

Preparation and Microwave Absorption Properties of Graphene/Pure Ferrite Composite Absorbers

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Title: Preparation and Microwave Absorption Properties of Graphene/Pure Ferrite Composite

Absorbers

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Abstract: To develop lightweight, broadband, and high-efficiency graphene-based microwave absorbing materials, graphene/pure ferrite composite absorbers with different mass ratios (N1–W3) were prepared via a hydrothermal method. The effects of the graphene-to-ferrite mass ratio on the electromagnetic parameters and absorption performance were systematically investigated. The microstructure was characterized by XPS, SEM, and TEM, and the absorption mechanisms were discussed. Results show that ferrite particles (0.1–0.5 μm) are uniformly loaded on the surface of graphene sheets, forming a composite structure. Electromagnetic parameter analysis indicates that with increasing ferrite content, the imaginary part of permeability increases, enhancing magnetic loss; the synergistic effect of dielectric loss and magnetic loss effectively improves impedance matching. At an absorber thickness of 3 mm, sample W3 (graphene: pure ferrite = 1:3) exhibits a maximum reflection loss of -23.8 dB at 11.8 GHz, with an effective absorption bandwidth of 3.7 GHz, demonstrating optimal performance. Microstructural analysis reveals that interfacial polarization, multiple reflections, and defect-induced polarization relaxation between graphene and ferrite are key to the excellent absorption properties. This study provides experimental evidence and design strategies for developing high-performance graphene-based composite absorbers.

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1 Introduction

Currently, common electromagnetic wave absorbing materials include carbon materials, magnetic metals, ferrites, conductive polymers, and multicomponent composites [1–3]. Among them, carbon-based materials possess low density, good electrical conductivity, and strong dielectric loss capability, attenuating electromagnetic waves via conduction loss, dipole polarization, and interfacial polarization [4]. Graphene and its derivatives possess a two-dimensional lamellar structure, large specific surface area, abundant defects, and tunable conductivity, enabling effective electromagnetic wave attenuation through conduction loss and interfacial polarization, thus attracting extensive attention in the field of microwave absorption [5]. In cement-based materials, a small amount of graphene can not only improve the microstructure of the matrix but also form conductive networks and multiple reflection paths, thereby enhancing dielectric loss capacity [6,7]. However, when graphene is used alone as an absorbing component, excessive content tends to cause overly strong conduction loss, leading to poor impedance matching, which causes incident waves to reflect at the material surface and hinders further improvement in absorption performance [8].

Ferrites are typical magnetic-loss-type absorbers, mainly including Fe_3O_4 , MnZn ferrite, NiZn ferrite, CoFe_2O_4 , and ZnFe_2O_4 . Ferrite materials exhibit good magnetic response, chemical stability, and cost advantages, and can attenuate electromagnetic energy through natural resonance,

exchange resonance, eddy current loss, and hysteresis loss [9]. Previous studies have shown that introducing ferrite into cement or mortar systems can improve electromagnetic wave absorption, especially in broadening the absorption band and enhancing magnetic loss contribution. However, single ferrite materials generally suffer from high density, insufficient dielectric loss, limited absorption bandwidth, and unsatisfactory impedance matching. Therefore, combining ferrite with carbon-based materials such as graphene can improve absorption performance through the synergy of magnetic loss and dielectric loss.

Ferrite/graphene composites can simultaneously exploit the magnetic loss advantage of ferrite and the dielectric loss advantage of graphene, representing an important direction in recent research on electromagnetic wave absorbers. Ferrite particles loaded on the surface of graphene sheets can, on one hand, inhibit graphene sheet stacking and improve ferrite particle dispersion; on the other hand, the numerous heterogeneous interfaces formed between the two components facilitate interfacial polarization and space charge accumulation, thus enhancing electromagnetic wave attenuation [10]. Moreover, the graphene sheet structure prolongs the propagation path of electromagnetic waves inside the composite, while ferrite particles provide magnetic loss centers; their combined action helps improve impedance matching and increase effective absorption bandwidth [11-13]. Relevant studies indicate that the composite of graphene with MnZn ferrite can significantly improve the electromagnetic absorption performance of mortar materials; multiwalled carbon nanotube/ferrite/GO composite systems also exhibit high microwave shielding and attenuation capabilities [14,15].

Based on the above background, this paper adopts a hydrothermal method to prepare graphene/pure ferrite composite absorbers with different mass ratios, systematically studies the

influence of the graphene/ferrite mass ratio on electromagnetic parameters and absorption performance, characterizes the microstructure using XPS, SEM, and TEM, and discusses the absorption mechanisms, aiming to provide experimental evidence and design strategies for developing high-performance graphene-based composite absorbers.

