

Recent research progress and prospect of graphene-based microwave absorbing materials

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Abstract. With the increasing use of electronic devices, electromagnetic wave pollution has become a hot topic of concern and discussion, and microwave absorption materials play an important role in civil and defense military fields. Therefore, the development of electromagnetic wave absorption materials with low density, thin thickness, wide effective bandwidth, and strong wave absorption performance has become the focus and goal of researchers. Graphene shows a unique two-dimensional structure, lightweight, large specific surface area, high electrical conductivity, and high thermal conductivity, which make it a promising electromagnetic wave-absorbing material. However, pure graphene material cannot meet the needs of practical applications due to bad impedance matching. Therefore, graphene is usually combined with other materials (such as carbon materials, magnetic metal, polymer, ferrite, and ceramic) to improve its wave absorption properties. In this review, we summarize recent research progress of different types of graphene-based microwave absorbing materials. In addition, the challenges and prospects for improving the microwave absorption properties of graphene-based composites are presented, which will be useful for those working in related fields.

Keywords: microwave absorbing materials, graphene, composite, dielectric loss

1. Introduction

The rapid development and popularity of electronic devices today have brought great convenience to people's lives, but the resulting electromagnetic pollution problem is also becoming increasingly serious. Therefore, in recent years, high-performance wave-absorbing materials that can effectively absorb electromagnetic waves have gradually become a research hotspot. In addition, in modern warfare, radar stealth technology has become an important technical means to improve the capability of weapons systems. As the core of radar stealth technology, the research of wave-absorbing materials has received great attention from the world's military powers. Therefore, the development of high-performance electromagnetic wave-absorbing materials with the characteristics of "thin, wide, light, and strong" is of great importance for both civil and national defense.¹⁻⁵

Electromagnetic waves, as particle waves, can effectively transmit energy and momentum, and their absorption capacity is closely related to electromagnetic wave energy loss. Wave-absorbing materials are a type of functional materials that can convert the electromagnetic waves incident on the inside of the material into other forms of energy through loss. High-performance electromagnetic wave-absorbing materials are determined by two main aspects: 1, impedance matching characteristics, that is, how many electromagnetic waves are into the material inside; 2, Attenuation characteristics, that is, how much of the electromagnetic wave entering the interior is consumed. Until now, ferrite, metal magnets, dielectric ceramics, conductive polymers, and their hybrids play a dominant role in the field of microwave absorption. However, these traditional absorbing materials suffer from high density, poor environmental stability, corrosion, narrow absorption band, and other drawbacks, which limit them in actual application. Reflection loss (RL): a measure of the absorption capacity of electromagnetic waves, the higher the absolute value the better the absorption performance. Effective absorption bandwidth (EAB): the ability of a material to absorb electromagnetic waves in a specific frequency range.

Graphene was first obtained by two scientists, Andre Geim and Konstantin Novoselov, in 2004. Subsequently, numerous studies have shown that graphene's unique two-dimensional structure,

lightweight, large specific surface area, high electrical conductivity, and high thermal conductivity make it a promising material for electromagnetic wave absorption.

With the development of science and technology, absorbing materials not only must meet the requirements of lightweight, thin thickness, strong absorption, and wide frequency band, but also must meet the requirements of maintaining good microwave absorption performance in harsh environments, that is, good chemical stability, and other functions.⁶⁻¹⁰ Based on this, the absorption of electromagnetic waves by a single graphene material can no longer meet the requirements of practical applications. Therefore, it must be combined with other functional materials to combine the advantages of the two to prepare a wave-absorbing material with excellent performance.

In recent years, there has been a gradual increase in the number of literature reports on graphene-based composite microwave-absorbing materials. In this paper, we summarize and discuss the preparation methods, mechanism of action, and absorption properties of different graphene-based microwave-absorbing materials. At the same time, we present our views and opinions on the challenges and development prospects of graphene-based composite microwave-absorbing materials in the future.

2. Main Kinds Of Graphene-Based Microwave Absorbing Materials

Graphene derivatives (such as graphene oxide) contain various oxygen-containing functional groups (hydroxyl, carboxyl, and epoxy groups) and are often used as precursors for graphene-based composites. Graphene oxide can be reduced to obtain reduced graphene oxide (RGO) after reduction. In recent years, various graphene-based absorbing materials have been developed and studied. According to the different types of composite with graphene derivatives, we can classify graphene-based microwave absorbing materials into five categories, namely graphene-based carbon materials, graphene-based magnetic metal nanocomposites, graphene-based ferrite composites, graphene-based polymer composites, and graphene-based ceramic composites.

