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Microwave absorption property of GO–Fe/FeO–NiO HNFs: GO decorated Fe/FeO–NiO hexagonal flakes with a 2D/0D/2D structure

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In this study, Fe/FeO–NiO HNFs in which 2D NiO hexagonal nanoflakes (NiO HNFs) were decorated by 0D Fe/FeO NPs were prepared by a facile hydrothermal method. Then, 0D/2D Fe/FeO–NiO HNFs were loaded on 2D GO sheets in three different weight ratios of GO (1:3), (1:4), and (1:5) to Fe/FeO–NiO HNFs and a novel GO–Fe/FeO–NiO HNF composite with a 2D/0D/2D structure was successfully produced. TEM images revealed the interesting morphology of the GO–Fe/FeO–NiO HNF composite in which individual FeO NPs with a narrow size distribution (~ 15 nm) were arranged in hexagonal NiO nanoflakes, decorated on the GO substrate. Since the morphology of nanomaterials has an important effect on their microwave absorption properties, designing a composite with an asymmetric morphology, which is the combination of zero, one, and two-dimensional nanostructures can be very efficient for adjusting the microwave absorption property. The microwave absorption ability of GO–Fe/FeO–NiO HNF composites was surveyed. All samples of Fe/FeO–NiO HNF composites exhibited superior microwave attenuation performance in terms of reflection loss with a suitable bandwidth. The minimum reflection losses for GO–Fe/FeO–NiO HNFs (1:3), (1:4), and (1:5) reached -75.22 , -53 , and -18 dB, respectively, and the effective absorption bandwidths ($RL \leq -10$ dB) for GO–Fe/FeO–NiO HNFs (1:3), (1:4), and (1:5) were 2, 3 and 3.2 GHz, respectively.

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1 Introduction

A microwave absorber is a matter that absorbs microwaves and electromagnetic energy and has low reflection and scattering.¹ Absorbent matters transmute input microwaves to heat or attenuate them through scattering, diffraction and interference.² Information safety and electromagnetic pollution are very significant in the radar system. In addition to being important materials in military and radar systems, high-performance electromagnetic absorbers can protect the environment and people from pollution. Protecting the environment and surrounding objects from GHz microwave radiation emitted by cell phones, LANs, radar systems, *etc.* is critical. We need a material that has low weight and thickness, high bandwidth, and excellent absorption.¹

Microwave energy absorbers remove undesirable electromagnetic reflections at discrete frequencies due to electrical or magnetic loss. Poor absorption performance of electromagnetic waves, narrow frequency bandwidth and low impedance, prevent their practical applications. Thus, the production of excellent-efficiency, lightweight EM wave absorbers is very important and should be on the agenda.³

Metal oxides MO ($M = \text{Ni}, \text{Cu}, \text{Zn}, \text{etc.}$) have been receiving much attention because of their high specific surface area, large number of surface reactive sites, and rich material transport channels. Nickel oxide (NiO) is used as an important semiconductor application material in lithium-ion batteries, electrocatalysis, thermal catalysis, gas sensing, electrochemistry, optical sensing, fuel cell electrodes, and biosensors.⁴

Nickel oxide (NiO) is a p-type semiconductor with a band gap of 3.6 eV. Nano-scale nickel oxide has attracted a lot of attention due to its excellent physical and chemical properties, and new nanostructures have been made of nickel oxide, *e.g.*, nanoparticles, nanosheets, and nanorods.⁵

Some kinds of morphologies of NiO showed excellent microwave absorption. For example, the microwave absorption property of flower-like nickel oxide@polyaniline (NiO@PANI) showed a maximum RL of -32.8 dB at 10.1 GHz and the effective absorption bandwidths covered 4.64 GHz (11.12–15.76 GHz) at 2.0 mm.⁶

1D flower-like yolk-shell $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{NiO}$ nano-chains were prepared by a simple hydrothermal method. The sample loaded with 30 wt% showed a minimum reflection loss value of -54.28 dB and an effective absorption bandwidth of 4.0 GHz at a thickness of 2.54 mm.⁷

Graphene oxide was extensively used due to its high thermal conductivity, good corrosion resistance, elasticity, and low

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density to make microwave absorber composites.^{8,9} Although bare GO has limited dissipation mechanisms and impedance mismatch due to poor electrical conductivity compared to rGO, and hence weak conduction loss,^{10–13} having a higher chemically reactive surface than rGO makes it a good option for use as the substrate for loading and stabilizing nanoparticles to improve their microwave absorption property as well as other physical properties.¹⁴ According to recent researches, it was found that loading various kinds of iron oxide nanoparticles onto GO or rGO sheets could result in creating a new composite with proper absorption properties.

