leaching during application. Furthermore, achieving a truly homogeneous distribution of NPs on the graphene surface can be

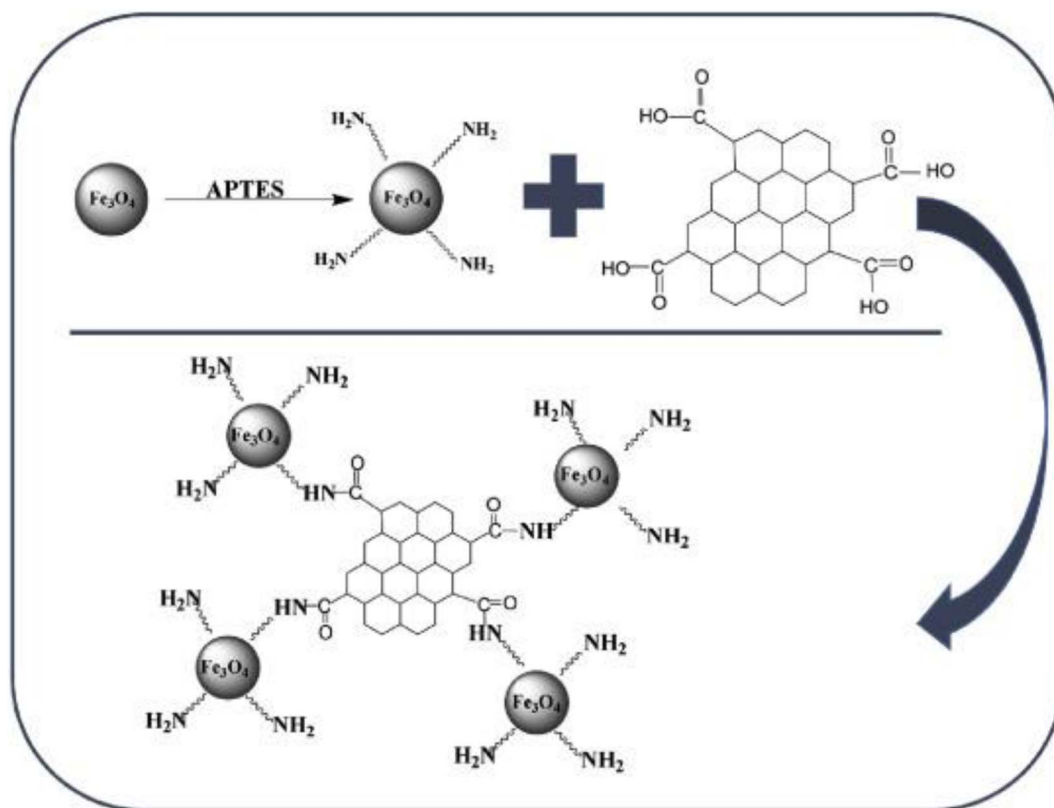


Fig. 2 Schematic representation of the synthesis of amine-functionalized Fe₃O₄/GO (Fe₃O₄-NH₂/GO) via a simple mixing and chemical cross-linking approach. Amine-functionalized Fe₃O₄ NPs form covalent amide bonds with the carboxylic acid groups on GO.⁵⁰



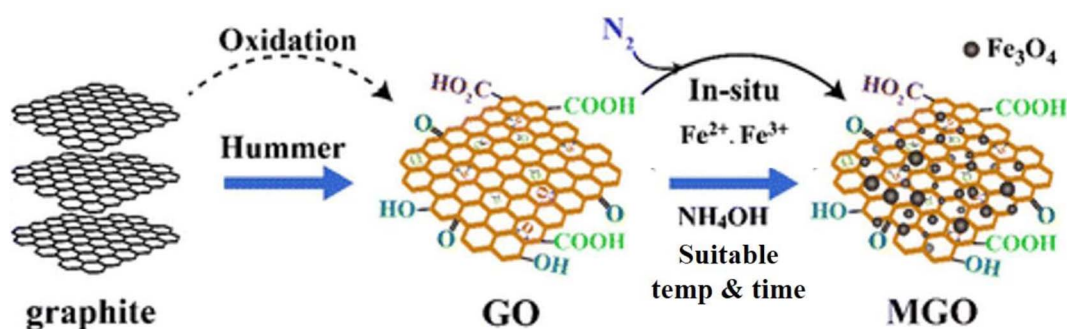


Fig. 3 Schematic illustration of the *in situ* co-precipitation method for preparing magnetic GO (MGO). Graphite is first oxidized to GO via a modified Hummers' method, followed by the precipitation of Fe_3O_4 NPs onto the GO surface from $\text{Fe}^{2+}/\text{Fe}^{3+}$ precursors.⁵⁷

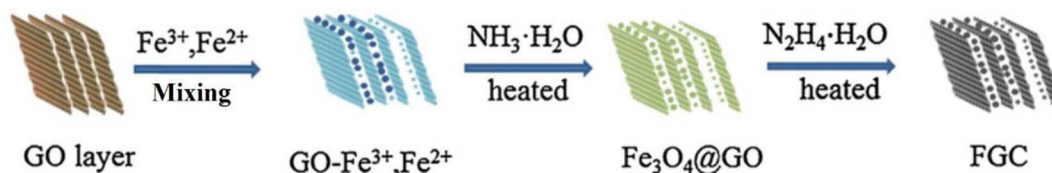


Fig. 4 Schematic diagram of an *in situ* co-precipitation process that includes a chemical reduction step. After the initial formation of $\text{Fe}_3\text{O}_4@\text{GO}$, a reducing agent ($\text{N}_2\text{H}_4\cdot\text{H}_2\text{O}$) is added to convert GO to rGO, yielding a final composite (FGC).³⁰

challenging, potentially leading to inconsistent material performance and underutilization of available surface area.

3.2 Co-precipitation

Co-precipitation is a widely used and highly effective *in situ* method where Fe_3O_4 NPs are nucleated and grown directly on the surface of dispersed graphene derivatives. In a typical procedure, GO is first exfoliated and dispersed in an aqueous solution containing a stoichiometric mixture of Fe^{2+} and Fe^{3+} salts (e.g., FeCl_2 and FeCl_3), usually in a 1 : 2 molar ratio to favor the formation of Fe_3O_4 . The subsequent addition of a base, such as NH_4OH or NaOH , rapidly increases the pH to 10–12. This change in pH induces the simultaneous hydrolysis and co-precipitation of iron ions as Fe_3O_4 NPs directly onto the functionalized GO sheets, which act as nucleation sites.^{4,29,33,34,38,40,43,49,52–56} The process is often conducted under an inert atmosphere (e.g., N_2) and at elevated temperatures (e.g., 80–90 °C) to promote the formation of highly crystalline

magnetite and prevent oxidation to less magnetic phases, such as maghemite ($\gamma\text{-Fe}_2\text{O}_3$) (Fig. 3).

In some variations, a chemical reducing agent, such as hydrazine hydrate, is introduced after the initial precipitation. This step facilitates the simultaneous reduction of GO to the more conductive rGO, yielding a final $\text{Fe}_3\text{O}_4@\text{rGO}$ composite with potentially different surface properties (Fig. 4).^{18,26,30,36} Co-precipitation generally results in a more uniform NP distribution and a stronger, more intimate interaction between the Fe_3O_4 and graphene components compared to simple mixing. However, the method's success hinges on precise control over reaction parameters, such as pH, temperature, stirring rate, and precursor concentration, to avoid uncontrolled NP agglomeration and ensure high batch-to-batch reproducibility.

3.3 Hydrothermal and solvothermal methods

Hydrothermal and solvothermal methods are powerful synthesis techniques that utilize high temperatures (typically

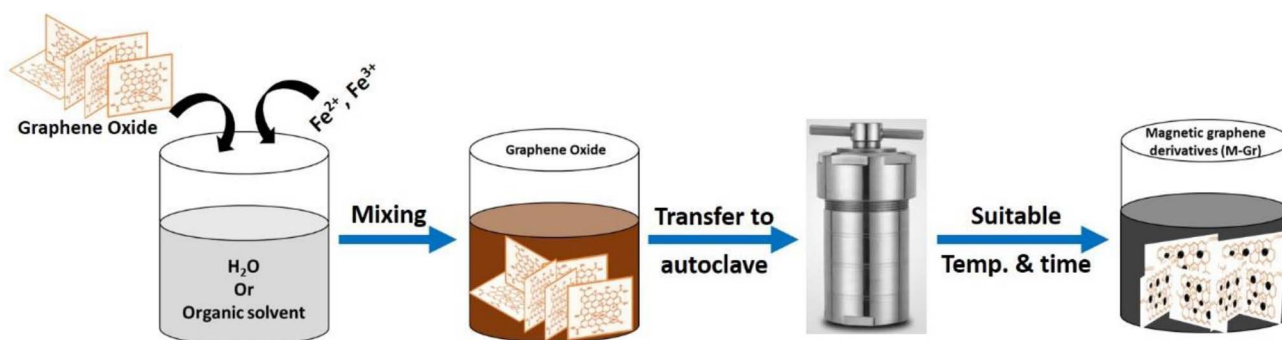


Fig. 5 General schematic of the hydrothermal or solvothermal synthesis process for the synthesis of magnetic graphene derivatives.



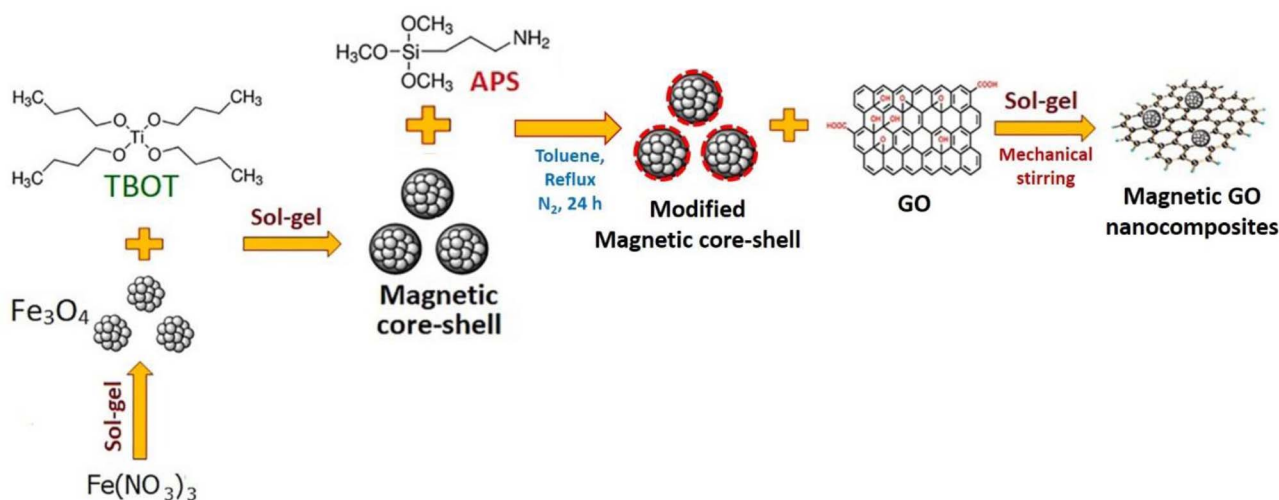


Fig. 6 Schematic of the sol-gel synthesis of a magnetic GO NC. Pre-formed Fe_3O_4 NPs are coated with a silica precursor (TBOT) and functionalized with an amine-containing silane (APS) before being integrated with GO through a sol-gel process.⁴⁶

120–200 °C) and the corresponding autogenous pressures within a sealed vessel (an autoclave) to produce highly crystalline and well-defined NCs (Fig. 5). In this process, GO is dispersed in a solution of iron salt precursors. The distinction lies in the solvent: the hydrothermal method uses water, while the solvothermal method employs an organic solvent with a high boiling point, such as ethylene glycol or diethylene

glycol. Under these solvothermal conditions, the iron precursors decompose, and the resulting ions undergo controlled hydrolysis and condensation to nucleate and grow Fe_3O_4 NPs directly on the graphene sheets.^{12,13,16,19–21,58}

These methods provide exceptional control over the size, shape (morphology), and crystallinity of the resulting NPs, which, in turn, lead to enhanced, more uniform magnetic

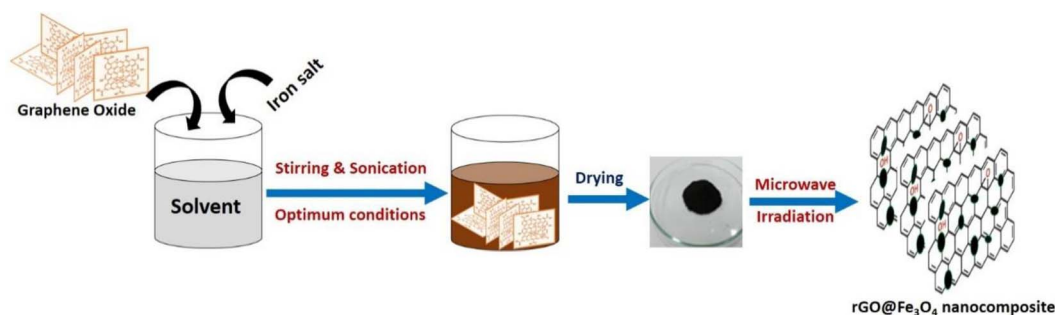


Fig. 7 General schematic of the microwave-assisted synthesis of $\text{rGO@Fe}_3\text{O}_4$ NCs. A mixture of GO and iron salt precursors is subjected to microwave irradiation, which simultaneously reduces the GO and forms the Fe_3O_4 NPs.