2.1 Graphene-based carbon material

Recently, graphene-based carbon materials consisting of only carbon materials have attracted attention in the field of electromagnetic wave absorption. Due to the identical electronegativity of graphene and the low number of defects in graphene, composites combining graphene and carbon can exclude their conductive and polarization losses, and have unique properties such as low mass and high chemical stability. Therefore, graphene/carbon composites are very promising in microwave absorption applications.

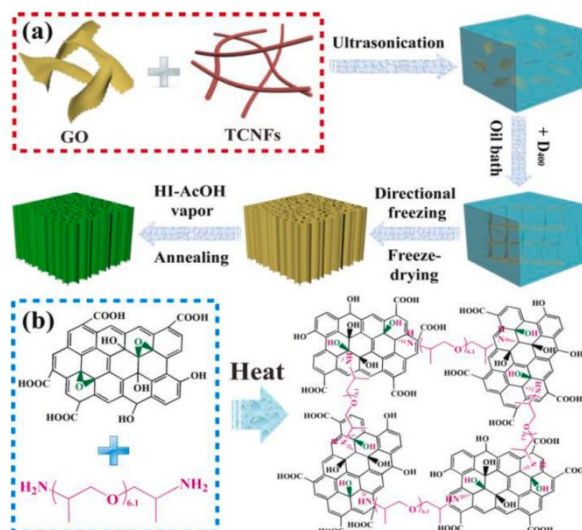


Figure 1: (a) Schematic illustration of constructing TGA; (b) cross-linked mechanism of GO nanosheets with D400 during the formation of TGOA13

Cao et al. obtained graphene/carbon fiber composite aerogels (CGAs) with lightweight and excellent mechanical properties through solvothermal reaction and freeze-drying methods. Specifically, a broad efficient absorption bandwidth of 8.72 GHz at 2 – 18 GHz and a high mean absorptivity of 97.4% at 0.3 – 1.5 THz are achieved, respectively. At the same time, the compressible CGA has excellent and stable EMI shielding properties, thermal insulation, and self-extinguishing properties. Generally, RL, EAB, and thickness of absorbers are the most important points for people to concern. During the research, Shu et al. found that dielectric loss and conductive loss were the most important influencing factors for microwave absorption performance.¹¹

In another work, Zhang et al. have prepared ultra-light and unidirectional honeycomb structured graphene/multi-walled carbon nanotube aerogel (GCA) with wide microwave absorption bandwidth by a simple directional freezing process and annealing treatment. The absorption bandwidth of less than 10 dB was 13.32 GHz in the frequency range of 2-18 GHz. The bandwidth of less than 20 dB reflection loss reached 13.5 GHz, covering the entire Ka-band (26.5-40 GHz) of existing 5G millimeter-wave communication technology. For the absorption mechanism, proper conductivity and dielectric tangent loss are the main causes of broadband absorption; polarization relaxation losses account for most of the dielectric losses. MWCNTs provide stability and flexibility for GCAs, improving conductivity and polarization.¹²

Kang et al. attached interwoven tubular carbon nanofibers (TCNFs) to a graphene backbone, chemically cross-linked, freeze-dried, and then reduced to obtain lightweight elastic TCNFs/graphene aerogels (TGAs) with anisotropic structures as shown in figure 1. It has the most outstanding microwave consumption, with a minimum reflection loss (RL_{min}) value of -46.1 dB and an effective absorption bandwidth (EAB) value of 7.1 GHz. an ultra-wide EAB of 11.5 GHz is achieved by adjusting the compression strain. For the mechanism, the hybrid aerogel with 80% compressibility has stronger polarization relaxation and sufficient multiple reflections and scattering in the vertical direction, thus obtaining an RL_{min} value of -17.7 dB and an EAB of 6.6 GHz. In addition to this, as the compressive strain increases, the conduction loss will strengthen and the multiple reflections and scattering will weaken.¹³