The minimum reflection loss of the γ -Fe₂O₃ nanotube/porous rGO composite reached -34.20 dB with a thickness of 2.0 mm and the absorption bandwidth was 4.59 GHz.¹⁵ The minimum reflection loss of the Fe₃O₄@rGO composites was -49.4 dB at 16.2 GHz with an effective bandwidth of 4 GHz at a thickness of 1.75 mm.¹⁶

The flaky morphology can restrain the eddy current effect when the thickness is smaller than the skin depth, as well as increase the interface of GO and Fe/FeO–NiO HNFs NPs and leads to an increase in interface polarization, which would result in less of the microwave being reflected. In this work, the FeO NPs which were arranged on the hexagonal NiO nanoflakes were decorated on the surface of GO sheets, and a GO–Fe/FeO–NiO HNF composite was designed.

Here, the GO–Fe/FeO–NiO HNF composite with a unique morphology was synthesized by a two-step method, in which flaky Fe/FeO–NiO HNFs were prepared and then decorated on the surface of GO sheets in weight ratios of 3, 4, and 5 times GO. Finally, GO–Fe/FeO–NiO HNFs (1:3), (1:4), and (1:5) composites with unique 2D/0D/2D structures were successfully produced. The GO–Fe/FeO–NiO HNF composites were characterized using an X-ray diffraction pattern. The morphology of

Fe/FeO–NiO HNFs and GO–Fe/FeO–NiO HNF composites were studied by SEM and TEM.

The SEM and TEM images illustrated that in GO–Fe/FeO–NiO HNF composites, the individual Fe/FeO NPs were uniformly placed on the hexagonal planes and then, Fe/FeO–NiO HNFs were decorated on the GO sheets to produce GO–Fe/FeO–NiO HNF composites with a flaky morphology. VSM analysis showed that the prepared GO–Fe/FeO–NiO HNF composites have ferromagnetic behavior with weak saturation magnetization at room temperature and suitable EM attenuation ability. The minimum reflection loss for GO–Fe/FeO–NiO HNFs (1:3), (1:4), and (1:5) flake composites reached -75.22 , -53 , and -18 dB at 4.4 mm, 4.6 mm, and 5 mm thicknesses with an effective absorption bandwidth of 0.75 GHz, 1.5 GHz and 3 GHz, respectively.¹⁷

2 Experimental

In this study, a flake composite was prepared using a controllable method including a two-step process. The flake composite constituents were prepared under controlled conditions and then combined in different weight ratios. At the end, the Fe/FeO–NiO HNFs composite was formed. Firstly, Fe/FeO–NiO HNFs were synthesized using the optimal method to attain an arbitrary morphology and narrow size distribution. Then, GO was used as a substrate to decorate the Fe/FeO–NiO HNFs.

2.1 Materials

Raw materials including NiCl₂·6H₂O, Fe(NO₃)₃·6H₂O, propylene glycol (purity > 98%), and NaOH ($\geq 97\%$) were acquired from MERCK, and graphene oxide with a minimum purity of 95% was purchased from Shenzhen Nanotech Port Co. Ltd.