2 Experimental

2.1 Preparation of Ferrite

Ferroferric oxide (Fe_3O_4) was prepared by a hydrolysis titration method. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ were mixed at a molar ratio of $\text{Fe}^{2+}:\text{Fe}^{3+} = 1.2:2$. Excess ammonia solution was added dropwise under continuous stirring to adjust the pH to 10–11. The mixture was stirred rapidly in a 50 °C water bath for 1.5 h, washed with deionized water to neutrality, and dried under vacuum for later use.

2.2 Preparation of Graphene

Graphene oxide (GO) was prepared by the Hummers method and then thermally reduced to obtain reduced graphene oxide (rGO). Graphite powder, sodium nitrate, and concentrated sulfuric acid were mixed in an ice bath. Potassium permanganate was added, and the reaction was carried out at low temperature, then heated to 35 °C and stirred for 6–12 h. After dilution, H_2O_2 treatment, centrifugation, washing, and drying, GO powder was obtained. It was then heat-treated in a tube furnace at 800–1000 °C for 1 h under argon protection to obtain rGO.

2.3 Preparation of Graphene/Pure Ferrite Composite Absorbers

The composite absorbers were prepared via a hydrothermal method. An appropriate amount of graphene was ultrasonically dispersed in 40 mL of deionized water. Self-prepared pure ferrite powder was added according to the ratios listed in Table 1, followed by ultrasonic stirring until

dissolution. Ammonia solution (25 wt%) was added dropwise to adjust the pH to 11. The mixture was transferred into a 100 mL Teflon-lined autoclave (70% filling degree) and reacted at 180 °C for 12 h. After natural cooling, the product was filtered and dried under vacuum at 60 °C for 12 h to obtain the composite absorber powder. For coaxial ring samples, the paraffin/powder mixture (4:1 by mass) was pressed at 10 MPa into rings with an inner diameter of 3.04 mm, an outer diameter of 7.00 mm, and a thickness of approximately 3.00 mm.

Table 1 Mass ratio table of grapheneloaded ferrite composite absorbers

Sample	Grapheneloaded:ferrite
N1	4:1
N2	3:1
N3	2:1
W1	1:1
W2	1:2
W3	1:3

3 Performance Study of Graphene/Ferrite Absorbers

3.1 Electromagnetic Parameter Analysis

Fig. 1(a), (b), (c), and (d) present the real part of permittivity, imaginary part of permittivity, real part of permeability, and imaginary part of permeability of the grapheneloaded pure ferrite composite absorbers, respectively. The real and imaginary parts of complex permittivity vary significantly with frequency. Sample N1, with the highest graphene content, exhibits the strongest dielectric energy storage and loss capability (ϵ' up to 43.57). In contrast, sample W2 shows the

lowest permittivity due to the weakened conductive network caused by increased ferrite proportion. Moreover, the imaginary part of permeability (μ'') gradually increases with higher ferrite content, indicating enhanced magnetic loss.

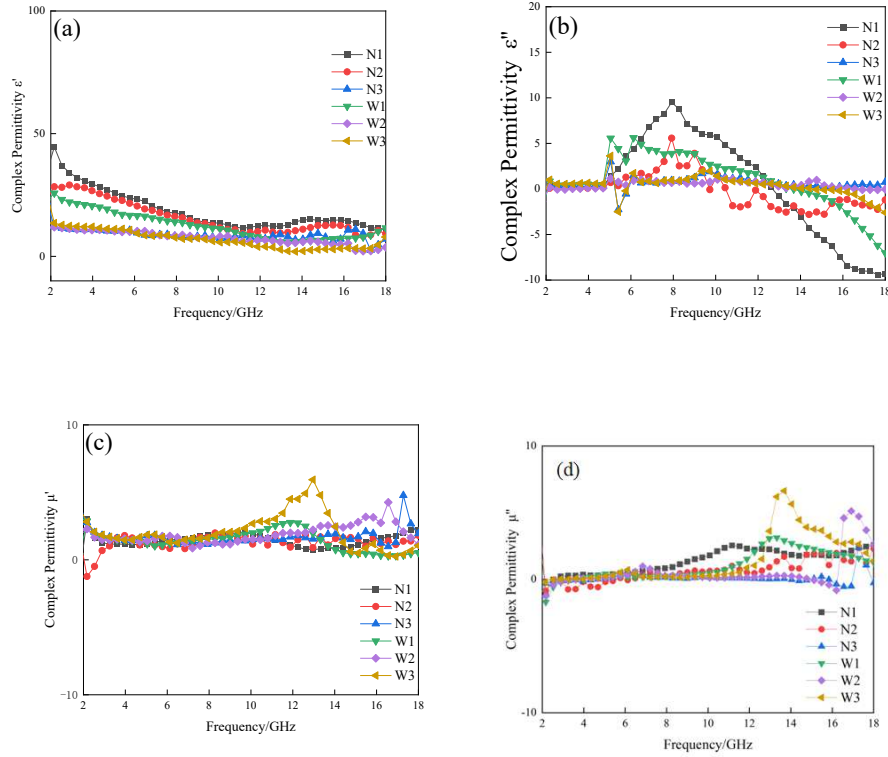


Fig. 1 Dielectric constant and permeability curves of graphene-loaded pure ferrite composite absorbers. (a) Real part of dielectric constant, (b) Imaginary part of dielectric constant, (c) Real part of permeability, (d) Imaginary part of permeability.