In addition to aerogel types, Lu et al. have successfully attached reduced graphene oxide (RGO) to cotton-derived flexible carbon fibers (CF) by electrostatic self-assembly, and then arranged two-dimensional layered double hydroxides (LDH) closely on the surface of CF/RGO to form a unique core-sheath structure. The composite shows an optimum reflection loss value of -60.9 dB at 10.3 GHz at a thickness of 2.5 mm; the effective absorption bandwidth value is 6.1 GHz when the fill rate is only 20wt%. By investigating the mechanism, the authors concluded that the combination of multiple loss mechanisms (dipole/interface polarization, conductive losses) and the optimum impedance characteristics due to the special structure gives CF/RGO/LDH excellent microwave absorption properties.¹⁴

Sun et al. have synthesized thin graphene-like multifold carbon nanosheets by a facile hydrothermal method and annealing of watermelon flesh. Their minimum reflection loss reaches -50.0 dB at a thickness of 3 mm and their effective absorption bandwidth reaches 3.51 GHz at a thickness of 2 mm. The authors found a significant improvement in the absorption properties of this material due to conductive losses, multiple reflections, and scattering as well as dipole polarization.¹⁵

2.2 Graphene-based magnetic metal nanocomposites

The combination of graphene-based and magnetic metal nanoparticles is characterized by the combination of graphene or graphene derivatives with magnetic metal nanoparticles (e.g. Fe, Co, Ni, and their alloys). Due to the high dielectric constant of graphene and the structure of magnetic metal nanoparticles, the interfacial interaction between the two will enable graphene to exhibit excellent electromagnetic wave absorption properties.

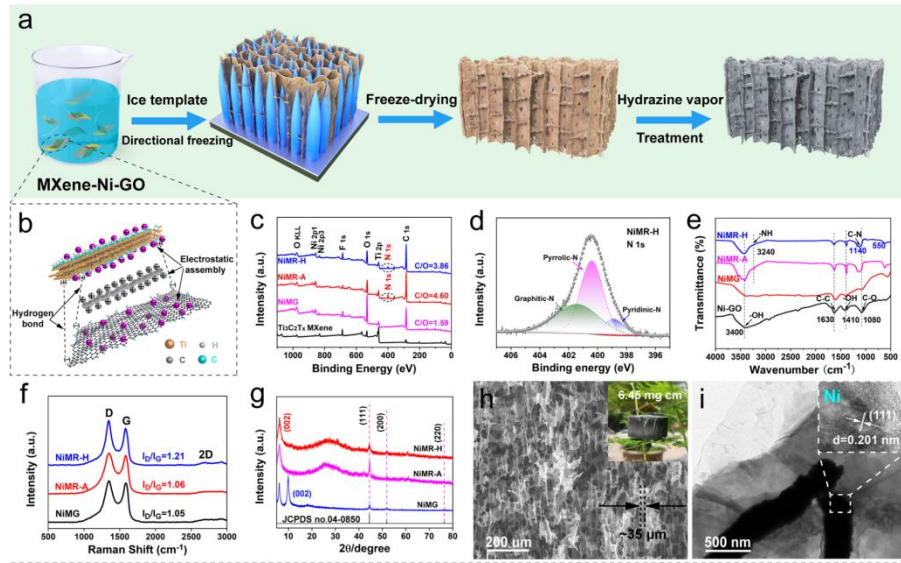


Figure 2: (a) Schematic illustration of fabricating a NiMR-H aerogel. (b) Electrostatic interaction and hydrogen-bonding effect between the MXene, Ni nanochain, and GO nanosheet. XPS spectra of (c) Ti₃C₂T_x MXene, NiMG, NiMR-A, and NiMR-H aerogels and (d) the NiMR-H aerogel at the N1s region. (e) FT-IR. (f) Raman spectra, and (g) XRD patterns of NiMR-H, NiMR-A, and NiMG aerogels. (h) SEM, photograph (inset). (i) TEM, HRTEM (inset).¹⁷

Hou et al. obtained reduced graphene oxide (RGO) modified with magnetic xNi/yNiO nanoparticles by a simple hydrothermal method, which exhibits a large reflection loss (RL) in the S-band. The maximum RL can reach -46.5 dB at 3.57 GHz with a thickness of 3.6 mm and a loaded wax ratio of 15wt%. In order to investigate the fundamental microwave absorption mechanism of the composite, the researchers have analyzed that good impedance matching and proper microwave damping processes in the optimized NNG composite are important to improve low-frequency attenuation and enhance absorption. Interfacial polarization resulting from charge aggregation on the surface of the NNG composite, and dipole polarization caused by functional groups and defects contribute to the excellent dielectric loss of the NNG composite. These properties make it simple to prepare, economical and cost-effective.¹⁶