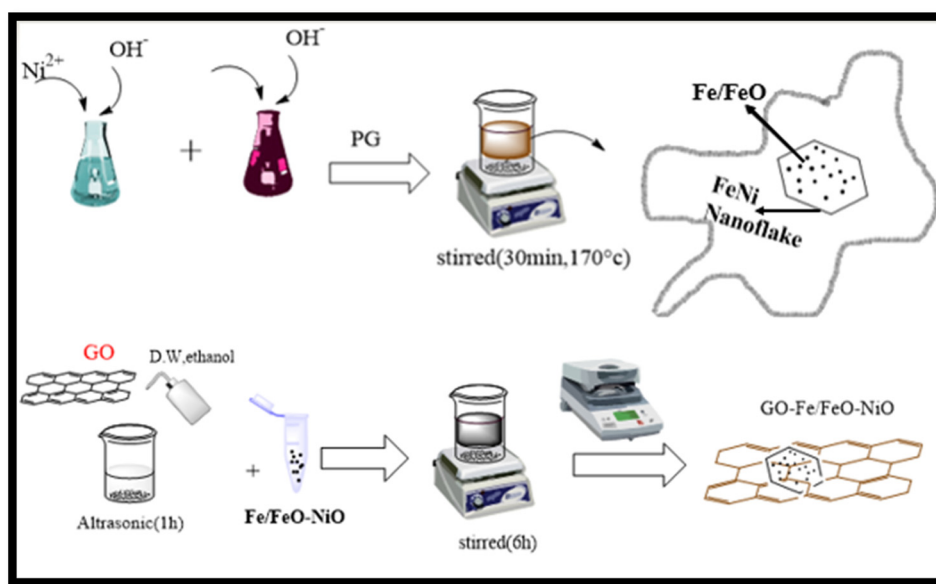


Fig. 1 Illustration of the synthesis process of Fe/FeO–NiO HNFs and GO–Fe/FeO–NiO HNF composites.

2.2 Synthesis of Fe/FeO–NiO HNFs

Fe/FeO–NiO HNFs were synthesized by the solvothermal method. Propylene glycol (PG) was used as a solvent and reducing agent as well as a protective agent to prevent aggregation during particle growth. Firstly, 40 mL of PG was heated under a temperature of 170 °C. Then, under strong magnetic stirring, a mixture of 1.4 g $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ + 0.7 gr NaOH pellets and a mixture of 0.120 g $\text{FeSO}_4 \cdot 4\text{H}_2\text{O}$ (Merck) + 0.7 gr NaOH pellets were separately prepared and added to the preheated propylene glycol (Merck, 99.0%), under magnetic stirring for 30 min. Then, the final product was collected after washing with methanol and drying at room temperature.

2.3 Synthesis of GO–Fe/FeO–NiO HNF composites

A scheme of the synthesis process of GO–Fe/FeO–NiO HNF composites is represented in Fig. 1. Firstly, GO was dispersed in 3/1 ethanol/water and then combined with Fe/FeO–NiO HNFs in the weight ratios of (3 : 1), (4 : 1), and (5 : 1), by ultrasonication and magnetic stirring for 6 h. Finally, GO–Fe/FeO–NiO HNF composites were collected by using a centrifuge and dried for 5 h at 60 °C.

3 Characterization

The successful synthesis of the GO–Fe/FeO–NiO HNF composite, chemical bonding, and crystalline structure of Fe/FeO–NiO HNFs were confirmed using FT-IR (Nicolet IS10), and X-ray diffraction (XRD, PANalytical), respectively. The magnetic behavior of products was studied in the presence of a magnetic field at room temperature using a vibrating sample magnetometer (VSM JDM-13). The TEM and FESEM techniques were applied for analysis of the morphology of Fe/FeO–NiO HNFs and GO–Fe/FeO–NiO HNF composites using an EM10C-100 KV. The flake composites' electromagnetic attributes and microwave absorption ability were specified using the transmission/reflection waveguide procedure on an Agilent vector network analyzer E8364B over 8–18 GHz.