From Fig. 1(c), the real part of permeability (μ') ranges from -1.13 – 2.99 , -1.23 – 9.03 , -8.98 – 7.36 , 0.28 – 2.88 , 0.87 – 3.26 , and -8.55 – 6.83 , respectively. From Fig. 1(d), the imaginary part of permeability (μ'') varies within -0.99 – 3.10 , -2.7 – 5.95 , -5.53 – 2.41 , -1.92 – 3.13 , -3.05 – 5.12 , and -5.63 – 6.63 , respectively. As the ferrite content increases, the magnetic loss value μ'' gradually rises, which is mainly attributed to the magnetic nature of ferrite and its strong magnetic loss capability.

To identify the dominant absorption forms, the dielectric loss tangent and magnetic loss tangent of the samples are constructed, as shown in Fig. 2. The loss tangent values are calculated from the real and imaginary parts of permittivity and permeability using Equation (1); a larger loss tangent indicates stronger loss.

Owing to the combined effects of frequency dispersion and resonance, the real part μ' and imaginary part μ'' of permeability exhibit different frequencydependent trends. The loss tangents are calculated as follows:

$$\tan \delta_E = \frac{\varepsilon''}{\varepsilon'}, \tan \delta_M = \frac{\mu''}{\mu'} \quad (1)$$

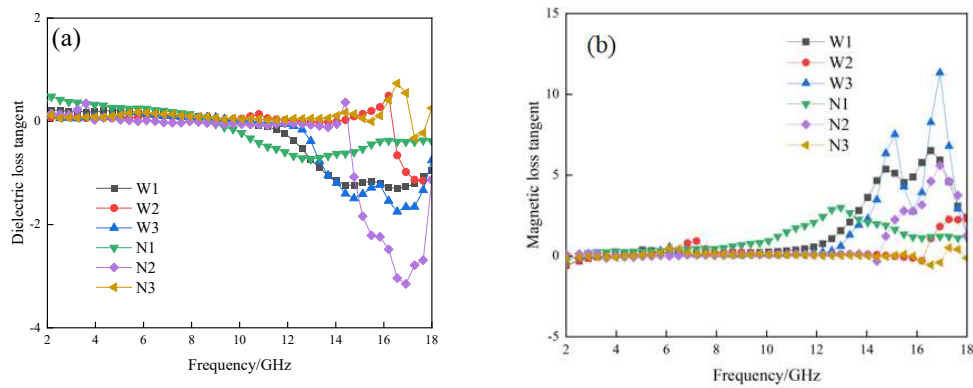


Fig. 2 Loss tangent values of graphene-loaded pure ferrite composite absorbers. (a) Dielectric loss tangent, (b) Magnetic loss tangent.

Based on the spectral response analysis in Fig. 2(a), in the 2–18 GHz range, the dielectric loss factor ($\tan \delta_E$) of each system exhibits significant frequency dispersion, and its magnitude decreases linearly with increasing ferrite doping (a 42% reduction from W1 to W3), which is

attributed to the gradient change in conductive network density caused by ferrite introduction. Comparing with the magnetic loss spectra in Fig. 2(b), the magnetic loss factor ($\tan\delta_\mu$) shows typical relaxation-enhanced behavior with increasing frequency, and the increase is more pronounced at higher ferrite contents, correlating with the natural resonance and eddy current suppression effects of ferrite. Notably, for sample W3, the dielectric loss tangent and magnetic loss tangent are relatively close, which not only improves impedance matching but also fully utilizes the dual roles of dielectric and magnetic losses, significantly enhancing absorption performance.

3.2 Microwave Absorption Performance Analysis

The reflection loss (RL) of the absorber can be calculated using Equations (2) and (3):

$$RL(\text{dB}) = 20 \lg \left| \frac{Z_{\text{in}} - Z_0}{Z_{\text{in}} + Z_0} \right| \quad (2)$$

$$Z_{\text{in}} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh \left[j \left(\frac{2\pi f d}{c} \right) \sqrt{\mu_r \epsilon_r} \right] \quad (3)$$

where c is the speed of light in air, f is the frequency, d is the sample thickness, ϵ_r and μ_r are the complex permittivity and permeability, Z_{in} is the input impedance, and Z_0 is the freespace impedance.

The reflection loss (RL) is a key parameter for evaluating absorption performance; lower RL

values indicate better absorption. An RL below -10 dB means that more than 90% of the incident wave is absorbed, which is a standard criterion for practical absorbers. Fig. 3 and Fig. 4 display the reflection loss curves and impedance matching of the graphene-loaded pure ferrite composite absorbers. From Fig. 3, it is evident that higher ferrite content significantly improves absorption performance. Moreover, all samples with different ratios show good absorption. Sample W3 exhibits a broad reflection loss threshold in the measured frequency range, indicating good absorption in the mid-to-high frequency region. At a matching thickness of 3 mm, sample W3 reaches a maximum reflection loss of -23.8 dB at 11.8 GHz.