In another work, Liang et al. have obtained a three-dimensional (3D) dielectric Ti₃C₂T_x MXene/reduced graphene oxide (RGO) aerogel with magnetic nickel nanochains anchored by a directional freezing method and hydrazine vapor reduction as shown in Figure 2. The prepared ultralight Ni/MXene/RGO (NiMR-H) aerogel (6.45 mg cm⁻³) provided the best EMA performance of any MXene-based absorbing material reported to date, with a minimum reflection loss (RL_{min}) of -75.2 dB (99.999 996% wave absorption) and the broadest EAB of 7.3 GHz. In addition, the composite has excellent structural robustness and mechanical properties, as well as high hydrophobicity and thermal insulation (close to air). The EMA mechanism of NiMR-H aerogels can be attributed to a combination of excellent impedance matching, multiple electromagnetic wave scattering, and synergistic electromagnetic loss effects.¹⁷

Additionally, Fe and Co also have attracted much attention for their microwave absorbing potential. Ding et al. have successfully fabricated FeCo/GO composites by a two-step method and subsequently measured the composites. The results show a maximum reflection loss (RL_{max}) value of -39.2 dB at 10.7 GHz, an effective bandwidth of 3.8 GHz (8.6-12.4 GHz) with RL_{max} less than -10 dB, and a thickness of 5.0 mm. For the mechanism, the dielectric pore structure of 2D graphene gives the composite a good impedance match and also enhances the dissipation strength of the energy absorbed by electromagnetic waves. A large number of carbon-air-magnetic powder interfaces formed strengthens the interfacial polarization of the composite, which results in increased dielectric and magnetic losses.¹⁸

2.3 Graphene-based ferrite composites

Combining non-corrodible ferrite with graphene to obtain a novel graphene-based material for electromagnetic wave absorption. Graphene-based ferrite composites not only have a wealth of physicochemical properties but also have synergistic effects resulting from the combination of graphene and magnetic metal nanoparticle composites. As a result, many graphene-based ferrite composites with excellent microwave absorption performance have been reported in recent years.

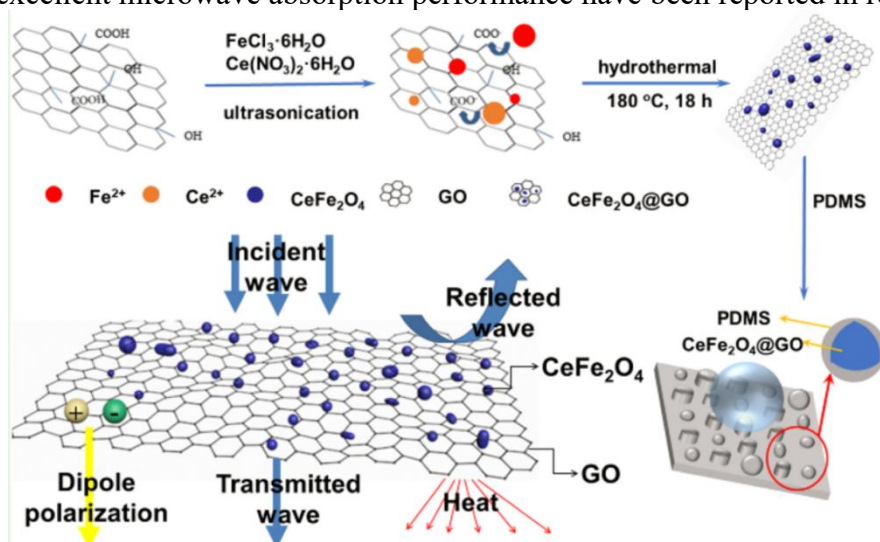


Figure 3: the process of a CeFe₂O₄/GO hybrid by a method of hydrothermal²²

Deng et al. prepared nitrogen-doped reduced graphene oxide/Ni ferrite composite foams through two-step solvothermal and hydrothermal methods. The minimum reflection loss was as high as 60.6 dB at an ultra-thin matched thickness of only 1.55 mm and the effective absorption bandwidth was up to 5.5 GHz at a thickness of 1.62 mm. For the microwave absorption mechanism of the composite foam, the authors have shown that impedance matching, conduction loss, dipole polarization, and interfacial polarization loss were important factors affecting the microwave absorption performance of the composite material¹⁹