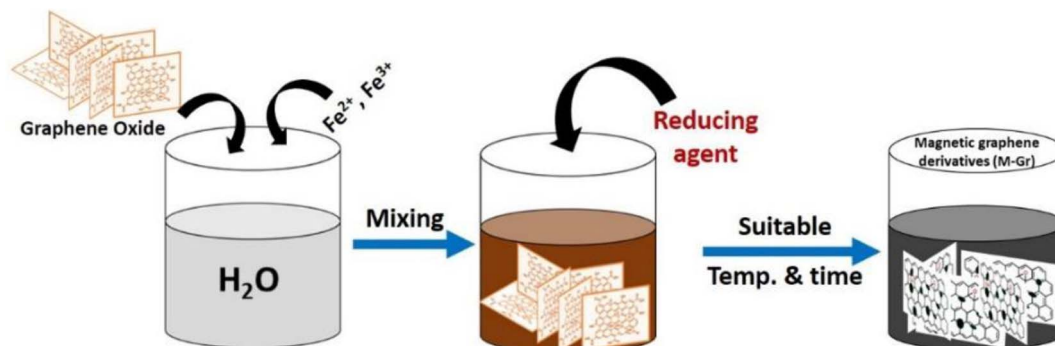


Fig. 8 Schematic diagram of the *in situ* chemical reduction method. A reducing agent is added to a mixture of GO and iron salt precursors, leading to the simultaneous formation of Fe_3O_4 NPs and reduction of GO to rGO.



Table 2 Summary of representative synthesis methods for Fe₃O₄@graphene-based NCs^a

| Composite name | Precursors | Conditions | Ref. |
|---|--|---|------|
| Synthesis method: simple mixing | | | |
| PB/Fe ₃ O ₄ /GO | GO, Fe ₃ O ₄ , FeCl ₃ , K ₄ [Fe(CN) ₆] | GO & Fe ₃ O ₄ ultrasonicated in 50 & 300 mL, mixed & stirred 10 min, FeCl ₃ added, K ₄ [Fe(CN) ₆] added dropwise and stirred 1 h | 25 |
| Fe ₃ O ₄ -NH ₂ /GO | FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ , APTES, GO, toluene | Fe ₃ O ₄ synthesized at 80 °C for 2 h, dried 60 °C 5 h, APTES functionalization in toluene under N ₂ at reflux 24 h, GO dispersed in water + Fe ₃ O ₄ -NH ₂ (ratios 1 : 1, 1 : 2, 1 : 3), stirred 30 min, NH ₄ OH added dropwise, stirred 2 h at 80 °C | 50 |
| Synthesis method: co-precipitation | | | |
| Fe ₃ O ₄ /GO | GO, ferric chloride, ferrous chloride | 50 °C for 30 min under N ₂ by constant mechanical stirring | 55 |
| (1) MGO | (1) GO, FeCl ₃ ·6H ₂ O and FeCl ₂ ·4H ₂ O | (1) 60 °C for 1 h, stirred under N ₂ atmosphere, pH 10 | 47 |
| (2) MGO <i>via</i> the cross-linking | (2) MGO, chitosan, NaTPP, glutaraldehyde | (2) RT for hydrogel formation, 40 °C drying, 6 h cross-linking at 150 rpm | |
| Fe ₃ O ₄ /GO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ | 90 °C for 8 h under N ₂ , pH adjusted to 10.5 | 4 |
| Magnetic GO | GO, FeCl ₃ ·6H ₂ O, FeSO ₄ ·7H ₂ O, NH ₄ OH | pH 10, RT, 4 h, N ₂ atmosphere | 54 |
| M-GO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ | Ultrasonicated at 85 °C, pH 10, 30 min sonication | 33 |
| M-GO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ | Alkaline pH | 29 |
| MGO | GO, (NH ₄) ₂ Fe(SO ₄) ₂ ·6H ₂ O, FeCl ₃ ·6H ₂ O, NH ₃ , HCl | 80 °C under N ₂ , stirred 1 h | 34 |
| (1) MGOs | (1) GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, H ₂ O ₂ | (1) 90 °C for 8.5 h under N ₂ , dried at 60 °C for 24 h | 1 |
| (2) CMC/MGOs <i>via</i> plasma-induced grafting | (2) MGOs, carboxymethyl cellulose (CMC), N ₂ gas | (2) Plasma activation: 45 min (10 Pa, 950 V, 120 W); grafting: 60 °C, 48 h | |
| SMGO | Disaccharide sugar, O ₃ , FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NaOH | 80 °C for 1 h, pH ≥ 10, under N ₂ atmosphere | 38 |
| MCGO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ , HCl, acetic acid | Sonication + 12 h stirring, pH adjusted to 8 under N ₂ | 40 |
| GO/Fe ₃ O ₄ /GC | GO, graphitic carbon (GC), FeSO ₄ ·7H ₂ O, NH ₃ ·H ₂ O | Ultrasonicated 3 h, pH 11, stirred 3 h | 49 |
| M/GO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ | 85 °C, pH 10, stirred 45 min | 52 |
| Fe ₃ O ₄ /GO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ | Room temp to 80 °C, pH 10; 45 min stirring | 56 |
| MGO | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₃ ·H ₂ O | pH pre-adjusted, stirred for 3 h at 80 °C | 57 |
| MGO | GO, FeCl ₃ , FeCl ₂ , NH ₄ OH, hydrazine hydrate | 80 °C, under N ₂ , stirred | 18 |
| (1) MGO | (1) GO, FeCl ₃ , FeCl ₂ , NH ₄ OH, hydrazine hydrate | (1) 80 °C under N ₂ , stirred | 26 |
| (2) PBMGO | (2) GO, Fe ₃ O ₄ , FeCl ₃ , K ₄ [Fe(CN) ₆] | (2) Room temp; stirred 1 h | |
| FGC | GO, FeCl ₃ ·6H ₂ O, FeCl ₂ ·4H ₂ O, NH ₄ OH, hydrazine hydrate | pH 10, 80 °C, stirred for 5 h | 30 |
| Fe ₃ O ₄ /rGO | GO, FeCl ₃ , FeCl ₂ , NH ₄ OH, hydrazine hydrate | 80 °C, stirred for 5 h | 35 |
| Synthesis method: hydrothermal | | | |
| Magnetic PANI/GO | GO, aniline, (NH ₄) ₂ S ₂ O ₈ , FeCl ₃ , FeCl ₂ , NH ₃ , hydrazine hydrate | 90 °C, stirred for 4 h under N ₂ , pH 10 | 17 |
| FGC | GO, FeCl ₃ , FeCl ₂ , 30% ammonia solution | 180 °C for 10 h | 12 |
| Fe ₃ O ₄ /rGO | GO, FeCl ₃ ·6H ₂ O, sodium acetate, aqueous ammonia, diethylene glycol | 180 °C for 12 h in autoclave | 16 |
| GN-Fe ₃ O ₄ | GO, FeCl ₃ ·6H ₂ O, ascorbic acid, hydrazine hydrate | 180 °C, 8 h (autoclave), then 500 °C calcination (4 h, N ₂) | 58 |
| Synthesis method: solvothermal | | | |
| MGONRs | GONRs, FeCl ₃ ·6H ₂ O, sodium acetate, ethylene glycol | 200 °C for 6 h, vacuum drying at 60 °C for 12 h | 13 |
| Fe ₃ O ₄ /GO | GO, ethylene glycol, FeCl ₃ ·6H ₂ O, NaAc | 190 °C for 6 h, vacuum oven at 60 °C for 24 h | 20 |
| EDTA-mGO | (1) GO, ethylene glycol, NaAc, FeCl ₃ ·6H ₂ O (2) mGO, HAC, ethylene glycol, EDTA | (1) 190 °C for 6 h (2) Stirred for 4 h at 60 °C, dried in a vacuum oven at 60 °C for 24 h | 21 |
| UiO-66/Fe ₃ O ₄ /GO | GO, Fe ₃ O ₄ , ZrCl ₄ , PTA, DMF | 120 °C for 24 h in autoclave; sonication + stirring pre-treatment | 19 |



Table 2 (Contd.)

| Composite name | Precursors | Conditions | Ref. |
|---|---|--|------|
| Synthesis method: sol-gel method | | | |
| TBOT-GO-Fe ₃ O ₄ | GO, Fe(NO ₃) ₃ ·9H ₂ O, ethylene glycol, TBOT, 3-aminopropyltrimethoxysilane, ammonia, nitric acid, ethanol, water, toluene | 100 °C, 6 h reflux, surface modification: reflux 24 h under N ₂ atmosphere, final drying at 80 °C | 46 |
| Fe ₃ O ₄ /graphene | Potassium humate, FeCl ₃ ·6H ₂ O, NaCl | Stir at 70 °C for 12 h, dry at 70 °C for 48 h, mix with NaCl, calcine at 500/600/700 °C for 4 h under Ar (10 °C min ⁻¹) | 59 |
| Synthesis method: microwave-assisted synthesis | | | |
| SPION-rGO | GO, Fe(acac) ₃ , benzyl alcohol | Disperse GO in benzyl alcohol, mix with Fe(acac) ₃ , microwave heating in 2-stage program | 39 |
| Fe ₃ O ₄ @SiO ₂ -GO | Fe ₃ O ₄ @SiO ₂ , 3-aminopropyltriethoxysilane, GO, NHS, EDC, isopropanol | Functionalize Fe ₃ O ₄ @SiO ₂ with silane, react with GO-EDC/NHS at 60 °C, 80 W for 4 h | 48 |
| rGO@Fe ₃ O ₄ | Graphite oxide, EtOH, FeCl ₃ ·6H ₂ O, diluted NH ₃ ·H ₂ O | Magnetic stirring, sonication, addition of Fe salt & NH ₃ ·H ₂ O, drying at 50 °C, microwave irradiation (900 W, 45 s) | 61 |
| Synthesis method: in situ chemical reduction | | | |
| Fe ₃ O ₄ /rGO | GO, FeCl ₃ , FeCl ₂ , NH ₃ ·H ₂ O, hydrazine hydrate | FeCl ₃ and FeCl ₂ added slowly to GO, NH ₃ added to precipitate Fe ₃ O ₄ , stirred 5 h at 80 °C | 35 |
| Fe ₃ O ₄ /RGO | GO, FeCl ₃ ·6H ₂ O, <i>Murraya koenigii</i> leaf extract | Stirred 1 h, refluxed 5 h at 80 °C | 42 |
| rGO/Fe ₃ O ₄ | GO, FeSO ₄ ·7H ₂ O, FeCl ₃ ·6H ₂ O, <i>Dolichos lablab</i> pod extract | Sonicated 1 h, extract added & stirred 1 h at 30 °C, NH ₃ adjusted pH10, heated 80 °C for 12 h | 62 |

^a PB, Prussian blue; PANI, polyaniline; NaTPP, sodium tripolyphosphate; APTES, 3-aminopropyltriethoxysilane; TBOT, tetrabutyl orthotitanate; DMF, dimethylformamide.

properties and a robust, stable interface between the two components. The high-temperature environment can also facilitate the partial reduction of GO to rGO. The primary drawbacks of these techniques are the need for specialized, costly high-pressure equipment, significantly longer reaction times (often several hours to a full day), and higher energy consumption, which can be limiting factors for large-scale industrial production.

3.4 Other synthesis methods

Several other advanced techniques have been successfully applied to the synthesis of Fe₃O₄@graphene NCs, each offering unique advantages and challenges.

The sol-gel method is a versatile bottom-up approach that involves transforming a liquid colloidal suspension ("sol") into a solid three-dimensional network ("gel"). In this context, molecular precursors (e.g., iron alkoxides) undergo hydrolysis and polycondensation in a sol containing a dispersed carbon source, such as GO. This process results in a gel where Fe₃O₄ NPs are homogeneously embedded within the interconnected graphene matrix. Subsequent aging, drying, and thermal treatment (calcination) yield the final porous composite (Fig. 6).^{46,59} This method offers exquisite control over the material's final composition and porosity, but is often a multi-step, complex process that can be prone to structural defects, such as cracking, during solvent removal.

Microwave-assisted synthesis represents a significant intensification strategy for process development. It uses microwave radiation to deliver energy directly and efficiently to the polar molecules in the reaction mixture, resulting in rapid, uniform

heating. This dramatically reduces reaction times (often from hours to minutes) and lowers overall energy consumption compared to conventional heating methods (Fig. 7). The technique promotes the rapid nucleation and controlled growth of Fe₃O₄ NPs on the graphene surface, offering excellent control over particle size and distribution.^{39,48,60,61} However, it requires specialized microwave reactor equipment, and the limited penetration depth of microwaves can pose a challenge for scaling up to large reaction volumes.

In situ chemical reduction is an elegant one-pot approach that simultaneously reduces GO to rGO and forms Fe₃O₄ NPs from their salt precursors. This is achieved by introducing a chemical reducing agent, which can range from conventional reagents, such as sodium borohydride or hydrazine hydrate, to more environmentally friendly "green" reagents like plant extracts, into the reaction mixture (Fig. 8).^{35,42,62}

This method is highly efficient and promotes the formation of strong chemical bonds between the components. The main challenge lies in carefully controlling the reduction kinetics to prevent the uncontrolled, rapid growth and subsequent agglomeration of the NPs.

A summary of various synthesis approaches and their corresponding conditions is provided in Table 2. A schematic overview of these methods is presented in Fig. 9.