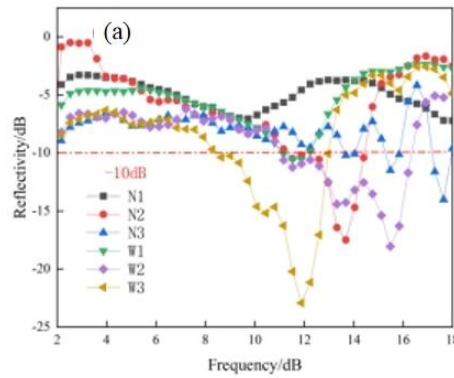


Fig. 3 Reflection loss curves of graphene/pure ferrite composite absorbers.

The broadband absorption of pure ferrite originates from its high magnetic loss, while the introduction of graphene enhances dielectric loss, forming a synergistic effect that yields excellent absorption. By selecting different ferrite substrates and combining them with graphene, the frequency response characteristics can be flexibly designed, providing a material basis for targeted electromagnetic shielding applications. At a matching thickness of 3 mm, sample W3 reaches a maximum reflection loss of -23.8 dB at 11.8 GHz.

Impedance matching is a core parameter characterizing the energy coupling efficiency at the electromagnetic wave–material interface; its essence is to reduce Fresnel reflection losses caused

by abrupt changes in wave impedance. According to transmission line theory, when the modulus of normalized wave impedance approaches unity, the minimum reflection coefficient $\Gamma = (Z_{in} - Z_0)/(Z_{in} + Z_0)$ is achieved. In practice, it is necessary to coordinate the synergy between dielectric and magnetic loss mechanisms: for dielectric loss dominated systems, excessively high ϵ'' often leads to impedance imaginary part mismatch, which can be mitigated by multilayer gradient impedance designs; while magnetic loss dominated materials ($\mu''/\epsilon'' > 3$) tend to exhibit overmatching at low frequencies due to a sharp increase in μ'' . Optimization should combine Cole–Cole model analysis of relaxation peak distributions and use genetic algorithms to invert the optimal ϵ – μ combinations, ultimately achieving quasi-ideal matching conditions with $|Z| \in [0.8, 1.2]$ over a broad band, providing sufficient energy dissipation paths for polarization relaxation and resonance losses.

Fig. 4 shows the impedance matching characteristic curves of the graphene/ferrite composite absorbers, with (a) for the graphene loaded pure ferrite system. The impedance modulus $|Z|$ ($= |Z_{in}/Z_0|$) is used as the evaluation index; when $|Z|$ approaches 1, the material exhibits optimal impedance matching, minimizing reflection at the interface and allowing efficient entry and dissipation of electromagnetic energy.

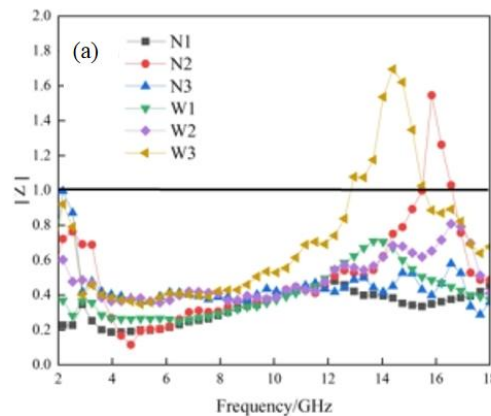


Fig. 4 Impedance matching curves of graphene/pure ferrite composite absorbers.

In Fig. 4(a), sample W3 shows a significant impedance matching advantage, with $|Z|$ reaching 1.008 at 12 GHz, exactly coinciding with the frequency of minimum reflection loss. This confirms that the composite of graphene and pure ferrite substantially improves impedance matching, providing a prerequisite for excellent absorption. It is worth noting that the improvement in impedance matching stems from the synergistic effect of graphene and ferrite in dielectric and magnetic losses; our results demonstrate that through compositional design and process optimization, the impedance matching characteristics of composite absorbers can be precisely tuned, which is of great guiding significance for developing novel broadband and high-efficiency absorbers

To further benchmark the overall performance of the prepared W3 sample, a comparative summary with recently reported similar graphene/ferrite-based composites is provided in Table 2.