Also, in a recent corresponding work, Wang et al. prepared graphite-coated multi-coiled NiFe₂O₄ composites by a facile two-step method. Overlapping conductive networks of flexible graphene configurations optimize the dielectric constants of the composites, and impedance matching proved to be best at lower filling concentrations, reaching the strongest reflection loss value of -48.1 dB. For the mechanism, the excellent performance of microwave absorbing was decided by conduction loss, multiple reflections, and scattering and magnetic-dielectric synergy effect. The composite material reached the strongest reflection loss of -48.1 dB with the best impedance matching in the result.²⁰

Additionally, Zhou et al. prepared three-dimensional patterned ZnFe₂O₄ ferrite (3D-ZFO) by two solution chemical precipitation processes, and then uniformly loaded the prepared 3D-ZFO onto graphene nanosheets (GNs) using ultrasound and heat treatment to obtain a new wave-absorbing material. The 3D-ZFO/GNs containing 5wt% GNs have an effective bandwidth of 5.56 GHz in the 2-18 GHz band and at a matched thickness of 1.5 mm. The authors proposed that the formation of ZnFe₂O₄ ferrite promoted magnetic loss and improved the impedance matching of a single dielectric material. the introduction of GNs promoted interfacial polarization and dipole polarization and enhances conductivity loss.²¹

Guo et al. successfully prepared a CeFe₂O₄/GO hybrid by a method of hydrothermal. Then they combined it with polydimethylsiloxane (PDMS) to get a composite material, which had excellent EM wave absorption ability and super-hydrophobicity. Finally, the CeFe₂O₄/GO hybrid showed a minimum reflection loss (RL) of -57 dB at 15.2 GHz and had an adequate absorption bandwidth of 5.2 GHz. Meanwhile, Researchers have synthesized superhydrophobic PDMS/CeFe₂O₄/GO nanocomposites by a simple mechanical mixing method. The nanocomposite has stable

electromagnetic wave absorption, reaching a minimum RL of -52dB at an absorption bandwidth of 2.1GHz. For the mechanism of action, the authors found that dielectric and magnetic losses are the most influential factors in the absorption capacity of microwaves.²² (Figure 3)

2.4 Graphene-based polymer composites

Based on what has been said above, magnetic metal nanoparticles and ferrite combined with graphene have been used as microwave-absorbing materials due to their magnetic loss qualities. However, they also have some disadvantages, such as high weight and poor corrosion resistance. In contrast, polymer composites offer more advantages, including low density, high flexibility, high corrosion resistance, and low cost. Therefore, in recent years, many conductive polymer matrix composites materials have been reported and attracted attention.

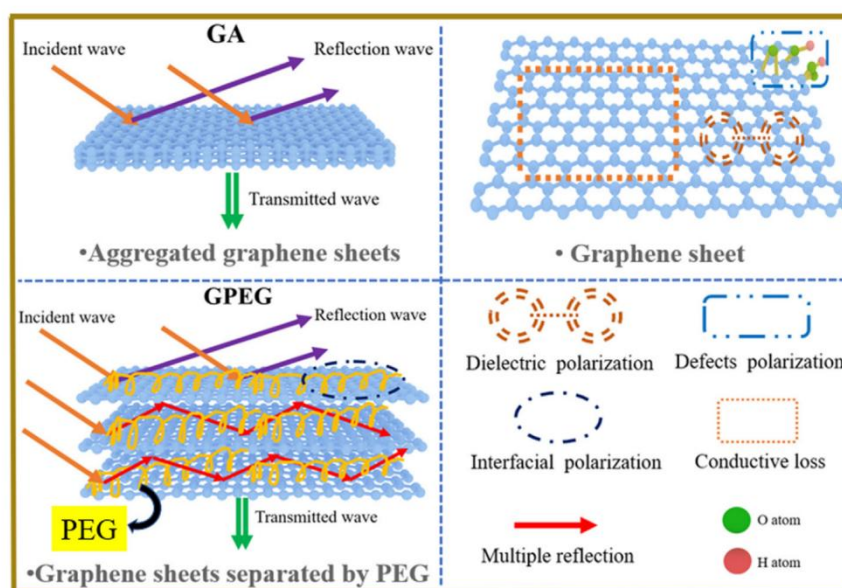


Figure 4: Schematic illustration for the microwave absorption mechanism of GPEG24