3.1 FTIR

FTIR spectroscopy can be used to characterize the chemical composition of materials. FTIR spectra of the GO–Fe/FeO–NiO HNFs (1 : 3) flake composite are shown in Fig. 2. It can be seen from Fig. 2 that the peaks at 1107.4 cm^{-1} and 3424.96 cm^{-1} were assigned to the stretching vibrations of the O–H and C–O groups of polyglycol. The absorption band at 3400 cm^{-1} in all samples corresponds to the stretching vibrations of the covalent O–H band of water molecules adsorbed on the surface and the absorption peak at 500 cm^{-1} belongs to the stretching vibration mode of Fe–O.²⁰

3.2 XRD analysis of GO–Fe/FeO–NiO HNFs

The XRD pattern of the GO–Fe/FeO–NiO HNFs (1 : 3) composite (Fig. 3) showed that the obvious diffraction peak of GO at $2\theta = 10.52^\circ$ can conform to the (001) plane with an interlayer spacing of 0.84 nm,²¹ and the specified peaks at the angles of

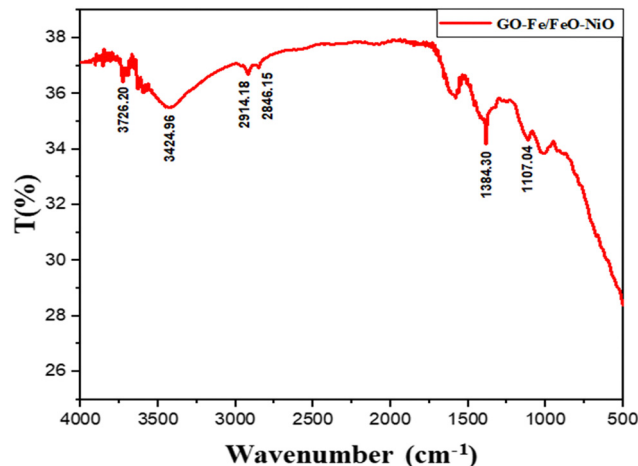


Fig. 2 FTIR spectra of the GO–Fe/FeO–NiO HNFs (1 : 3) composite.

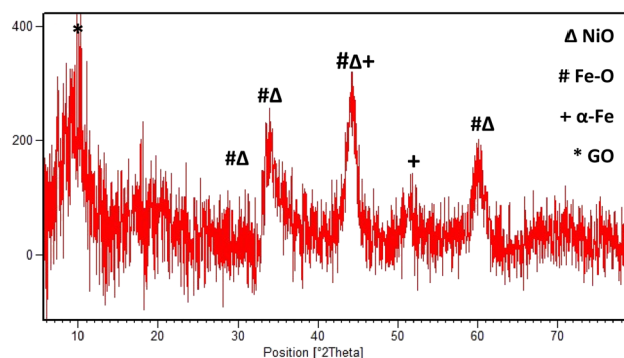


Fig. 3 X-Ray diffraction patterns of the synthesized GO–Fe/FeO–NiO HNFs (1 : 3) composite.

34, 44, and 60.5° correspond to the (111), (200) and (220) of FeO and NiO planes, respectively. Diffraction peaks at $2\theta = 34, 44,$ and 61° correspond to (111), (200), and (220) planes of NiO nanosheets, respectively. The diffraction peaks of NiO and FeO are the same.²⁰ The presence of FeO in the composite is also proved by FTIR spectra and EDS. Since the tendency of iron to oxidize is more than that of nickel, besides NiO, FeO must be present in the composite. Furthermore, the diffraction peak at $2\theta = 52^\circ$ shows the residual of α -Fe in the composite,²¹ but FeO is the predominant compound.

The average size of the crystals was calculated to be around 17.8 nm using the Debye–Scherrer equation.

$$D = 0.9\lambda/(\beta \cos \theta) \quad (1)$$

D represents the average size of nanocrystals, λ is the wavelength of the X-ray, B is the width at half the maximum value of the height, and θ is the Bragg angle.¹²

3.3 FESEM images

FESEM images of Fe/FeO–NiO HNFs are shown in Fig. 4a and c. The spherical agglomerated nanoparticles showed an irregular mass shape with a rough surface. The SEM image of the GO–Fe/