Table 2 Comparison of microwave absorption performance of W3 with recently reported graphene/ferrite-based composite absorbers

Absorber material	Thickness (mm)	Peak RL (dB)	Effective		Ref.
			bandwidth	Frequency	
			(GHz) (RL < -10 dB)	range (GHz)	
Graphene/pure ferrite (W3)	3.0	-23.8	3.7	2-18	This work
GO/Cr-substituted Co-Ni ferrite	2.5	-45.0	4.0	Ku band (12-18)	[4]

N-rGO/ZnFe ₂ O ₄ porous foam	2.0	−55.4	6.4	2–18	[8]
Ferrite-rGO (covalent bonding)	2.8	−38.2	4.2	2–18	[12]
GO-ferrite hybrid framework	2.5	−35.0	3.8	8–18	[14]

3.3 Microstructural Analysis

3.3.1 Xray Photoelectron Spectroscopy (XPS) Analysis

XPS, SEM, and TEM were jointly used to characterize the prepared absorbers. XPS was employed to investigate the surface elemental composition and chemical states. Table 3 lists the atomic percentages of elements in the composites determined by XPS.

Table 3 Atomic percentage content of elements in composite materials determined by XPS

Elemental content (%)	C	O	Fe	Ni
N1	82.03	16.9	1.07	0
W3	85.92	11.47	2.62	0

Table 3 shows that the materials are mainly composed of carbon (82.03% and 85.92%), with small amounts of oxygen (16.9% and 11.47%) and iron (1.07% and 2.62%). The XPS spectra in Fig. 5 further indicate that carbon exists mainly as C–C/C=C, C–O, and O–C=O functional groups, while

iron is present as Fe_3O_4 .

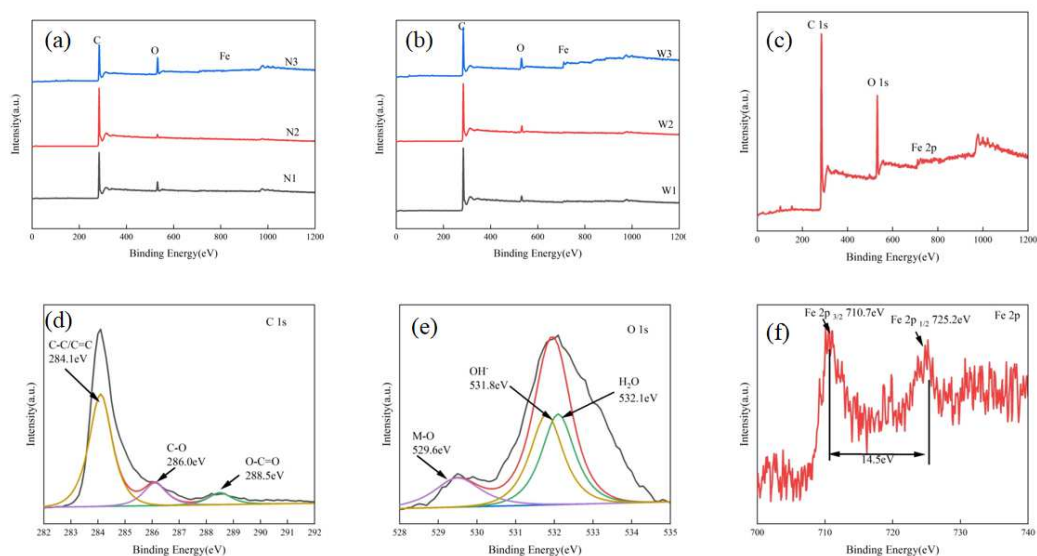


Fig. 5 XPS spectra of graphene-loaded pure ferrite composite absorbers. (a,b) Survey spectra of samples N1, N2, N3, W1, W2, W3; (c) Full spectrum of W3; (d) C 1s spectrum of W3; (e) O 1s spectrum of W3; (f) Fe 2p spectrum of W3.

Figs. 5(a) and (b) show the XPS survey spectra of samples N1, N2, N3, W1, W2, and W3. Fig. 5(c) is the full spectrum of W3, Fig. 5(d) is the C 1s spectrum, Fig. 5(e) is the O 1s spectrum, and Fig. 5(f) is the Fe 2p spectrum of W3. The spectra reveal that the graphene contains only carbon, oxygen, and iron. The C 1s spectrum of W3 (Fig. 5d) can be deconvoluted into three characteristic peaks at 284.1 eV, 286.0 eV, and 288.5 eV, corresponding to C-C/C=C, C-O, and O-C=O, respectively. The C-C/C=C peak intensity is much higher than the others, indicating that the carbon surface contains oxygen-rich groups, which facilitate strong bonding with metal oxides.

3.3.2 Scanning Electron Microscopy (SEM) Analysis

SEM was used to analyze the morphology of samples N1 and W3. As shown in Fig. 6, ferrite particles (size about 0.1–0.5 μm) are sparsely attached to the wrinkled graphene surface in some regions, with minimal agglomeration. This indicates that the modified ferrite particles can be

uniformly dispersed and bonded on the graphene sheets, attributed to reactions between modified ferrite and surface functional groups of graphene, enhancing dispersion and interaction. As ferrite content increases (Fig. 7 for W3), more particle accumulation and clearer interfaces are observed, suggesting weaker binding forces, yet still relatively uniform mixing.