4.0 Characterization of Fe₃O₄@graphene NCs

A comprehensive and multifaceted characterization of Fe₃O₄@graphene NCs is essential to fundamentally understand



their structure–property relationships, which, in turn, enables the optimization of their performance in targeted applications. A suite of complementary analytical techniques is typically employed to probe their crystallographic, morphological, chemical, and magnetic properties in detail.

4.1 Crystallographic analysis: X-ray diffraction (XRD)

XRD is a fundamental and indispensable technique for identifying crystalline phases in a material and determining key structural properties. For Fe_3O_4 @graphene NCs, XRD patterns consistently confirm the successful formation of the desired iron oxide phase. They typically exhibit a set of characteristic

diffraction peaks at 2θ values of approximately 30.1° , 35.4° , 43.1° , 57.0° , and 62.5° . These peaks can be indexed to the (220), (311), (400), (511), and (440) crystallographic planes, respectively, which is the definitive fingerprint of the inverse spinel crystal structure of magnetite (Fe_3O_4) (JCPDS file no. 19-0629).^{30,33,41} Interestingly, the characteristic broad diffraction peak of GO, which arises from the regular stacking of oxidized sheets and is typically observed around $2\theta = 10\text{--}11^\circ$, is often significantly diminished, broadened, or even completely absent in the final composite's XRD pattern (Fig. 10). The observation is highly informative, indicating that the integration of Fe_3O_4 NPs has effectively disrupted the regular layer-by-layer stacking of GO sheets, leading to a more exfoliated and disordered

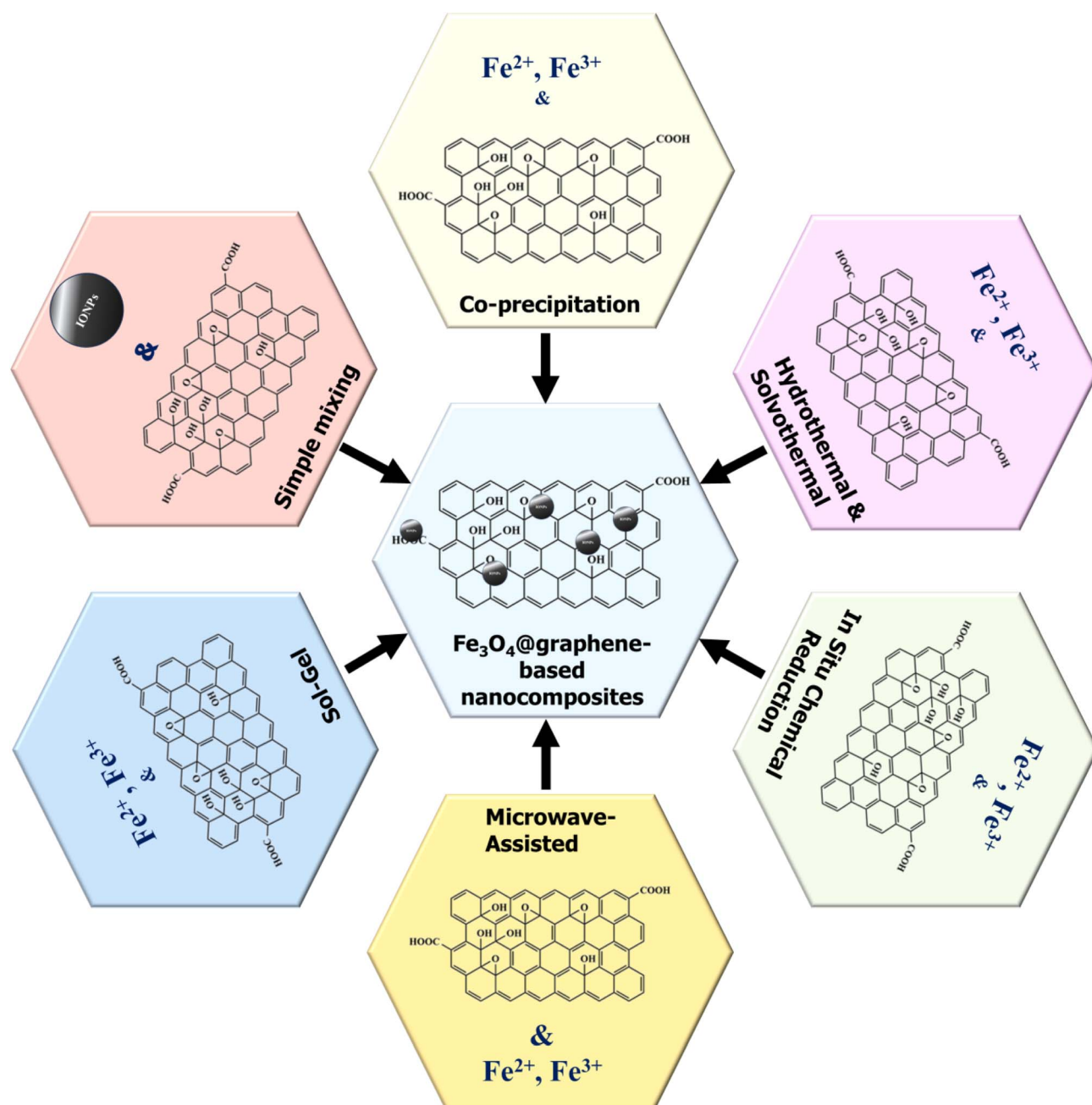


Fig. 9 Schematic overview of the primary synthesis methods for producing Fe_3O_4 @graphene-based NCs.



structure.^{12,18} In composites where GO is deliberately reduced to rGO, a new broad peak corresponding to the restacked rGO sheets may appear at a higher angle, around $2\theta = 24^\circ$.³⁵ Furthermore, the width of the Fe_3O_4 diffraction peaks can be used in conjunction with the Scherrer equation to provide a reliable estimate of the average crystallite size, which typically falls within the desired nanometer range.^{30,41} A summary of XRD findings from a range of studies is presented in Table 3.

4.2 Morphological and microstructural analysis: electron microscopy (SEM and TEM)

Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) are indispensable tools for visualizing the morphology, particle size, and spatial distribution of the components within the NC. SEM provides detailed information about the surface topography, revealing how the Fe_3O_4 NPs are distributed across the larger, often wrinkled, sheet-like structure of the graphene matrix (Fig. 11). An ideal morphology, typically sought after, is a uniform, dense NP decoration, as this configuration is most effective at preventing graphene sheet restacking, thereby maximizing the accessible surface area for sorption.^{12,20,61}

TEM offers significantly higher resolution, enabling the detailed analysis of individual NP size and shape, as well as a clear visualization of their interface with the graphene support. TEM images characteristically show the translucent, veil-like sheets of graphene decorated with dark, electron-dense Fe_3O_4 NPs (Fig. 12). At very high magnifications, High-Resolution TEM (HRTEM) can even resolve the lattice fringes of the individual crystalline Fe_3O_4 NPs, providing direct visual confirmation of their high crystallinity and structural integrity.⁴¹ Collectively, these powerful microscopy techniques confirm the synergistic architecture of the composite: the graphene sheets act as a high-surface-area support that prevents the magnetic NPs from agglomerating, while the NPs, in turn, act as spacers that prevent the graphene sheets from collapsing and restacking. This mutual stabilization is a key factor in the

material's high performance. Table 4 summarizes morphological observations from various representative studies.

4.3 Surface and textural properties: BET surface area analysis

The Brunauer–Emmett–Teller (BET) method, based on the physical sorption of nitrogen gas at cryogenic temperatures, is the standard technique for quantifying the specific surface area and pore characteristics of porous materials. These parameters are of paramount importance for sorption applications, as they directly relate to the number of available binding sites. As detailed in Table 5, the specific surface areas of these NCs can vary dramatically, from as low as $\sim 40 \text{ m}^2 \text{ g}^{-1}$ to well over $400 \text{ m}^2 \text{ g}^{-1}$, with this variation directly determined by the chosen synthesis conditions and the resulting material architecture.^{12,38} The analysis also yields critical information on the total pore volume and the average pore diameter. The data often reveal a predominantly mesoporous structure (pores between 2 and 50 nm), which is considered ideal for environmental remediation applications because it allows unhindered diffusion of hydrated ions from the bulk solution to the internal sorption sites.^{12,21,61} For example, Zhao *et al.*¹² reported a BET surface area of $42.53 \text{ m}^2 \text{ g}^{-1}$ and an average pore diameter of 15.71 nm for their magnetic composite, confirming its mesoporous nature. In stark contrast, a composite synthesized by Pu *et al.*⁵⁷ under different conditions exhibited a much higher surface area of $407 \text{ m}^2 \text{ g}^{-1}$, highlighting the tunability of these materials. These textural properties are crucial inputs for accurately modeling and predicting the material's ultimate sorption capacity.

4.4 Chemical and functional group analysis: FTIR and Raman spectroscopy

Fourier Transform Infrared (FTIR) spectroscopy is a powerful technique for identifying the chemical functional groups present on the surface of the NC and for confirming the successful integration of the constituent components. The FTIR

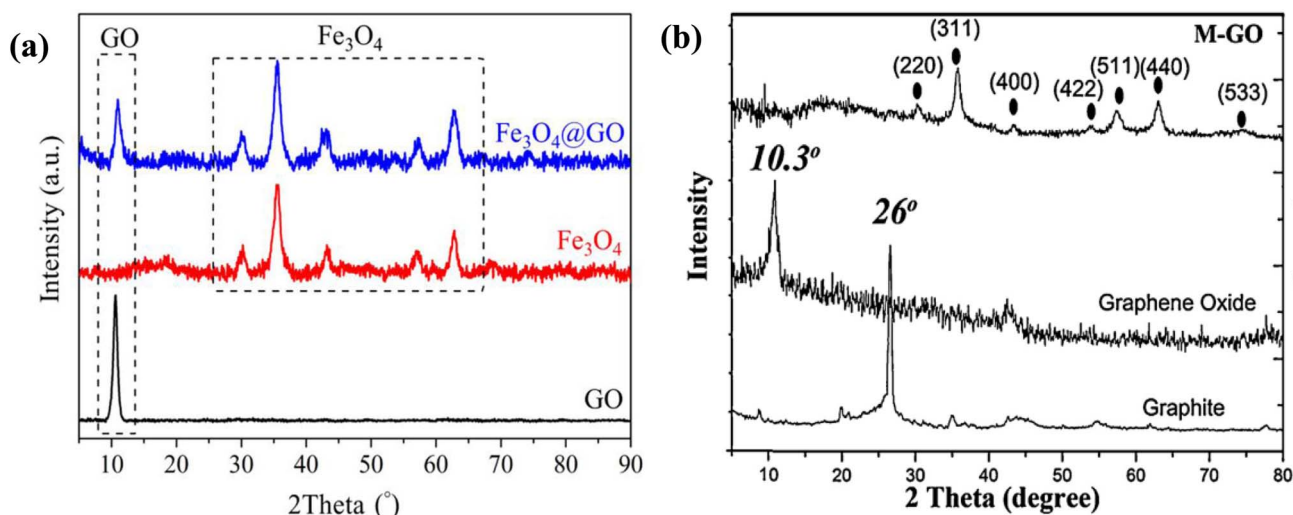


Fig. 10 Representative XRD patterns of (a) graphite, GO, and a magnetic GO (M-GO) composite, and (b) a different M-GO hybrid, demonstrating the characteristic peaks of the Fe_3O_4 phase and the change in the GO peak upon composite formation.^{33,41}



spectrum of a typical Fe₃O₄@graphene composite displays the characteristic absorption bands associated with GO, including a broad peak for hydroxyl (–OH) stretching vibrations around 3400 cm^{–1}, a sharp peak for carboxyl (C=O) stretching at ~1730 cm^{–1}, vibrations from aromatic (C=C) bonds at ~1620 cm^{–1}, and absorptions from epoxy (C–O) groups at ~1220 cm^{–1}. A definitive indicator of successful composite formation is the appearance of a new, strong absorption band in the low-wavenumber region, typically around 580 cm^{–1}, which is the characteristic vibrational mode of the Fe–O bond within the Fe₃O₄ crystal lattice (Fig. 13). Furthermore, a noticeable reduction in the intensity of the peaks corresponding to oxygen-containing functional groups in the composite, when compared to the spectrum of pure GO, can provide evidence of both chemical interaction with the Fe₃O₄ NPs and the partial

reduction of GO during the synthesis process.^{12,13} A summary of key FTIR peaks reported in the literature is provided in Table 6.