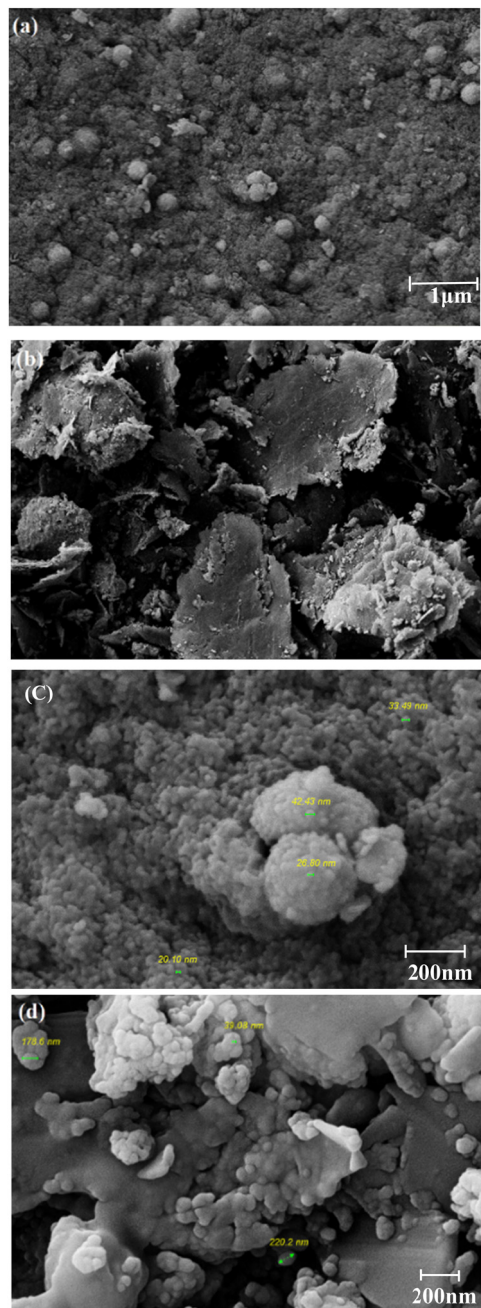


Fig. 4 FESEM images of composite materials: Fe/FeO-NiO HNFs (a and c) and GO-Fe/FeO-NiO HNFs (1:3) composite (b and d), respectively.

FeO-NiO HNF composite shows the GO sheets decorated with dispersed nanoparticles (Fig. 4b and c). The SEM image didn't give more information about the morphology of Fe/FeO-NiO HNFs because the size of NiO nanosheets is very small compared to graphene sheets and is out of the scale range of SEM.

3.4 EDS

In this analysis, all the elements such as oxygen, nickel, and iron are seen, and also the weight percentage of nickel is more than that of iron; the presence of the seen elements indicates the successful synthesis of Fe/FeO-NiO HNFs Fig. 5.

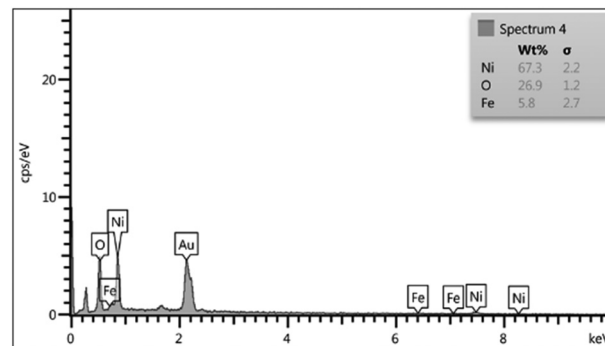


Fig. 5 EDS analysis of the GO-Fe/FeO-NiO HNFs (1:3) flake composite.

3.5 TEM images

The TEM images which represent more detail about the unique morphology of Fe/FeO-NiO HNFs and GO-Fe/FeO-NiO HNFs (1:3) composite are shown in Fig. 6. This figure obviously shows that individual Fe/FeO NPs with a very small size of less than 15 nm are uniformly distributed on the NiO hexagonal flakes. Fe/FeO NPs are also seen in clusters with an average size of 75 nm, at the edges of the hexagons. In the image of GO-Fe/FeO-NiO HNF composites (Fig. 6b), the random spreading of hexagonal flakes at the surface of GO sheets can be clearly observable. Moreover, graphene sheets can be seen as flat and wide sheets (with small folds) without creases and fractures at the edges, which shows the high quality of graphene. Decoration of GO with higher amounts of hexagonal flakes containing Fe/FeO NPs is shown in the TEM image of the GO-Fe/FeO-NiO HNFs (1:5) composite with a large scale of 1 μm (Fig. 6d). Hexagonal flakes are also obviously observable in this image. As expected, the agglomeration of hexagonal flakes increases with an increment in the amount of Fe/FeO-NiO HNFs. As shown in Fig. 6b-d the number and size of hexagonal flake clusters were increased with increasing the weight ratio of Fe/FeO-NiO HNFs.