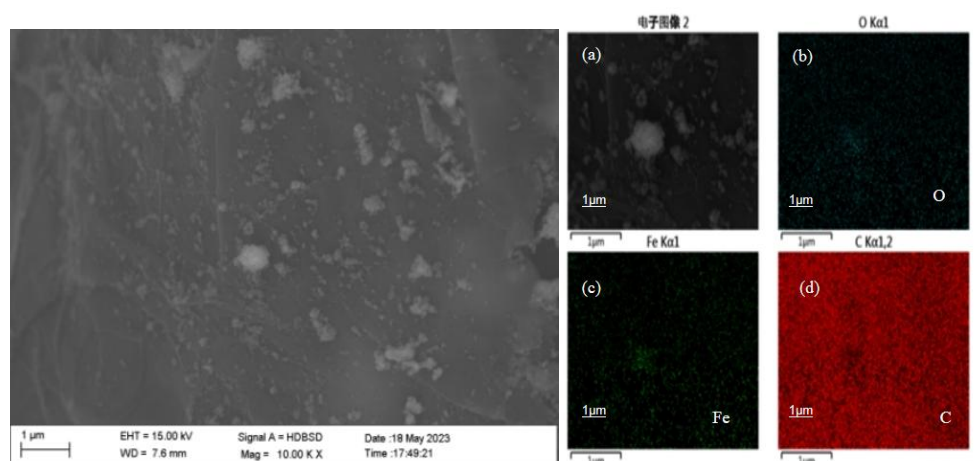


Fig. 6 SEM images of grapheneloaded pure ferrite absorber N1. (a) Electron image; (b,c,d) O, Fe, C element mappings.

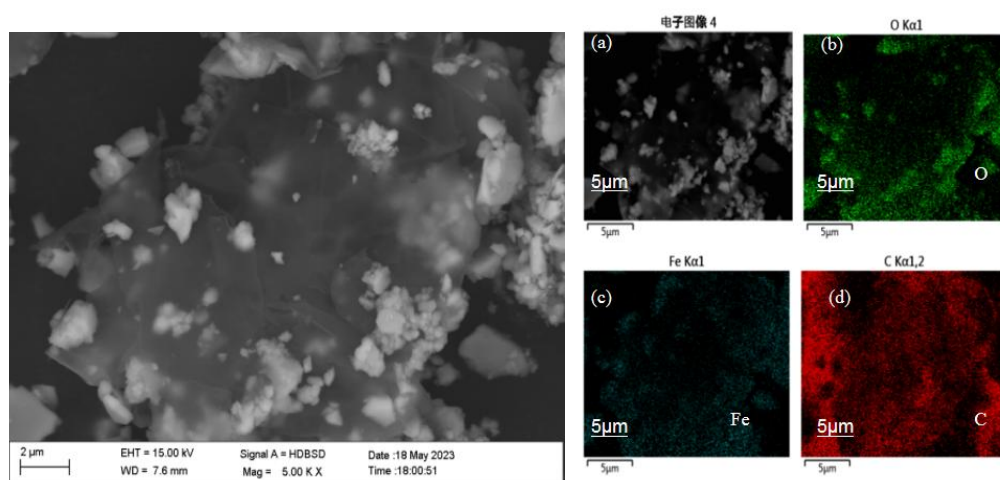


Fig. 7 SEM images of grapheneloaded pure ferrite absorber W3. (a) Electron image; (b,c,d) O, Fe, C element mappings.

SEM analysis (Figs. 6 and 7) shows that ferrite particles (0.1–0.5 μm) are mainly distributed on the graphene sheet surfaces without deep embedding. Sample N1 exhibits better particle

dispersion with a few particles attached to graphene wrinkles, indicating some interaction between modified ferrite and graphene surface groups; while sample W3, with higher ferrite content, shows obvious particle accumulation and weaker interfacial bonding, suggesting that simple physical mixing is insufficient to achieve homogeneous composite formation. These morphological features suggest that enhancing interfacial chemical bonding between ferrite and graphene would further improve absorption performance.

3.3.3 Transmission Electron Microscopy (TEM) Analysis

To further investigate the composite structure, TEM was performed on samples N1 and W3, as shown in Figs. 8 and 9.

Fig. 8 displays TEM images and EDS spectra of sample N1. From Fig. 8(a), a relatively uniform composite structure of graphene and ferrite particles is observed. Figs. 8(b) and (c) present EDS region mapping and total elemental distribution; Figs. 8(d–f) show C, Fe, and O element mappings, revealing widespread graphene distribution with local aggregation of ferrite, indicating that ferrite particles are wrapped by the graphene matrix.

TEM analysis of N1 reveals uniformly dispersed nanoscale particles with irregular polyhedral or nearspherical shapes, smooth surfaces, and good dispersion. EDS mapping shows homogeneous distribution of C, Fe, and O, with Fe closely matching particle morphology, confirming that iron-based species are the main active components, while uniform C distribution suggests a carbon coating structure. Such uniform elemental distribution and controllable morphology indicate potential magnetic applications.