Raman spectroscopy is an exceptionally sensitive tool for probing the structural integrity, electronic properties, and degree of disorder within the carbon lattice of the graphene component. The Raman spectrum of graphene-based materials is characteristically dominated by two prominent peaks: the D band (at ~1350 cm^{–1}), which is activated by structural defects, edges, and sp³-hybridized carbon atoms, and the G band (at ~1580 cm^{–1}), which arises from the in-plane vibrational mode of the sp²-hybridized carbon atoms that form the graphitic lattice. The intensity ratio of these two bands (*I*_D/*I*_G) is a widely accepted metric for quantifying the level of defects and disorder in the graphene structure. An increase in the *I*_D/*I*_G ratio in the composite relative to the starting GO is often observed, which can be interpreted as the formation of new, smaller graphitic

Table 3 Summary of XRD data for various Fe₃O₄@graphene-based NCs

| No. | Composite name | 2θ (°) values | Planes (<i>hkl</i>) | Crystal nature | Crystallite size (nm) | Particle size (nm) | Ref. |
|-----|---|--|--|--------------------|-----------------------|--------------------|------|
| 1 | Fe ₃ O ₄ @graphene | 10.3 (GO), 18.27, 30.1, 35.4, 43.05, 56.94, 62.51, and 73.95 | (001) GO, (111), (220), (311), (400), (422), (511), (553) | Inverse spinel | | ~32 | 30 |
| 2 | Fe ₃ O ₄ @GO | 11.0 (GO), 30.1, 35.5, 43.1, 57.0, and 62.7 | (002) GO, (220), (311), (400), (511), and (440) | | 8–9 | | 41 |
| 3 | Fe ₃ O ₄ /RGO (9 : 1) | 23.9 (RGO), 18.7, 30.1, 35.5, 43.2, 54.1, 57.3, and 62.9 | (002) RGO, (111), (220), (311), (400), (422), (511), and (440) | Cubic | | ~30 | 35 |
| 4 | M-GO | 30.1, 35.4, 37.1, 43.1, 53.4, 56.9, and 62.5 | (220), (311), (222), (400), (422), (511), and (440) | | | | 33 |
| 5 | Magnetic Fe ₃ O ₄ /graphene | 29.8, 35.2, 43.1, 53.9, 57.2, and 62.9 | (220), (311), (400), (422), (511), (440) | | | | 12 |
| 6 | MGO | 30.2, 35.6, 43.3, 53.7, 57.1, and 62.8 | | Cubic spinel | 11.2 ± 8 | | 18 |
| 7 | EDTA-mGO | 34.3, 43.2, 53.3, 57.5, and 63.2 | (220), (311), (400), (422), (551), and (440) | | | | 21 |
| 8 | RGO/Fe ₃ O ₄ | 30.2, 35.5, 43.2, 53.5, and 62.6 | (220), (311), (400), (422) and (440) | Face-centred cubic | | | 42 |
| 9 | MGO/chitosan | 20.8 (chitosan), 30.1, 35.4, 43.2, 53.6, 57.1, and 62.6 | (220), (311), (400), (422), (511), and (440) | Face-centred cubic | | | 47 |
| 10 | SMGO | 30.0, 35.4, 43.1, 53.6, 57.2, and 62.7 | (220), (311), (400), (422), (511), and (440) | Cubic | 12.1 | | 38 |
| 11 | Fe ₃ O ₄ /GO | 30.23, 35.21, 43.15, 53.43, 57.22, and 62.52 | (220), (311), (400), (422), (511), and (440) | Face-centred cubic | | | 20 |
| 12 | PB/Fe ₃ O ₄ /GO | 30.2, 35.6, 43.3, 53.7, 57.3, 62.8, 74.9, 17.4 (PB), 24.8 (PB), 35.3 (PB), and 39.5 (PB) | (220), (311), (400), (422), (511), (440), and (533) | Cubic spinel | | | 31 |
| 13 | Fe ₃ O ₄ -NH ₂ /GO (3 : 1) | 18.1, 30.0, 35.4, 43.3, 53.4, 57.1, and 62.7 | (111), (220), (311), (400), (422), (511), and (440) | Inverse-spinel | 12.13 | | 50 |
| 14 | (mGOi) | 18.27°, 30.1°, 35.4°, 43.05°, 56.94°, 62.51°, and 73.95° | (111), (220), (311), (400), (422), (511), and (553) | Spinel | 18.4 | | 56 |
| 15 | Fe ₃ O ₄ -GO | 26.5, 30.42, 35.06, 43.48, 53.22, 57.78, and 63.06 | | Cubic spinel | | | 43 |
| 16 | FeNPs/rGO | 26 (rGO), 44 (Fe ₃ O ₄) | | | | 80–85 | 63 |
| 17 | Fe ₃ O ₄ @SiO ₂ @GO | 18.31, 30.12, 35.39, 43.06, 54.93, 57.53, 63.45, and 73.92 | (111), (220), (311), (400), (422), (511), (440), and (553) | | | | 48 |
| 18 | rGO/Fe ₃ O ₄ | 21 (GO), 33.09, 35.64, 43.44, 53.91, 57.41, and 62.9 | (002) rGO, (220), (311), (400), (422), (511), and (440) | Face-centred cubic | 38 | | 64 |



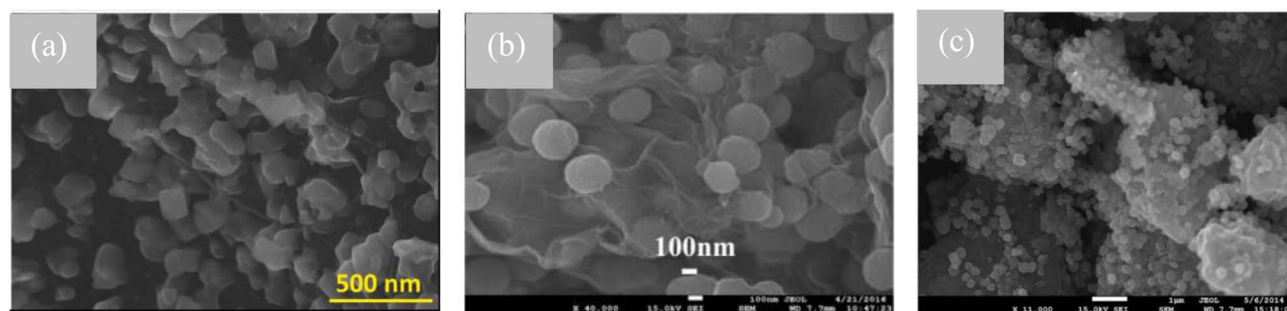


Fig. 11 Representative SEM images of various magnetic graphene composites (a) rGO@Fe₃O₄, (b) Fe₃O₄/graphene, and (c) Fe₃O₄/GO, showing Fe₃O₄ NPs distributed on the surface of graphene sheets.^{12,20,61}

domains or an increase in defect sites during chemical processing and NP integration.¹² Conversely, a decrease in this ratio may indicate a specific chemical interaction between the NPs and the existing defect sites on the GO surface, which can restrict the vibrational freedom of those atoms.³³ The Raman spectrum of the composite may also exhibit characteristic vibrational modes of Fe₃O₄ (e.g., a peak around 670 cm⁻¹), providing further confirmation of its presence (Fig. 14). Table 7 summarizes Raman spectral data from various studies.

4.5 Compositional and stability analysis: XPS and TGA

X-ray photoelectron spectroscopy (XPS) is a surface-sensitive analytical technique that provides quantitative information about the elemental composition and, crucially, the chemical (oxidation) states of the atoms on the material's surface. A survey scan of the composite readily confirms the presence of the expected elements: C, O, and Fe. High-resolution scans of specific elemental regions provide deeper insight. The C 1s region can be deconvoluted into multiple peaks corresponding to different carbon–oxygen functional groups (e.g., C–O, C=O, O–C=O), enabling a quantitative assessment of surface chemistry. The Fe 2p spectrum, with its characteristic Fe 2p_{3/2} and Fe 2p_{1/2} spin–orbit split peaks (located at binding energies of ~711 eV and ~724 eV, respectively), confirms the presence of iron in a mixed +2 and +3 oxidation state, which is consistent with the Fe₃O₄ stoichiometry.^{25,31,33,52,58} Most importantly, a careful analysis of the O 1s spectrum can often reveal

a distinct chemical environment associated with the formation of Fe–O–C bonds. This provides direct spectroscopic evidence of a covalent linkage between the iron oxide NPs and the graphene support, which is essential for the long-term stability and integrity of the composite (Fig. 15).³³ Table 8 summarizes key XPS findings from the literature.

Thermogravimetric Analysis (TGA) is used to assess the thermal stability of the NCs and to determine their approximate composition. The TGA curve plots the percentage weight loss of a sample as it is heated at a controlled rate. A typical TGA curve for a Fe₃O₄@-graphene composite shows several distinct weight-loss stages. An initial weight loss below ~150 °C is attributed to the desorption of physically adsorbed water. This is followed by a more significant weight loss, typically occurring between ~200 °C and 500 °C, which corresponds to the thermal decomposition of the labile oxygen-containing functional groups and, at higher temperatures, the combustion of the carbon skeleton of the graphene component. The mass that remains at the end of the analysis at high temperatures corresponds to the thermally stable inorganic component, Fe₃O₄ (which may be oxidized to Fe₂O₃ if the study is conducted in an air atmosphere). By comparing the total weight loss to that of pure GO under the same conditions, the relative mass percentage of Fe₃O₄ in the composite can be reliably estimated. These analyses generally show that the composites exhibit significantly enhanced thermal stability compared to pure GO, a result of the protective and stabilizing effects of the integrated iron oxide NPs.^{1,12,36,49} Table 9 summarizes TGA results from various studies.

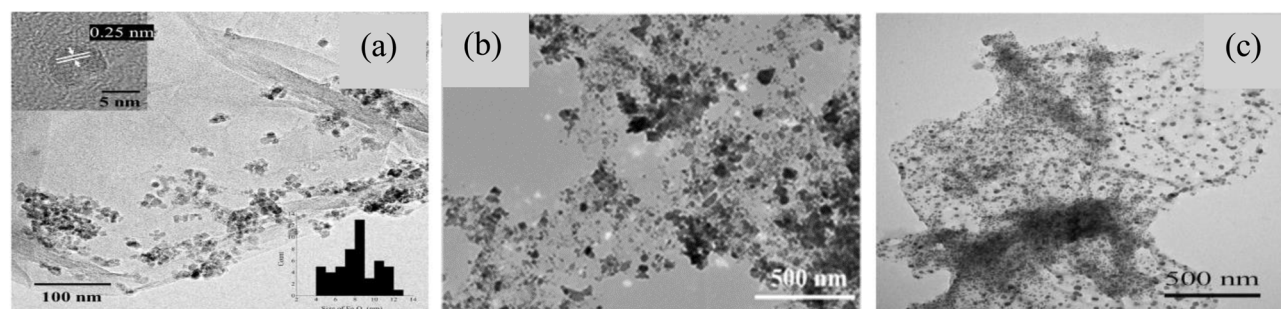


Fig. 12 Representative TEM images of magnetic graphene composites (a) Fe₃O₄@GO, (b) Fe₃O₄/GO, and (c) Fe₃O₄/rGO, providing a higher-resolution view of the uniform dispersion of Fe₃O₄ NPs on the translucent graphene sheets. The inset in (a) is an HRTEM image showing the crystalline lattice of an Fe₃O₄ NP.^{41,55,65}



Table 4 Summary of morphological characteristics of Fe₃O₄@graphene-based NCs observed via SEM and TEM^a

| No. | Composite name | Technique | Observed morphology | Particle size/layer thickness | Ref. |
|-----|---|-----------|---|-------------------------------|------|
| 1 | Fe ₃ O ₄ /GO | TEM | Fe ₃ O ₄ NPs are uniformly and homogeneously decorated on GO flakes | | 55 |
| 2 | MGO/chitosan | SEM | Fe ₃ O ₄ particles are anchored on a much rougher surface than the GO surface | | 47 |
| 3 | SMGO | TEM | Small Fe ₃ O ₄ particles are dispersed on a transparent and thin carbon sheet | 16 nm | 38 |
| 4 | GO/Fe ₃ O ₄ | SEM | Fe ₃ O ₄ particles are embedded on GO sheets | | 20 |
| 5 | Fe ₃ O ₄ /graphene | SEM & TEM | Fe ₃ O ₄ MNPs are dispersed on the surface of graphene homogeneously | 340 nm | 12 |
| 6 | PB/Fe ₃ O ₄ /GO | TEM | PB coating Fe ₃ O ₄ NPs inhomogeneously anchored on the surface of the GO sheets | 15 nm | 31 |
| 7 | UiO-66/Fe ₃ O ₄ /GO | SEM & TEM | UiO-66/Fe ₃ O ₄ NPs are disorderly dispersed in the wrinkles of the GO sheet | | 19 |
| 8 | PMMA/GO-Fe ₃ O ₄ | SEM | Fe ₃ O ₄ particles are embedded in PMMA/GO | | 53 |
| 9 | Fe ₃ O ₄ -NH ₂ /GO (2 : 1) | SEM | Fe ₃ O ₄ NPs loaded on GO sheets | 25 nm | 50 |
| 10 | Fe ₃ O ₄ @GO | TEM | Fe ₃ O ₄ NPs uniformly dispersed in GO sheets | 8.9 ± 0.9 nm | 41 |
| 11 | Fe ₃ O ₄ @SiO ₂ @ GO | TEM | GO forms the outermost shell layer on the SiO ₂ -coated | 500 nm | 48 |
| 12 | (GO)/Fe ₃ O ₄ | TEM | Elliptical and circular Fe ₃ O ₄ NPs were uniformly dispersed on the translucent GO | 9.8 nm | 57 |
| 13 | rGO@Fe ₃ O ₄ | SEM | rGO nanosheets fully loaded with faceted-like Fe ₃ O ₄ NPs | 100–150 nm | 61 |
| 14 | rGO/Fe ₃ O ₄ | SEM & TEM | The Fe ₃ O ₄ NPs were homogeneously assembled on the surface of RGO sheets | 12 nm | 42 |
| 15 | Graphene-Fe ₃ O ₄ | TEM | Two-dimensional graphene sheets densely decorated with spherical Fe ₃ O ₄ NPs | 7 nm | 58 |
| 16 | rGO/Fe ₃ O ₄ | FESEM | Spherical Fe ₃ O ₄ NPs are uniformly anchored on randomly dispersed rGO. | 9.75 to 14.85 nm | 62 |
| 17 | rGO/Fe ₃ O ₄ | FESEM | Crumpled rGO sheets with closely packed quasi-spherical Fe ₃ O ₄ NPs covering the surface | <10 nm | 16 |
| 18 | PB/Fe ₃ O ₄ /GO | SEM & TEM | PB/Fe ₃ O ₄ NPs inhomogeneously anchored on GO sheets | 17 nm | 25 |

^a PB, Prussian blue; PMMA, polymethyl methacrylate.