3.6 Vibrating sample magnetometer (VSM) analysis

The magnetic properties of the Fe/FeO-NiO HNFs were represented by magnetic hysteresis curves (Fig. 7), which were measured in an applied magnetic field from -15 kOe to 15 kOe. This illustrates that more than one magnetic domain existed in a single crystal particle, exhibiting typical ferromagnetic behavior. The Fe/FeO-NiO HNFs exhibits a very weak saturation magnetization (M_s) of 4.25 emu g⁻¹ and coercivity (H_c) of 13.9 Oe. By adding Fe/FeO-NiO HNFs to GO sheets, the GO-Fe/FeO-NiO HNFs (1:3) composite has a saturation magnetization (M_s) of 0.9 emu g⁻¹ and a coercivity (H_c) of 13.8 Oe. Fig. 7 also compares the hysteresis loops of the Fe/FeO-NiO HNFs and GO-Fe/FeO-NiO HNFs (1:3) composite. This result proves that the Fe/FeO-NiO HNFs has a very weak magnetization property.²²

3.7 Electromagnetic parameters

The electromagnetic (EM) parameters of the GO-Fe/FeO-NiO HNF composite were computed to assess their absorption

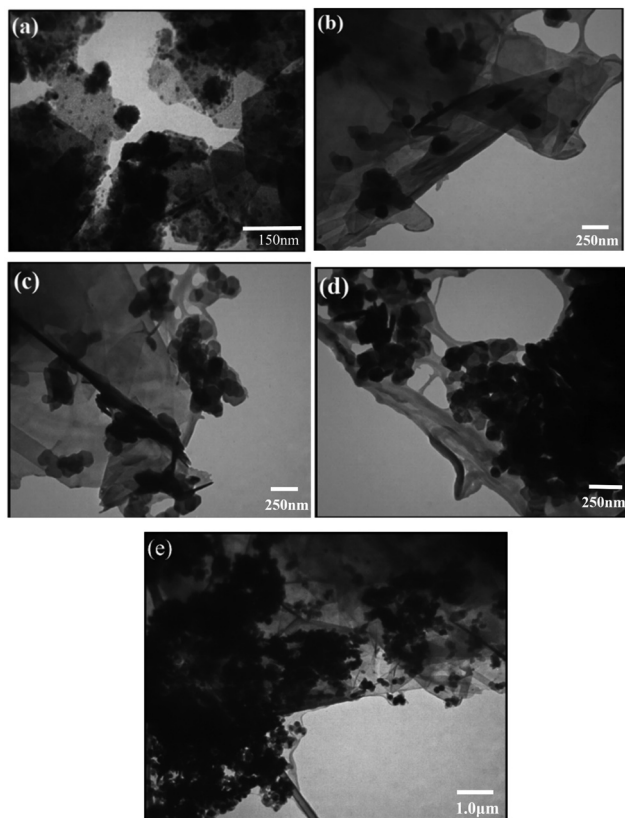


Fig. 6 TEM images of Fe/FeO-NiO HNFs (a), GO-Fe/FeO-NiO HNFs (1:3) (b), GO-Fe/FeO-NiO HNFs (1:4) (c), and GO-Fe/FeO-NiO HNFs (1:5) (d and e).