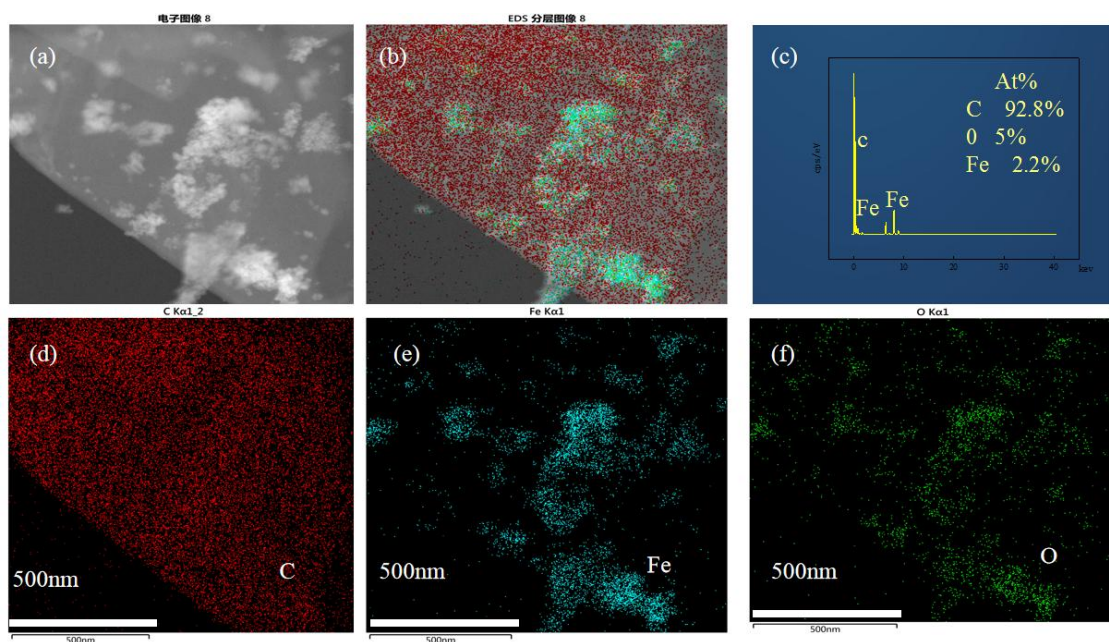


Fig. 8 TEM images of sample N1. (a) Electron image; (b) EDS region image; (c) Total element map; (d–f) EDSDMapping of C, Fe, O.

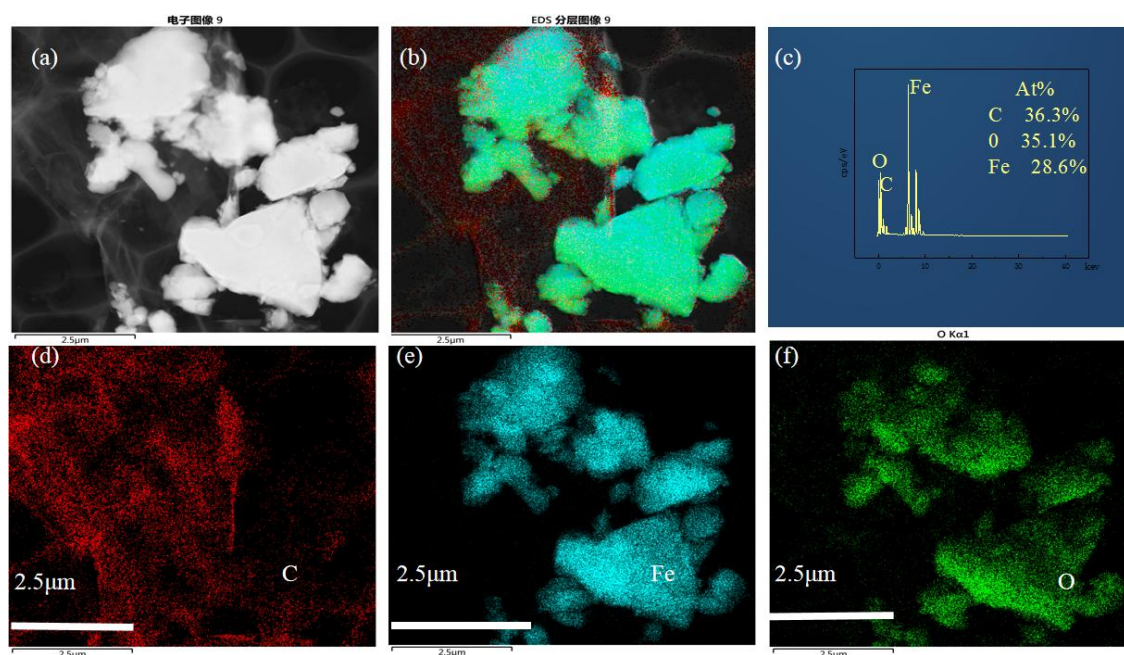


Fig. 9 TEM images of sample W3. (a) Electron image; (b) EDS region image; (c) Total element map; (d–g) EDSDMapping of C, Fe, O.