4.6 Magnetic properties: vibrating sample magnetometry (VSM)

VSM is the standard and most direct technique for characterizing the bulk magnetic properties of the NCs. The instrument measures the sample's magnetic moment as a function of an applied external magnetic field, generating a magnetic hysteresis loop. For well-synthesized Fe₃O₄@graphene NCs, these plots typically exhibit a characteristic "S" shape, with negligible coercivity (the field required to return the magnetization to zero) and remanence (the residual magnetization at zero field). It is the definitive signature of super-paramagnetic behavior. This result is critical, as it confirms that the material is strongly magnetic only when a field is applied and does not retain any permanent magnetism when the field is removed. This property is crucial for preventing magnetic aggregation and facilitating the easy redispersion of the sorbent in solution for reuse.

The saturation magnetization (M_s), which represents the maximum achievable magnetization and is measured from the

hysteresis loop plateau, is a key performance parameter. As shown in Table 10, the M_s values for these composites typically range from approximately 10 to over 60 emu g⁻¹.^{4,28,43,50,52,58} As expected, this value is lower than that of bulk Fe₃O₄ due to the presence of the non-magnetic graphene component. However, it is more than sufficient for rapid, effective magnetic separation from an aqueous solution using a conventional permanent magnet.

5.0 Application in radionuclide sorption

The unique constellation of properties inherent to Fe₃O₄@graphene NCs—namely, high surface area, abundant and tunable surface functional groups, and rapid magnetic separability—makes them exceptionally effective sorbents for the challenging task of removing radioactive ions from contaminated water. These materials offer significant performance advantages over many traditional remediation technologies.



Table 5 Summary of textural properties of various Fe₃O₄@graphene-based NCs determined by BET analysis

| No. | Composite name | BET surface area (m ² g ⁻¹) | Pore volume (cm ³ g ⁻¹) | Average pore diameter (nm) | Pore structure | Ref. |
|-----|---|--|--|----------------------------|----------------|------|
| 1 | MGO/chitosan | 169.7 | | | | 47 |
| 2 | SMGO | 428 | 0.146 | | | 38 |
| 3 | Magnetic-PANI/GO | 86.53 | | | | 17 |
| 4 | EDTA-mGO | 54.32 | | 16.5 | Mesoporous | 21 |
| 5 | Fe ₃ O ₄ /graphene | 42.53 | 0.26 | 15.71 | Mesoporous | 12 |
| 6 | UiO-66/Fe ₃ O ₄ /GO | 171.79 | 0.11162 | 8.9332 | | 19 |
| 7 | GO-Fe ₃ O ₄ | 124.37 | | 0.386 | | 43 |
| 8 | rGO@Fe ₃ O ₄ | 241 | 0.39 | 6.4 | Mesoporous | 66 |
| 9 | GO/Fe ₃ O ₄ | 407.1267 | | | | 57 |
| 10 | rGO@Fe ₃ O ₄ | 113 | 0.0068 | 4–20 | Mesoporous | 61 |
| 11 | M/GO | 126 | | | | 60 |
| 12 | RGO/Fe ₃ O ₄ | 58 | | | | 42 |
| 13 | GN-Fe ₃ O ₄ | 221 | 0.299 | | Mesoporous | 58 |
| 14 | M2-Fe ₃ O ₄ /RGO | 326 | 0.303 | | | 65 |
| 15 | 3D MPBRGO | 402.62 | | 4.34 | Mesoporous | 45 |
| 16 | PB/Fe ₃ O ₄ /GO | 152.91 | | | | 25 |
| 17 | MNGO | 124.37 | | 0.386 | | 67 |

5.1 Sorption performance for key radionuclides

A substantial body of research has demonstrated the high sorption capacity of these NCs for a range of environmentally and radiologically significant radionuclides, including uranium (as the uranyl cation, U(vi)), cesium (Cs(i)), strontium (Sr(ii)), and thorium (Th(iv)). As summarized in the comprehensive Table 11, the maximum sorption capacities (q_{\max}) reported in the literature are often substantial and highly competitive. For example, a functionalized FeWO₄/GO composite has shown an exceptional capacity for U(vi) removal, reaching an impressive 625 mg g⁻¹.²⁸ Similarly, a novel magnetic Prussian blue/GO aerogel, which combines the ion-sieving properties of Prussian blue with the magnetic graphene scaffold, exhibited a remarkable capacity of 484.12 mg g⁻¹ for the selective capture of Cs(i).⁴⁵

Kinetic studies consistently show that the sorption process is well-described by the pseudo-second-order kinetic model. It

indicates that the rate-limiting step is chemisorption, which involves valence forces through the sharing or exchange of electrons between the sorbent and the radionuclide. Furthermore, the equilibrium data often exhibit an excellent fit to the Langmuir isotherm model, which assumes monolayer sorption onto a finite number of identical, homogeneous active sites. Collectively, these findings underscore the immense potential of these materials for the efficient and high-capacity sequestration of hazardous radioactive contaminants from aqueous solutions.

5.2 Underlying sorption mechanisms

The efficient removal of radionuclides by Fe₃O₄@graphene NCs is not the result of a single process but rather a complex interplay of multiple physical and chemical interaction mechanisms, as illustrated schematically in Fig. 16. A thorough and nuanced understanding of these mechanisms is crucial for the

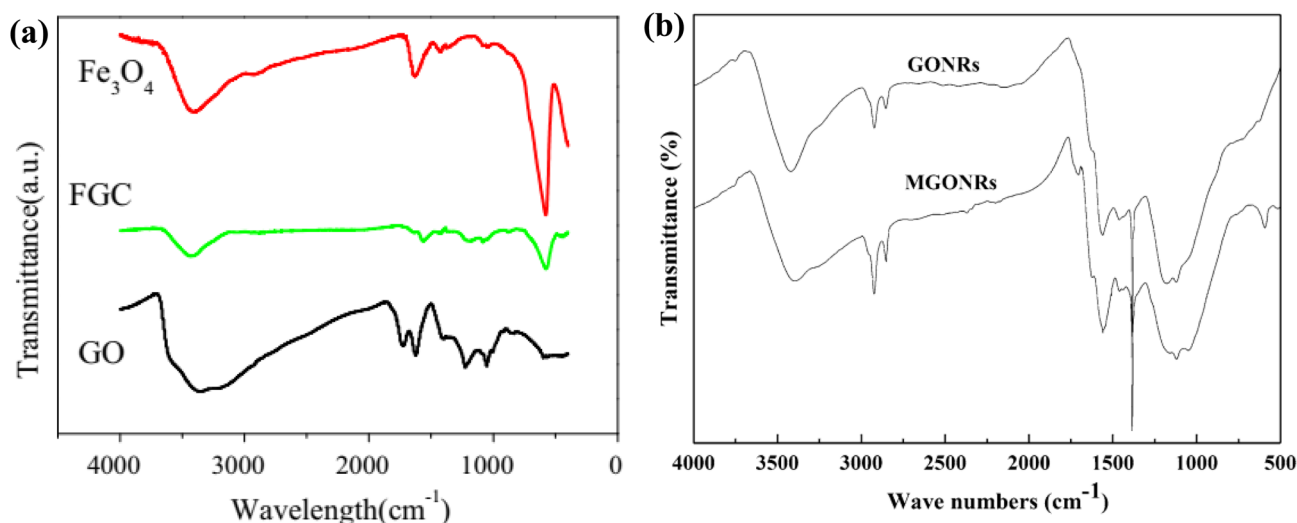


Fig. 13 Representative FTIR spectra of (a) GO and a magnetic composite (FGC), and (b) GO nanoribbons (GONRs) and their magnetic composite (MGONRs). The appearance of a peak around 580 cm⁻¹ in the composites confirms the presence of Fe–O bonds from Fe₃O₄.^{12,13}



Table 6 Summary of characteristic functional groups in Fe₃O₄@graphene-based NCs identified by FTIR spectroscopy

| No. | Composite name | Prominent FTIR peaks (cm ⁻¹) | Designated functional groups | Observed shifts | Ref. |
|-----|---|--|--|--|------|
| 1 | RGO/Fe ₃ O ₄ | 3328, 2952, 1658, 1449, 1122, and 585 | –OH, C–H (methyl), C=O, C=C, C–OH, and Fe–O | O–H band shifted, weakened, new Fe–O peak at 585 | 42 |
| 2 | MGO/chitosan | 3400, 2750–3600, 1724, 1660, 1620, 1402, 1377, 1068, 1053, and 580 | O–H, –NH/–OH (broad), C=O (COOH), amide (–NHCO), C=C/C–OH/O–H bending, C–H (–CH ₃), C–O–C, and Fe–O (Fe ₃ O ₄) | Slight intensity changes in 1660, 1377, and 1068 peaks; Fe–O peak at 580 retained | 47 |
| 3 | MGONRs | 3440, 2920, 2850, 1731, 1558, 1397, 1095, and 586 | –OH, –CH (symmetric & asymmetric), C=O, C–C (graphite), C–O, and Fe–O | Fe–O peak at 586 cm ⁻¹ appears in MGONRs | 13 |
| 4 | mGO–PP | 592, 2931, 2849, 2245, 1656, and 941 | Fe–O, CH ₂ , CH (PEI), C≡N (PAN), C=N, and N–O | Disappearance of C≡N (2245 cm ⁻¹); new peaks C=N and N–O after hydroxylamine treatment | 68 |
| 5 | Fe ₃ O ₄ /GO | 3420, 1578, 1225, and 580 | O–H (H ₂ O), C=C (aromatic), C–O (epoxy), and Fe–O | Fe–O peak at 580 cm ⁻¹ appears in Fe ₃ O ₄ /GO but not in GO | 20 |
| 6 | Magnetic GOs | 3450, 1730, 1620, 1220, and 1100 | O–H (adsorbed water), C=O, C=C (skeletal), O=C–O (carboxyl), and C–O (alkoxy) | Broad –OH stretching; oxygen-containing groups confirmed | 54 |
| 7 | Magnetic PANI/GO | 3400, 1750, 1630, 1420, 1180, 790, and 585 | –OH, C=O, C=C, C–C, C–O/N, and Fe–O | Fe–O bands at 790 & 585 cm ⁻¹ confirm magnetite | 17 |
| 8 | EDTA–mGO | 3435, 3260, 1733, 1401, 1384, 1122, and 581 | –OH, –NH, C=O, C–N, C–OH, C–NH ₂ , and Fe–O | GO C=O (1726) shifts to 1564 in mGO (COO ⁻) | 21 |
| 9 | Magnetic Fe ₃ O ₄ /graphene | 3450, 1625, 1251, and 580 | –OH, C–O, CO–H, and Fe–O | Intensity of –OH and Fe–O decreased in the composite | 12 |
| 10 | UiO-66/Fe ₃ O ₄ /GO | 3423, 1577–1654, 1095, and 571 | –OH, C=C, C–O, and Fe–O | Intensity of –OH, Fe–O increased | 19 |
| 11 | PMMA/GO–Fe ₃ O ₄ | 3430, 1730, 1444, 1244, and 584 | –CH ₂ stretching vibrations, C=O stretching, bending vibration of the C–H bonds of the –CH ₃ group, and C–O–C stretching vibration | Fe–O peak at 584 cm ⁻¹ appears in PMMA/GO–Fe ₃ O ₄ | 53 |
| 12 | Fe ₃ O ₄ /RGO | 1642, 675 | –COOH, Fe–O | Stretching vibration of Fe–O bond is also shifted to 675 cm ⁻¹ compared to that of 570 cm ⁻¹ reported for the stretching mode of Fe–O in bulk Fe ₃ O ₄ | 35 |

rational design of next-generation sorbents with enhanced selectivity and capacity. The primary mechanisms at play are discussed in this section.