Dai et al. have prepared Fe₃O₄ reduced graphene oxide (FRGO)/waterborne polyurethane (WPU) composites with controlled structure and high microwave absorption. The WPU with 2% FRGO achieves a minimum reflection loss value of 67.8 dB and the combination of two composites with different dielectric constants can achieve an effective bandwidth of more than 14 GHz. For the mechanism, the authors proposed that dielectric loss, magnetic loss, impedance matching, and interfacial polarization were the main factor for the strongest wave absorption performance.²³

In another work, Ji et al. obtained graphene/polyethylene glycol (GPEG) composite aerogels by the reductive assembly of graphene oxide (GO) in polyethylene glycol 400 (PEG400), followed by freeze-drying. When GO is added to the absorber at only 7.5wt%, a minimum reflection loss (RL) of -43.2 dB at 13.8 GHz can be achieved, with an effective absorption bandwidth (RL < -10 dB) of 5.30 GHz (9.6-14.9 GHz) and an absorber thickness of 2.35 mm. By investigating the mechanism, it was found that the excellent electromagnetic wave absorption performance of this composite comes from these key factors, including good impedance matching, strong dielectric loss, and high electrical conductivity, as well as a special 3D porous network structure as shown in Figure 4.²⁴

Moreover, to get lighter-weight composite materials for wave-absorbing, Cheng et al. have developed ultra-lightweight graphene/polyaramid composite foams for broadband electromagnetic wave absorption in the gigahertz and terahertz bands, for use at temperatures up to 300 °C. The composite foam has an ultra-low density (0.0038 g/cm³), showing a minimum reflection loss (RL) of -36.5 dB and an effectual absorption bandwidth (EAB) of 8.4 GHz between the 2-18 GHz band. At the same time, the composite has excellent terahertz (THz) absorption with an EAB covering the entire 0.2-1.6 THz range. For the mechanism, the authors proposed that the conductive loss,

polarization loss, and impedance marching were the main factor for the excellent wave absorption performance.²⁵

2.5 Graphene-based ceramic composites

For the magnetic metallic materials and conducting polymers combined with graphene composites mentioned above, their poor stability in high-temperature environments has resulted in the materials being damaged by harsh environments. Research has shown that some ceramic materials (mainly zinc oxide, aluminosilicate, alumina, silicon nitride, etc.) have high corrosion resistance, electrical insulation, and excellent thermal stability, and can still perform their electromagnetic wave absorbing properties in harsh environments.

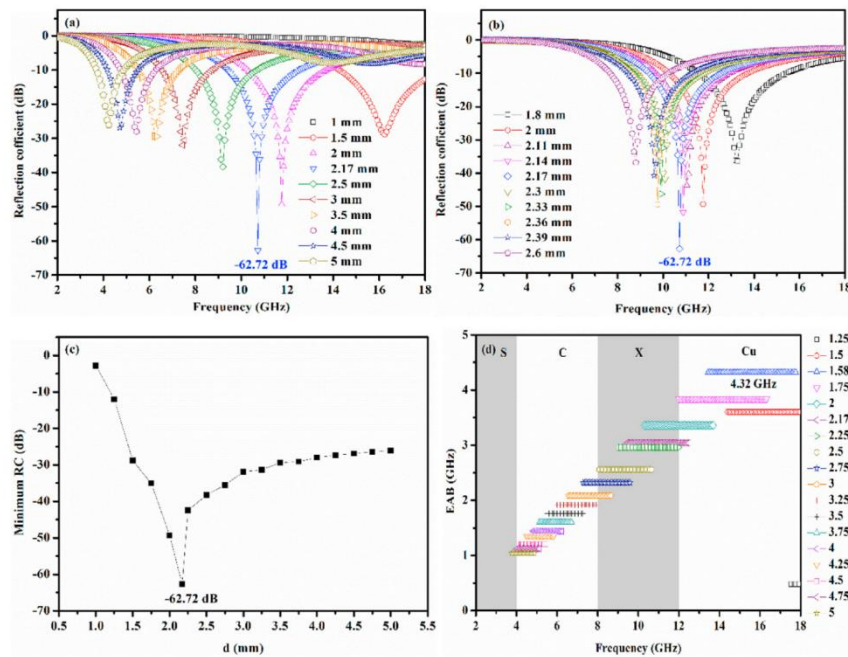


Figure 5: The electromagnetic wave absorption performance of the as-synthesized SiBCN-rGO sample: (a), (b) RC vs testing frequency; (c) minimum RC vs thickness; (d) EAB vs testing frequency²⁸