Combining XPS, SEM, and TEM results, Fig. 10 proposes an electromagnetic wave absorption

mechanism model for the graphene/pure ferrite composite, as follows:

- (1) Enhanced interfacial polarization: During hydrothermal synthesis, ferrite particles loaded on graphene introduce lattice defects and dislocations (e.g., oxygen vacancies), complicating the surface structure. These defects act as polarization centers, inducing interfacial polarization under the electromagnetic field, causing charge accumulation at the graphene–ferrite interface and forming dipoles.
- (2) Multiple reflections and resonance losses: Upon incidence, the graphene sheets and heterogeneous interfaces induce multiple reflections and scattering of electromagnetic waves, prolonging the propagation path. Meanwhile, ferrite particles provide natural resonance and exchange resonance losses, converting electromagnetic energy into heat.
- (3) Synergistic dissipation mechanism: Magnetic loss of ferrite (hysteresis, eddy current) and dielectric loss of graphene (dipolar polarization, conduction loss) cooperate, converting electromagnetic energy into heat via lattice friction and carrier migration. Lattice defects further increase carrier concentration and accelerate electron migration, thereby enhancing overall absorption.

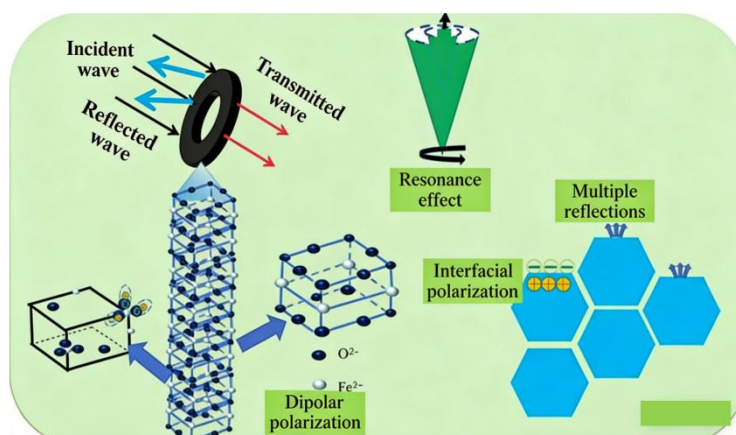


Fig. 10 Schematic diagram of electromagnetic wave absorption mechanism of the composite absorber.

Due to impedance mismatch between the absorber and air, partial reflection occurs at the graphene surface upon wave incidence, but most waves penetrate into the material. During propagation, waves undergo repeated reflection and scattering by the multilayer graphene structure, leading to continuous energy dissipation. Additionally, iron and carbon elements induce multiple scattering effects, synergistically enhancing attenuation. Ultimately, the excellent absorption arises from: (i) multimechanism synergy—coupling of magnetic loss (ferrite) and dielectric loss (graphene + interfacial polarization); (ii) efficient defect engineering—controllable defects serve as polarization centers and also optimize impedance matching via band tuning; (iii) structural advantages—combination of nanoparticle dispersion and graphene conductive network achieves a balance between "broadband absorption" and "strong attenuation".

4 Conclusions

In military applications, graphene/ferrite composite absorbers can be used as surface coatings for stealth equipment and military facilities to reduce radar crosssection and enhance stealth effectiveness. Their excellent absorption performance and tunable electromagnetic properties offer broad application prospects in stealth technology. This study prepared graphene/ferrite composite absorbers via a hydrothermal method, obtaining materials with good microwave absorption. The main conclusions are:

(1) Graphene in the absorber primarily functions through dielectric loss, while ferrite mainly contributes via magnetic loss. Their synergistic effect significantly improves the electromagnetic wave absorption performance. The absorption capability depends not only on the attenuation ability of the material but also on impedance matching and other factors.

(2) In the 2–18 GHz frequency range, sample W3 (graphene: pure ferrite = 1:3, thickness 3 mm) exhibits the best absorption performance, with a maximum reflection loss of –23.8 dB at 11.8 GHz and an effective absorption bandwidth of 3.7 GHz. The excellent performance is attributed to the synergistic effect of dielectric and magnetic losses, as well as good impedance matching.

Future work will focus on enhancing interfacial chemical bonding between ferrite and graphene through surface modification, and on evaluating the absorption performance over a broader frequency range (e.g., 1–40 GHz).

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Declaration of competing 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.

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CRediT authorship contribution statement

B.D. Cui: Writing -- original draft. X.G.Wang: Writing -- review & Editing and Investigation.