5.2.1 Electrostatic attraction. The surface of GO and rGO is densely populated with oxygen-containing functional groups

(e.g., carboxyl, hydroxyl, epoxy). Under typical environmental pH conditions (neutral or slightly acidic), the carboxyl and hydroxyl groups are deprotonated, resulting in a significant negative charge on the graphene surface. This negative surface charge facilitates a strong, long-range electrostatic attraction of

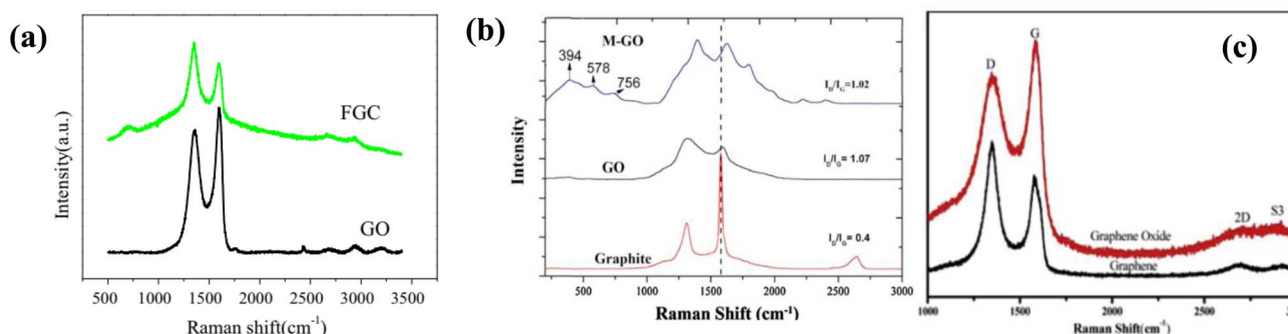


Fig. 14 Representative Raman spectra of magnetic graphene derivatives (a) GO and Fe₃O₄/graphene, (b) graphite, GO, and Fe₃O₄/GO, and (c) graphene and GO.^{12,33,54}



Table 7 Summary of Raman spectral characteristics of various Fe₃O₄@graphene-based NCs

| No. | Material | D-Band (cm ⁻¹) | G-Band (cm ⁻¹) | I _D /I _G | Remarks for I _D /I _G | Ref. |
|-----|--|----------------------------|----------------------------|--------------------------------|--|------|
| 01 | SMGO | ~1356 | ~1603 | 1.28 | Presence of localized sp ³ defects | 38 |
| 02 | CMC/MGOs | 1332 | 1580–1602 | 1.06 | Notably increased after using the plasma-grafted technique | 1 |
| 03 | Magnetic GOs | 1350 | 1580 | 0.83 | Indicate the defects of magnetic GOs | 54 |
| 04 | Fe ₃ O ₄ /graphene | ~1355 | ~1597 | 1.11 | New graphitic domains created | 12 |
| 05 | Fe ₃ O ₄ /graphene | 1344 | 1582 | 0.92 | Indicating the existence of graphitic carbon | 59 |
| 06 | M-GO hybrid | 1350 | 1580 | 1.02 | Anchoring of the magnetite NPs to the surface of graphene | 33 |
| 07 | Fe ₃ O ₄ @GO | 1352 | 1582 | 0.89 ± 0.01 | Increase in defects on the GO sheet due to the attachment of Fe ₃ O ₄ | 41 |
| 08 | Magnetic anatase–GO NC | 1345 | 1585 | | | 46 |
| 09 | rGO@Fe ₃ O ₄ | 1345 | 1570 | 1.01 | Representing an increase of defect density | 61 |
| 10 | M-GO | 1346 | 1595 | 1.0 | Increased intensity of both D and G bands attributed to the presence of Fe ₃ O ₄ | 60 |
| 11 | rGO/Fe ₃ O ₄ | 1334 | 1600 | | | 42 |
| 12 | Binary ferberite–graphene NCs | 1345 | 1603 | | | 28 |
| 13 | rGO/Fe ₃ O ₄ | 1306.92 | 1591 | 1.19 | Magnetite NPs were decorated on the surfaces of rGO sheets | 62 |
| 14 | rGO/Fe ₃ O ₄ | 1341 | 1596 | | | 16 |
| 15 | GO–MGH–chitosan | 1295–1347 | 1596–1609 | 1.16 | Involvement of oxygen-containing groups with the functional groups of CS and MGH | 69 |

positively charged radionuclide species, such as the hydrated cations UO₂²⁺, Cs⁺, and Sr²⁺, drawing them from the bulk solution to the sorbent surface.⁷⁴

5.2.2 Surface complexation. Beyond simple electrostatic attraction, oxygen-containing functional groups can act as powerful Lewis bases (ligands), forming stable coordination complexes with radionuclide ions. It can occur *via* the formation of inner-sphere complexes, in which the radionuclide ion binds directly to the surface functional groups without intervening water molecules, or outer-sphere complexes, in which the ion remains fully or partially hydrated and is bound *via* electrostatic forces. This mechanism is particularly dominant for actinides such as U(vi) and Th(iv), which have a high affinity for oxygen-donating ligands.^{6,74}

5.2.3 Ion exchange. The protons on the acidic carboxyl and hydroxyl groups, or other mobile counter-ions (like Na⁺ or K⁺) present on the NC surface, can be stoichiometrically exchanged for radionuclide ions from the solution. This mechanism is particularly relevant for the sorption of alkali and alkaline earth metals, such as Cs⁺ and Sr²⁺, which have a lower tendency to form strong covalent complexes.^{6,27}

5.2.4 Physical sorption and coagulation. The vast, delocalized π -electron system and the high specific surface area of the graphene sheets provide numerous sites for weaker physical sorption forces, such as van der Waals interactions. Furthermore, GO has been observed to act as a coagulant, effectively enmeshing and precipitating certain radionuclides, thereby contributing to their removal from the aqueous phase.^{3,14} The magnetic core of the NC then facilitates the rapid aggregation and magnetic separation of these coagulates.³⁰

5.3 Key factors influencing sorption efficiency

The practical performance of Fe₃O₄@graphene NCs as sorbents is highly susceptible to various operational parameters. Optimizing these factors is crucial for achieving maximum removal efficiency in real-world applications.

5.3.1 pH of the solution. The solution pH is arguably the most critical factor, as it simultaneously governs the sorbent's surface charge (specifically, its relationship to the point of zero charge, PZC) and the chemical speciation of the dissolved radionuclides. For U(vi), sorption is typically optimal at slightly acidic to neutral pH values of 4.0–7.0. At lower pH, the high concentration of H⁺ ions protonates the sorbent surface, leading to electrostatic repulsion with the cationic UO₂²⁺ species and intense competition for binding sites. At higher pH levels (>7.0), uranium speciates into stable, anionic uranyl carbonate or hydroxide complexes, which are repelled by the negatively charged sorbent surface, thereby reducing sorption.^{4,12,49,73} In contrast, the sorption of monovalent Cs⁺ is generally favored at neutral to alkaline pH, where the sorbent surface is more strongly deprotonated and negatively charged, and competition from H⁺ ions is minimal.^{19,25}

5.3.2 Initial ion concentration. The efficacy and kinetics of the sorption process are critically influenced by the initial concentration of the target radionuclide and the contact time between the sorbent and the contaminated solution. The initial concentration establishes the driving force for mass transfer from the aqueous phase to the sorbent surface. Generally, the sorption capacity increases with a higher initial ion concentration, as more ions are available to occupy the active binding sites. However, this trend continues only until the sorbent reaches saturation, at which point the capacity plateaus. The



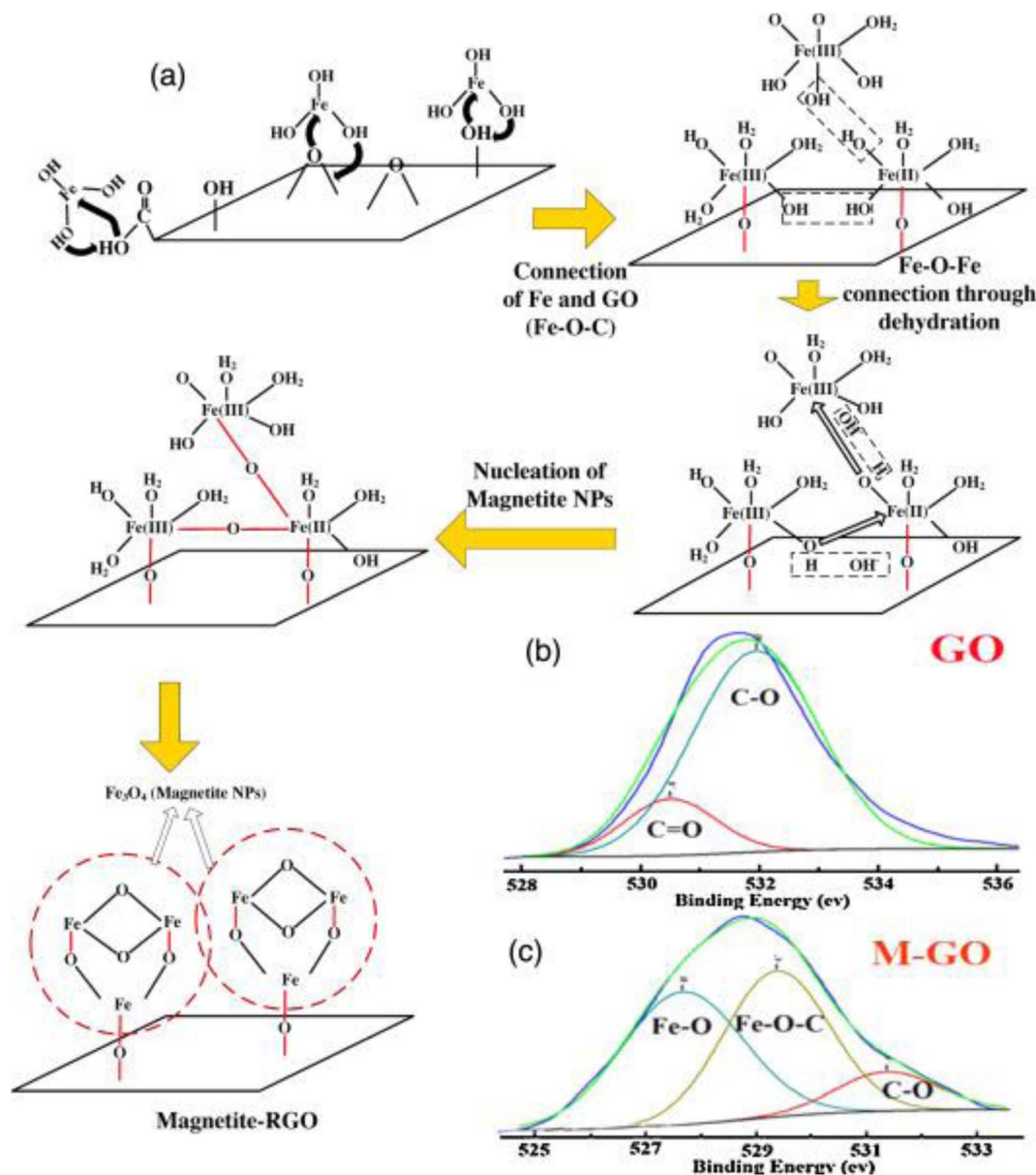


Fig. 15 (a) Proposed mechanism for the formation of Fe_3O_4 NPs on the GO surface, highlighting the formation of Fe-O-C bonds. High-resolution XPS O 1s spectra of (b) GO and (c) the M-GO composite, where the appearance of a peak corresponding to Fe-O-C confirms the chemical bonding.³³

relationship between initial concentration and equilibrium uptake is often modeled using sorption isotherms, such as the Langmuir or Freundlich models.^{13,18,43} For instance, studies on a magnetic graphene oxide NC showed that for an initial Sr^{2+} concentration range of 25–125 mg L^{-1} , the maximum sorption (29.98%) was achieved at 50 mg L^{-1} , while for Cs^+ in a range of 200–500 mg L^{-1} , the peak removal (57.2%) occurred at 250 mg L^{-1} .⁴³ Similarly, the sorption of Th(IV) on magnetic graphene oxide nanoribbons (MGONRs) increased with initial concentrations from 5 to 150 mg L^{-1} , reaching a maximum capacity of 31.4 mg g^{-1} at 150 mg L^{-1} .¹³ For Cs^+ removal using a $\text{UiO-66/Fe}_3\text{O}_4/\text{GO}$ composite, the sorption capacity peaked at 60 mg g^{-1} when the initial concentration was 100 mg L^{-1} .¹⁹

5.3.3 Contact time. The contact time required to reach sorption equilibrium is another crucial parameter that varies with the specific radionuclide, its initial concentration, and the sorbent properties. The sorption process is typically biphasic, beginning with a rapid initial uptake phase in which radionuclides are adsorbed onto readily accessible external surface sites. It is followed by a much slower, diffusion-controlled phase as the ions gradually migrate into the composite's interior porous structure. For example, a $\text{GO/Fe}_3\text{O}_4/\text{GC}$ composite demonstrated high-speed kinetics for U(VI) , removing over 98% within 30 min.⁴⁹ In another case, a $\text{rGO/Fe}_3\text{O}_4/\text{TW}$ composite adsorbed 96% of U(VI) in under one minute, although it took 2 h to reach final equilibrium.⁷³ In contrast, the sorption of Th(IV) onto MGONRs was slower, reaching equilibrium after 6 h.¹³ For