Shu et al. prepared a reduced graphene oxide/zinc oxide (RGO/ZnO) composite aerogels by means of a one-pot hydrothermal route. They found that the optimal minimum reflection loss was -79.1 dB with a thickness of 2.49 mm. Its effective absorption bandwidth reached 7.7 GHz under a thickness of 1.97 mm. This material also exhibited outstanding lightweight, flame resistance, and compression resilience properties. For the mechanism, the authors proposed that multiple reflection and scattering, dipole polarization, interfacial polarization, and conduction loss were the main factors for excellent wave absorption performance.²⁶

In addition, Zhang et al. prepared carbon-based composite foams decorated with ZnO@reduced graphene oxide (MF/ZnO@G) as microwave absorbers by in-situ crystallization combined with impregnation methods. This material exhibits excellent microwave absorption performance with a maximum reflection loss of -63.2 dB and an effective absorption bandwidth of 6.2 GHz. For the mechanism, firstly, the excellent electromagnetic wave absorption performance benefits from the three-dimensional porous structure and suitable impedance matching; in addition, polarization relaxation losses caused by functional groups, structural defects, N-doping, and heterogeneous interfaces significantly increase microwave dissipation.²⁷

Significantly, in order to improve the properties in the long term, Chen et al. prepared SiBCN-reduced graphene oxide (SiBCN-rGO) ceramic composites with different rGO contents by a polymer-derived ceramic route. The SiBCN-rGO ceramic composite with 6wt% GO has the best

microwave absorption properties, achieving a minimum reflection coefficient (RC_{min}) of 62.71 dB at 10.72 GHz with a thickness of 2.17 mm. Adjusting its thickness to 1.58 mm gives a maximum effective absorption bandwidth of 4.32 GHz (RC<10 dB) as shown in Figure 5. The authors found that the main factors affecting the microwave absorption mechanism of the prepared SiBCN-rGO ceramic composites were conductive loss and polarization loss.²⁸

Based on graphene-based binary composites (with ferrite, magnetic metal nanoparticles, polymeric materials, etc.), the combination of graphene with multi-component multifunctional composites has also attracted extensive attention.

Zhang et al. have created three-dimensional (3D) nitrogen-doped carbon nanotube (NCNT)/reduced graphene oxide heterogeneous nanostructures (3D NiFe/N-GCTs) using reduced graphene oxide as a substrate and nickel-iron as a catalyst. In general, the optimized 3D NiFe/N-GCT shows excellent minimum reflection loss (-40.3 dB) and an outstandingly efficient absorption bandwidth (4.5 GHz), exceeding most reported absorbers. Three-dimensional heterogeneous nanostructures have an interconnected network structure, a large specific surface area, and enhanced dielectric loss resulting in excellent electromagnetic wave absorption properties.²⁹

3. Conclusion

Based on the above overview of graphene-based microwave absorbing materials, it can be concluded that graphene and graphene-based composites have become more and more widely used in the field of electromagnetic wave absorption, so the study of graphene-based microwave absorbing materials has become particularly important. In practical production applications, we need to consider how to manufacture excellent-performance graphene-based microwave-absorbing materials on a large scale through cost-effective process models.

By studying the preparation methods and mechanisms of different types of graphene-based composites, it can be found that most graphene-based microwave-absorbing materials have different advantages. Graphene/carbon composites have unique properties such as low mass and high chemical stability. Graphene-based magnetic metal nanocomposites and graphene-based ferrite composites have a wide absorption band. Polymer composites offer many advantages, including low density, high flexibility, high corrosion resistance, and low cost. In addition, graphene-based ceramic composites can perform electromagnetic wave-absorbing properties in harsh environments.

Graphene has low density, large interface, high specific surface area, and high light transmission, and can be combined with other components with excellent properties to form graphene-based composites; therefore, the development of high-performance wave-absorbing materials with "thin, wide, light and strong" characteristics is of great importance for both civil and national defense. How to improve the synergy between graphene and other materials in terms of absorbing properties is now a key issue for researchers. Researchers should make use of the properties of different absorbing materials (e.g. their adaptability to different environments) to improve the performance of different graphene-based composites in a targeted manner.

Nowadays, we have already made great achievements in graphene-based composite wave-absorbing materials, and we believe that with the continuous efforts of researchers, new graphene-based composite materials with excellent wave-absorbing properties will have a broad development space and prospects in the future.

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