Table 8 Summary of elemental composition and chemical state information for Fe₃O₄@graphene-based NCs from XPS analysis

| No. | Composite name | Element | XPS peak | Binding energy (eV) | Chemical state | Ref. |
|-----|--|-------------|---|---|--|------|
| 1 | Fe ₃ O ₄ /GO | C, O, Fe | C 1s, O 1s, Fe 2p _{3/2} , Fe 2p _{1/2} | C 1s: 285.0, O 1s: 530.4, Fe 2p _{3/2} : 711.12, Fe 2p _{1/2} : 724.79 | C-C/C=C (sp ²) Anionic O ²⁻ from Fe ₃ O ₄ Fe ²⁺ /Fe ³⁺ (Fe ₃ O ₄ phase) | 55 |
| 2 | SMGO | C, O, Fe | C 1s, O 1s, Fe 2p _{3/2} , Fe 2p _{1/2} | C 1s: 284.5 (C-C), 286.0 (C-O), 288.2 (C=O), O 1s: 531.4 (Fe ₃ O ₄), 533.0 (C=O), 533.5 (C-O), Fe 2p: 710.7, 713.0 (Fe ²⁺ , Fe ³⁺), 724.4, 726.6 (Fe ²⁺ , Fe ³⁺), 719.3, 733.3 (satellites from γ-Fe ₂ O ₃) | C-C (sp ²), C-O (hydroxyl/epoxy), C=O (carbonyl/carboxyl) O ²⁻ in Fe ₃ O ₄ Fe ²⁺ /Fe ³⁺ in Fe ₃ O ₄ , minor γ-Fe ₂ O ₃ contribution | 38 |
| 3 | EDTA-mGO | Fe, N | Fe 2p, N 1s | Fe 2p: 710.2, N 1s: 400.3 | Fe ³⁺ in Fe ₃ O ₄ -NH or -N in EDTA anchored to mGO | 21 |
| 4 | Fe ₃ O ₄ /graphene | Fe, O | Fe 2p, O 1s | Fe 2p _{3/2} : 710.1, Fe 2p _{1/2} : 724.3, O 1s: 530.1 (O ²⁻), 531.5 (-COOH), 532.1 (-OH) | Fe ₃ O ₄ (Fe ²⁺ /Fe ³⁺) Oxygen in iron oxide, carboxylic acid, and hydroxyl groups | 12 |
| 5 | PB/Fe ₃ O ₄ /GO | C, O, N, Fe | C 1s, Fe 2p, N 1s | C 1s: 284.7 (C-C/C=C), 285.6 (C-O), 286.8 (C=O), 287.6 (C≡N), 288.9 (O-C=O), Fe 2p _{3/2} : 711.3, Fe 2p _{1/2} : 724.8, N 1s: 399.0, 398.6, 397.3 | Aromatic C, epoxy, carbonyl, cyano group (C≡N), carboxyl Fe ₃ O ₄ [Fe(CN) ₆] ⁴⁻ cyano nitrogen | 31 |
| 6 | M-GO | C, O, Fe | C 1s, O 1s, Fe 2p | C 1s: 285.0 (C-C/C=C), 286.2 (C-OH), 287.1 (C-O-C), 289.1 (O-C=O), Fe 2p: ~711.0, O 1s: 532.0 | Magnetic Fe ₃ O ₄ presence confirmed (Fe 2p ~ 711 eV) Oxygen functional groups reduced (~55%) | 33 |
| 7 | GO-Fe ₃ O ₄ | C, O, Fe | C 1s, O 1s, Fe 2p _{3/2} , Fe 2p _{1/2} | C 1s: ~285.0, O 1s: ~530.0, Fe 2p _{3/2} : 711.12, Fe 2p _{1/2} : 724.79 | sp ² carbon (C-C/C=C). Oxygenated groups Fe ₃ O ₄ (Fe ²⁺ /Fe ³⁺) | 43 |
| 8 | rGO/Fe ₃ O ₄ | C, O, Fe | C 1s, O 1s, Fe 2p _{3/2} , Fe 2p _{1/2} | C 1s: 284.4 (C-C/C=C), 287.4 (C=O), Fe 2p: 711.1 (2p _{3/2}), 724.7 (2p _{1/2}), O 1s: 530.5 (Fe-O), 532.0 (C=O), 533.4 (C-O) | Graphene backbone (C-C), carbonyl Fe ₃ O ₄ confirmed <i>via</i> Fe 2p, with Fe-O anchoring to rGO and oxygenated groups | 61 |
| 9 | rGO/Fe ₃ O ₄ | C, O, Fe | Fe 2p, O 1s, C 1s | Typical Fe 2p _{3/2} : 710–711 eV (not restated here) | Fe ₃ O ₄ and rGO functional groups | 65 |
| 10 | rGO/Fe ₃ O ₄ | C, O, Fe | C 1s, O 1s, Fe 2p | C 1s: 285, O 1s: 531, Fe 2p: 725 | sp ² C (graphene), oxygenated groups Fe ₃ O ₄ phase | 16 |
| 11 | 3D MPBRGO | C, O, N, Fe | C 1s, Fe 2p, N 1s | C 1s: 284.5 (C-C/C=C), 285.7 (C≡N, from [Fe(CN) ₆] ⁴⁻), 286.5 (C-O), 287.8 (C=O), 288.9 (O-C=O), Fe 2p: 708.3 (Fe in PB), 711.3 (Fe ₃ O ₄ 2p _{3/2}), 724.9 (Fe ₃ O ₄ 2p _{1/2}), N 1s: 397.0, 398.0, 399.2 | sp ² C (graphene), cyano group (PB), epoxy, carbonyl, carboxylic Prussian blue ([Fe(CN) ₆] ⁴⁻), magnetite (Fe ₃ O ₄) Cyano nitrogen from [Fe(CN) ₆] ⁴⁻ | 45 |
| 12 | PB/Fe ₃ O ₄ /GO | C, O, N, Fe | C 1s, Fe 2p, N 1s | C 1s: 284.8 (C=C/C-C/C-H), 285.8 (C-O), 286.8 (C=O), 287.6 (C≡N), 288.9 (O-C=O), Fe 2p: 711.3 (Fe 2p _{3/2}), 724.8 (Fe 2p _{1/2}), 708.3 (Fe 2p _{3/2} in [Fe(CN) ₆] ⁴⁻), N 1s: 397.5, 398.4, 399.6 | Aromatic graphitic carbon, epoxy, carbonyl, nitrile (C≡N), and carboxylic groups Fe ₃ O ₄ (magnetite) and Prussian blue ([Fe(CN) ₆] ⁴⁻) coexist C≡N from [Fe(CN) ₆] ⁴⁻ confirming PB presence | 25 |

Cs⁺ sorption on a UiO-66/Fe₃O₄/GO composite, 50% of the removal occurred within the first 10 min, with equilibrium being approached after 2 h.¹⁹ These examples highlight the rapid sorption kinetics characteristic of these materials, a key advantage for practical water treatment applications.

5.3.4 Temperature. The influence of temperature on the sorption process provides valuable thermodynamic insight. For many radionuclides, sorption onto these composites is endothermic, meaning that increasing temperature enhances sorption capacity. It can be attributed to several factors, including an increase in the diffusion rate of the ions from the bulk solution to the sorbent surface, potential swelling or changes in the sorbent's pore structure at higher temperatures, and an increase in the chemical activity and mobility of the radionuclide species in solution.^{12,17,19,20,25,73}

5.3.5 Presence of competing ions. Real-world wastewater is rarely a simple solution of a single contaminant. It is typically a complex matrix of coexisting ions that can compete with the target radionuclides for active sorption sites, thereby reducing overall removal efficiency. Cations with a higher charge density (*e.g.*, Al³⁺, Ca²⁺) or a smaller hydrated radius often exhibit stronger competition and can significantly reduce the removal efficiency for the target radionuclide. For example, the presence of common groundwater ions, such as K⁺ and Na⁺, can slightly decrease Cs⁺ sorption, whereas trivalent cations, such as Al³⁺, can significantly inhibit U(VI) sorption.^{19,26,49} Conversely, the presence of certain anions, such as CO₃²⁻, can in some cases enhance sorption by modifying the sorbent surface charge or by forming ternary surface complexes.¹⁰



Table 9 Summary of thermal stability and decomposition behavior of Fe₃O₄@graphene-based NCs from TGA analysis

| No. | Material | Stage | Temp. range (°C) | Weight loss (%) | Interpretation | Ref. |
|-----|--|-------|------------------|-----------------|---|------|
| 01 | CMC/MGOs | 1 | 120–350 | 73 | | 1 |
| 02 | EDTA–mGO | 1 | Before 200 | | Absorbed water | 21 |
| | | 2 | Higher than 200 | | Decomposition of EDTA | |
| 03 | FGC | 1 | 30–720 | 18.3 | Oxygen functional groups | 12 |
| | | 2 | 735–772 | 26.8 | Carbon skeleton | |
| 04 | Fe ₃ O ₄ /graphene | 1 | Till 300 | 0.03 | Evaporation of trace H ₂ O | 59 |
| | | 2 | 350–500 | | Organic molecules | |
| | | 3 | After 500 | | Formation of Fe ₃ O ₄ | |
| 05 | PB/Fe ₃ O ₄ /GO | 1 | Below 150 | | Loss of water | 31 |
| | | 2 | Around 200 | | Removal of coordinating water, oxygen functional groups | |
| | | 3 | 300–550 | | Combustion of GO and the decomposition of the cyano group | |
| | | 4 | Elevated to 950 | | Pyrolysis of the carbon skeleton | |
| 06 | NPs@GO | 1 | Below 120 | | Evaporation of adsorbed water | 36 |
| | | 2 | At 200 | | Decomposition of labile oxygen-containing functional groups | |
| 07 | FGC | 1 | Below 120 | | Evaporation of adsorbed water molecules | 30 |
| | | 2 | 120–350 | | Oxygen-containing functional groups | |
| | | 3 | 350–520 | | Decomposition of the carbon skeleton | |
| 08 | Fe ₃ O ₄ @GO | 1 | At 120 | | Evaporation of adsorbed water | 41 |
| | | 2 | 120–300 | | Organic functional groups | |
| | | 3 | 300–600 | | Combustion of the carbon skeleton | |
| 09 | APS | 1 | 200–400 | 28.4 | Organic molecules are decomposed partially | 46 |
| | | 2 | 400–600 | 18 | Siloxane bond breaking | |
| 10 | Fe ₃ O ₄ /RGO | 1 | At 100 | | Loss of moisture | 65 |
| | | 2 | 150–400 | | Functional group | |
| | | 3 | 400–500 | | rGO was converted into carbon dioxide, and Fe ₃ O ₄ was completely oxidized into Fe ₂ O ₃ | |
| | | 4 | After 500 | | Fe ₂ O ₃ particles | |

6.0 Challenges and future perspectives

6.1 Current challenges

While the scientific literature convincingly demonstrates the significant promise of Fe₃O₄@graphene NCs, several formidable challenges must be systematically addressed to facilitate the transition of this technology from the controlled environment of the laboratory to large-scale, real-world industrial applications. Overcoming these issues will be the central focus of future research and development in this field.

6.1.1 Scalability and cost-effectiveness. A significant impediment to commercialization is the scalability of production. Many of the current synthesis methods reported in the literature are complex, time-consuming, multi-step processes that are difficult to scale up while maintaining high quality and batch-to-batch reproducibility. Furthermore, they often rely on expensive and hazardous precursor materials, notably high-purity GO and various chemical reagents.^{75–77} Developing simple, continuous, and economically viable production processes is a prerequisite for the practical implementation.

6.1.2 Long-term stability and durability. The performance of these NCs must be reliable and predictable over extended operational periods and under harsh, variable chemical

conditions (*e.g.*, extreme pH, high salinity, presence of oxidizing/reducing agents) characteristic of many radioactive waste streams.^{78–80} Ensuring the long-term structural, chemical, and mechanical stability of the material and preventing the leaching of embedded iron oxide NPs remain key challenges.

6.1.3 Selective sorption. Real-world effluents are chemically complex, containing a diverse mixture of coexisting ions (*e.g.*, Na⁺, K⁺, Ca²⁺, Mg²⁺) that are often present at concentrations orders of magnitude higher than the target radionuclides. These ions can compete for active sorption sites, significantly reducing the efficiency and capacity for radionuclide removal.^{80,81} Enhancing the intrinsic selectivity of the NCs for specific high-risk contaminants, such as cesium-137 or strontium-90, is a major scientific and engineering hurdle.

6.1.4 Environmental and health impact. A comprehensive and rigorous life-cycle assessment of these nanomaterials is urgently required. The potential for the unintended release and leaching of the constituent NPs into the environment during use, regeneration, or final disposal must be thoroughly investigated. The long-term toxicity and ecological impact of these materials are still not fully understood,⁸² and it is imperative to ensure that the proposed remediation solution does not inadvertently create a new environmental problem.



Table 10 Summary of magnetic properties of various Fe₃O₄@graphene-based NCs from VSM analysis

| No. | Material | M_s (emu g ⁻¹) | M_r (emu g ⁻¹) | Magnetic behavior | Ref. |
|-----|---|------------------------------|------------------------------|--------------------|------|
| 01 | Fe ₃ O ₄ /GO | 48.6 | 0.785 | Super-paramagnetic | 55 |
| 02 | MGOC | 22.55 | | | 47 |
| 03 | SMGO | 41.38 | 3.5 | Super-paramagnetic | 38 |
| 04 | MGONRs | 33.7 | Neglectable | | 13 |
| 05 | mGO-PP | 14.63 | | | 68 |
| 06 | EDTA-mGO | 39.47 | | | 21 |
| 07 | Fe ₃ O ₄ /graphene | 45.6 | | | 12 |
| 08 | UiO-66/Fe ₃ O ₄ /GO | 15.53 | | | 19 |
| 09 | Fe ₃ O ₄ /RGO (5 : 1) | 10 | | | 35 |
| | Fe ₃ O ₄ /RGO (9 : 1) | 24 | | | |
| 10 | Fe ₃ O ₄ -NH ₂ /GO (3 : 1) | 58.26 | 0 | Super-paramagnetic | 50 |
| 11 | M-GO (stirred method) | 41 | | | 33 |
| | M-GO (ultrasound method) | 30 | | | |
| 12 | Fe ₃ O ₄ @GO | 40.4 | | Super paramagnetic | 41 |
| 13 | mGOi | 65 | | Super-paramagnetic | 56 |
| 14 | M-GO | 10.74 | | | 43 |
| 15 | Fe ₃ O ₄ @GO | 16.4 | | Super-paramagnetic | 70 |
| 16 | TBOT-GO-Fe ₃ O ₄ (hydrothermal) | 5.8 | 0.42 | Super-paramagnetic | 46 |
| | TBOT-GO-Fe ₃ O ₄ (sol-gel) | 2.8 | 0.20 | Super-paramagnetic | |
| 17 | MGO | 5.76 | 0 | Super-paramagnetic | 71 |
| 18 | Mag GO | 36.77 | 0 | Super-paramagnetic | 72 |
| 19 | MGO | 17.38 | Nearly zero | Super-paramagnetic | 57 |
| 20 | RGO/Fe ₃ O ₄ | 12.6 | 0 | Super paramagnetic | 42 |
| 21 | GN-Fe ₃ O ₄ | 10.23 | 0.03 | Super-paramagnetic | 58 |
| 22 | rGO/Fe ₃ O ₄ | 42 | | Super-paramagnetic | 62 |
| 23 | rGO/Fe ₃ O ₄ | 18.6 | Low | Super-paramagnetic | 16 |
| 24 | MNGO | 10.74 | | Super-paramagnetic | 67 |
| 25 | rGO/Fe ₃ O ₄ /TW | 10 | | Super-paramagnetic | 73 |
| 26 | Fe ₃ O ₄ /GO | 31.2 | | | 4 |
| 27 | MCGO | 30.21 | | | 40 |
| 28 | M/GO | 31 | | | 52 |

6.2 Future research directions

Future research efforts should be strategically directed at overcoming the challenges mentioned above through concerted innovation in material design, process engineering, and safety assessment.

6.2.1 Advanced and sustainable synthesis. There is a pressing need to develop and validate green, sustainable synthesis routes that use non-toxic, renewable, and low-cost precursors. The implementation of advanced manufacturing processes, such as continuous flow reactors or aerosol-based synthesis, could dramatically enhance scalability, reduce production costs, and improve the consistency and quality of the final product.^{83,84}

6.2.2 Tailored surface functionalization and hierarchical design. A highly promising avenue for future research is the rational design of the NC surface to create binding sites with exceptionally high affinity and selectivity for specific target radionuclides. This could involve advanced surface engineering techniques such as grafting specific chelating agents, developing ion-imprinted polymers on the surface, or creating hierarchical composites with other advanced materials like metal-organic frameworks (MOFs).^{85,86}

6.2.3 Process optimization and predictive modeling. Systematic optimization of the entire sorption-desorption process is needed to maximize performance and minimize

operational costs. The increasing use of advanced computational tools, such as density functional theory (DFT) and molecular dynamics (MD) simulations, can provide unprecedented insights into sorption mechanisms at the molecular level.^{87,88} This fundamental understanding can, in turn, guide the rational design of more effective and efficient sorbent materials.

6.2.4 Sustainable and efficient regeneration strategies. The long-term sustainability of this technology depends on the ability to regenerate and reuse the sorbent multiple times. Research into novel, less chemically intensive regeneration methods, such as electrochemical desorption, thermal swing, or microwave-assisted desorption, could significantly enhance overall efficiency and reduce secondary waste generated during regeneration.⁷⁸ The primary goal is to develop strategies that allow for dozens or even hundreds of reuse cycles without a significant loss of performance.

6.2.5 Integration and system-level development. Future work should move beyond batch-scale experiments and focus on integrating these magnetic NCs into practical, continuous-flow water treatment systems. It could involve their use in magnetically stabilized fluidized-bed reactors, incorporation into reactive membrane systems, or deployment in packed-bed columns.^{89,90} Developing portable, modular, and scalable



Table 11 Summary of the sorption performance of various magnetic graphene derivatives for the removal of radionuclides^a

| Materials | Tested radionuclides | Sorption conditions | Kinetic model, isotherm model | Maximum sorption capacity | Ref. |
|---|----------------------|---|---|--|------|
| CMC/MGOs | U(vi) | pH = 5.5, $T = 301$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 7.94×10^{-4} mol g ⁻¹ | 1 |
| Fe ₃ O ₄ /GO | U(vi) | pH = 5.5, $T = 293$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 69.49 mg g ⁻¹ | 4 |
| PB/Fe ₃ O ₄ /GO | Cs(i) | pH = 7.0, $T = 298$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 55.56 mg g ⁻¹ | 25 |
| Fe ₃ O ₄ /GO | U(vi) | pH = 5.5, $T = 298$ K, $t = 40$ min | Pseudo-second-order, Langmuir | 176.47 mg g ⁻¹ | 12 |
| Fe ₃ O ₄ /GO | Th(iv) | pH = 3.0, $T = 298$ K, $t = 6$ h | Pseudo-second order, Freundlich | 36.54 mg g ⁻¹ | 13 |
| Fe ₃ O ₄ /PANI/GO | Sr(ii) | pH = 3.0, $T = 293$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 37.17 mg g ⁻¹ | 17 |
| rGO/Fe ₃ O ₄ | Cs(i) | pH = 7.0, $T = 298$ K, $t = 6$ h | Pseudo-second-order, Freundlich | 128.2 mg g ⁻¹ | 16 |
| | Sr(ii) | | | 384.6 mg g ⁻¹ | |
| UiO-66/Fe ₃ O ₄ /GO | Cs(i) | pH = 7.0, $T = 298$ K, $t = 12$ h | Pseudo-second-order, Langmuir | 62.07 mg g ⁻¹ | 19 |
| Fe ₃ O ₄ /GO | U(vi) | pH = 5.0, $T = 318$ K, $t = 5$ h | Pseudo-second-order, Langmuir | 3.50×10^{-4} mol g ⁻¹ | 20 |
| Fe ₃ O ₄ /EDTA/GO | U(vi) | pH = 5.5, $T = 298$ K, $t = 100$ min | Pseudo-second-order, Langmuir | 277.43 mg g ⁻¹ | 21 |
| Fe ₃ O ₄ /PB/GO | Cs(i) | pH = 7.0, $T = 298$ K, $t = 24$ h | Langmuir | 362 mg g ⁻¹ | 26 |
| FeWO ₄ /GO | U(vi) | pH = 6.0, $T = 298$ K, $t = 1-2$ h | Pseudo-second-order, Langmuir | 455 mg g ⁻¹ (1 h) 625 mg g ⁻¹ (2 h) | 28 |
| PB/Fe ₃ O ₄ /GO | Cs(i) | pH = 7.0, $T = 298$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 43.52 mg g ⁻¹ | 31 |
| Fe ₃ O ₄ /GO | Sr(ii) | pH = 7.0, $T = 298$ K, $t = 5$ h | Pseudo-second-order, Langmuir | 13.7 mg g ⁻¹ | 33 |
| Fe ₃ O ₄ /chitosan/GO | U(vi) | pH = 5.0, $T = 313$ K, $t = 100$ min | Pseudo-second-order, Langmuir | 204.1 mg g ⁻¹ | 40 |
| GO-Fe ₃ O ₄ | Cs(i) | pH = 10, pH = 4.0, $T = 298$ K, $t = 2$ h | Pseudo-second-ordered, Dubinin-Radushkevich, Freundlich | 148.77 mg g ⁻¹ 17.92 mg g ⁻¹ | 43 |
| Fe ₃ O ₄ /PB/GO aerogel | Cs(i) | pH = 7.0, $T = 303$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 484.12 mg g ⁻¹ | 45 |
| Fe ₃ O ₄ /chitosan/GO | Co(ii) | pH = 7.0, $T = 298$ K, $t = 6$ h | Pseudo-second-order, Langmuir | 59.82 mg g ⁻¹ | 47 |
| GO/Fe ₃ O ₄ /GC | U(vi) | pH = 5.0, $T = 298$ K, $t = 30$ min | Pseudo-second-order, Langmuir | 390.70 mg g ⁻¹ | 49 |
| Fe ₃ O ₄ /GO | Sr(ii), Cs(i) | pH = 4.0, $T = 293$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 14.706 mg g ⁻¹ 9.259 mg g ⁻¹ | 54 |
| rGO/Fe ₃ O ₄ /TW | U(vi) | pH = 5.0, $T = 298$ K, $t = 24$ h | Pseudo-second-order, Langmuir | 104.95 mg g ⁻¹ | 73 |

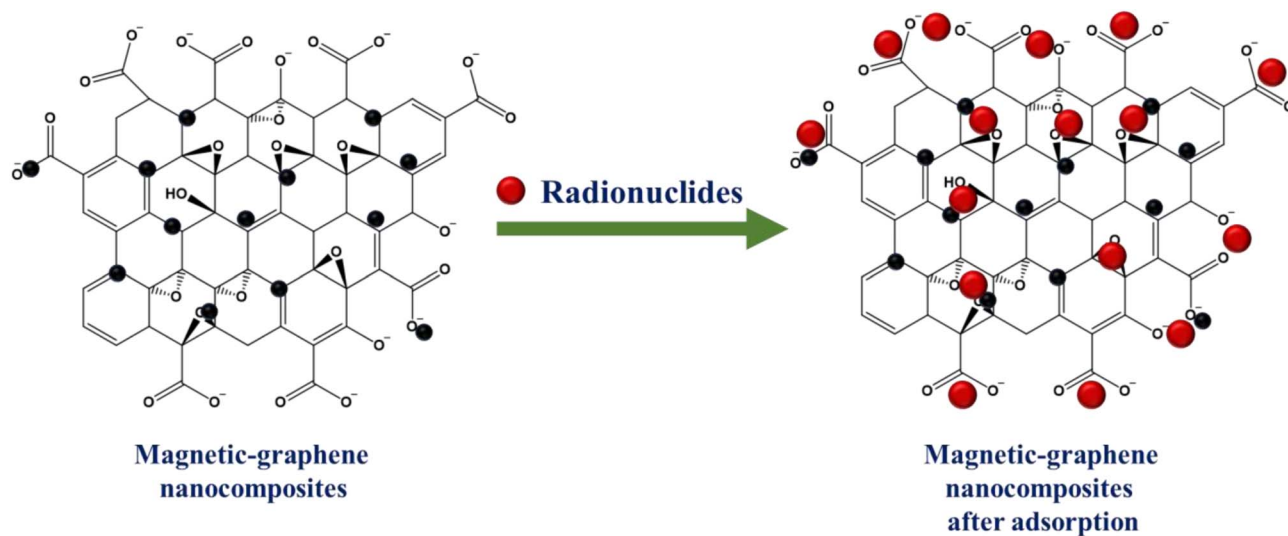
^a PB, Prussian blue; PANI, polyaniline.

Fig. 16 Schematic representation of the primary mechanisms for radionuclide removal by magnetic-graphene NCs, including electrostatic attraction, surface complexation, and ion exchange.

systems for on-site treatment could provide rapid and practical solutions for the remediation of contaminated sites.

6.2.6 Comprehensive safety assessment and regulatory frameworks. A concerted effort is required to conduct comprehensive toxicological and ecotoxicological studies to ensure the safe application of these materials throughout their entire life

cycle. The data is essential for establishing clear, robust regulatory guidelines for the production, handling, use, and ultimate disposal of these NCs, which will be critical for gaining public acceptance and facilitating their widespread, responsible adoption.⁹¹



7.0 Conclusion

Magnetic NCs based on Fe_3O_4 @graphene derivatives represent a significant and auspicious advancement in materials science, poised to address some of the most pressing challenges in radioactive waste management. Their unique and rationally designed architecture, which combines the vast, functionalizable surface of a graphene matrix with the powerful magnetic responsiveness of embedded Fe_3O_4 NPs, results in a multi-functional sorbent capable of efficiently capturing and removing a wide range of hazardous radionuclides from contaminated environments. This review systematically examines the key facets of these advanced materials, from the nuances of their synthesis and characterization details to the fundamental mechanisms that govern their sorptive performance. The high efficacy of these composites is clearly driven by a powerful synergy of electrostatic attraction, surface complexation, and ion exchange, with their practical performance being highly dependent on the optimization of operational conditions such as pH, temperature, and the composition of the aqueous matrix. Despite their well-demonstrated potential, the path from promising laboratory-scale materials to practical large-scale implementation is contingent on overcoming several significant and interconnected challenges. Critical issues of scalable, cost-effective synthesis, ensuring long-term stability and durability in harsh chemical environments, and, perhaps most importantly, enhancing sorption selectivity in complex, multi-component ionic matrices remain paramount. Furthermore, a rigorous assessment of their potential environmental and health impacts throughout their life cycle is essential for responsible, sustainable technological development. Future research must therefore be strategically focused on innovative and green synthesis techniques, the rational design of surface functionalities for highly targeted radionuclide capture, and the development of robust and efficient regeneration protocols. By systematically addressing these challenges through interdisciplinary research, Fe_3O_4 @graphene NCs can evolve from a scientific curiosity into a pivotal, deployable technology for mitigating the pervasive risks of radioactive contamination, thereby contributing to a safer, more sustainable future.

Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All data generated or analyzed during this study are included in this article.

Acknowledgements

Funding for this research was provided by the Japan Society for the Promotion of Science (JSPS) KAKENHI (Grant No. 24K15337 to IR), the Research and Innovation Centre (RIC) at Khulna

University (Memo No. KU/RIC-04/2000-323 to MM & IR), and the Environmental Radioactivity Research Network Center (ERAN) at Fukushima University (Grant No. I-25-10 to MM & IR). During manuscript preparation, Gemini Advanced, QuillBot, and Grammarly were employed to assist with paraphrasing and language editing. The authors have verified the content and accept full responsibility for the published work.

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