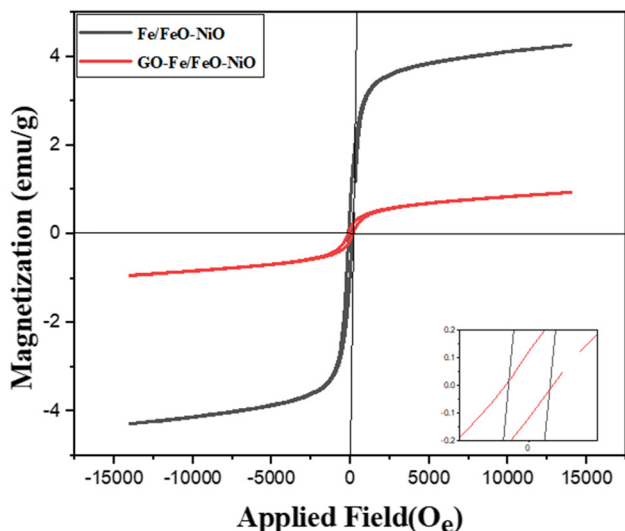


Fig. 7 Room temperature magnetization hysteresis loops: Fe/FeO-NiO HNFs and GO-Fe/FeO-NiO HNFs (1:3) composite.

performance. For this purpose, composites were homogeneously dispersed in a resin epoxy with a 25 wt% filler content. The real and imaginary parts of electrical permittivity (ϵ' and ϵ'')

and the real and imaginary parts of magnetic permeability (μ' and μ'') of GO-Fe/FeO-NiO flake composites were attained using a network vector analyzer (Anritsu.37269D) in the 8–18 GHz. Since the GO-Fe/FeO-NiO HNF composite showed very weak magnetization, nonmagnetic mode was used to calculate EM parameters.

The reflection loss values of the GO-Fe/FeO-NiO HNF composite with various thicknesses were calculated based on the transmission line theory, according to the following relations:

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}} \tan h \left(j \frac{2\pi d f}{c} \sqrt{\epsilon_r \mu_r} \right) \quad (2)$$

$$RL(dB) = 20 \log \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (3)$$

where μ_r and ϵ_r are the relative complex permeability and permittivity, respectively, Z_{in} is the input impedance of the microwave absorbing material, and Z_0 is the characteristic impedance of free space. The parameters f , c , j and d are the microwave frequency, the speed of the electromagnetic wave in the vacuum, the proportionality constant and the thickness of the absorbent material layer, respectively. When Z_{in} (the input impedance of the absorbing material) is equal to Z_0 (the impedance of the free space), the absorption by the absorbent material shows the maximum value. In this case, all the energy of electromagnetic waves is absorbed by the absorbent material and converted into heat by magnetic and dielectric losses.^{18–23}

Complex permittivity ($\epsilon_r = \epsilon' - i\epsilon''$) and complex permeability ($\mu_r = \mu' - i\mu''$) are EM parameters, which are an intrinsic property of each absorbent material, and determine the ability of a matter to absorb waves. ϵ' and μ' , the real part of transmittance and permeability, measure the capability to store electric and magnetic energy, which represents the degree of the polarizability of absorbent materials. The electric and magnetic energy of the incident wave inside the material is represented by ϵ'' and μ'' , respectively, which are also identified as dielectric and magnetic losses.^{24,25}

The imaginary parts of permittivity and permeability (ϵ'' , μ''), represent the polarization relaxation and spin rotation relaxation (leading to the alignment of dielectric and magnetic dipoles with electric and magnetic fields of incident waves), respectively. ϵ'' is due to several polarization including the electron, nucleus, dipole, and interfacial polarization relaxation (space accumulated charge polarization). The main factor in the dielectric loss is the space-charge polarization, which occurs because of additional polarization in the higher frequency range.²⁶

At the interface of materials with different dielectric constants, surface polarization happens as an effect of space-charge accumulation. ϵ'' in conductive materials generally originates from conductivity loss and polarization relaxation.²⁷

Fig. 8a shows that the ϵ' values of GO-Fe/FeO-NiO HNFs (1:3), (1:4), and (1:5) flake composites varied from 2.4 to 0.32, 5.14 to 0, and 10.54 to 3.23, respectively; by increasing the frequency, ϵ' has decreased. For the entire frequency range, ϵ' of GO-Fe/FeO-NiO HNFs (1:3) is the smallest compared to that of the two other samples. Totally, this figure shows that by

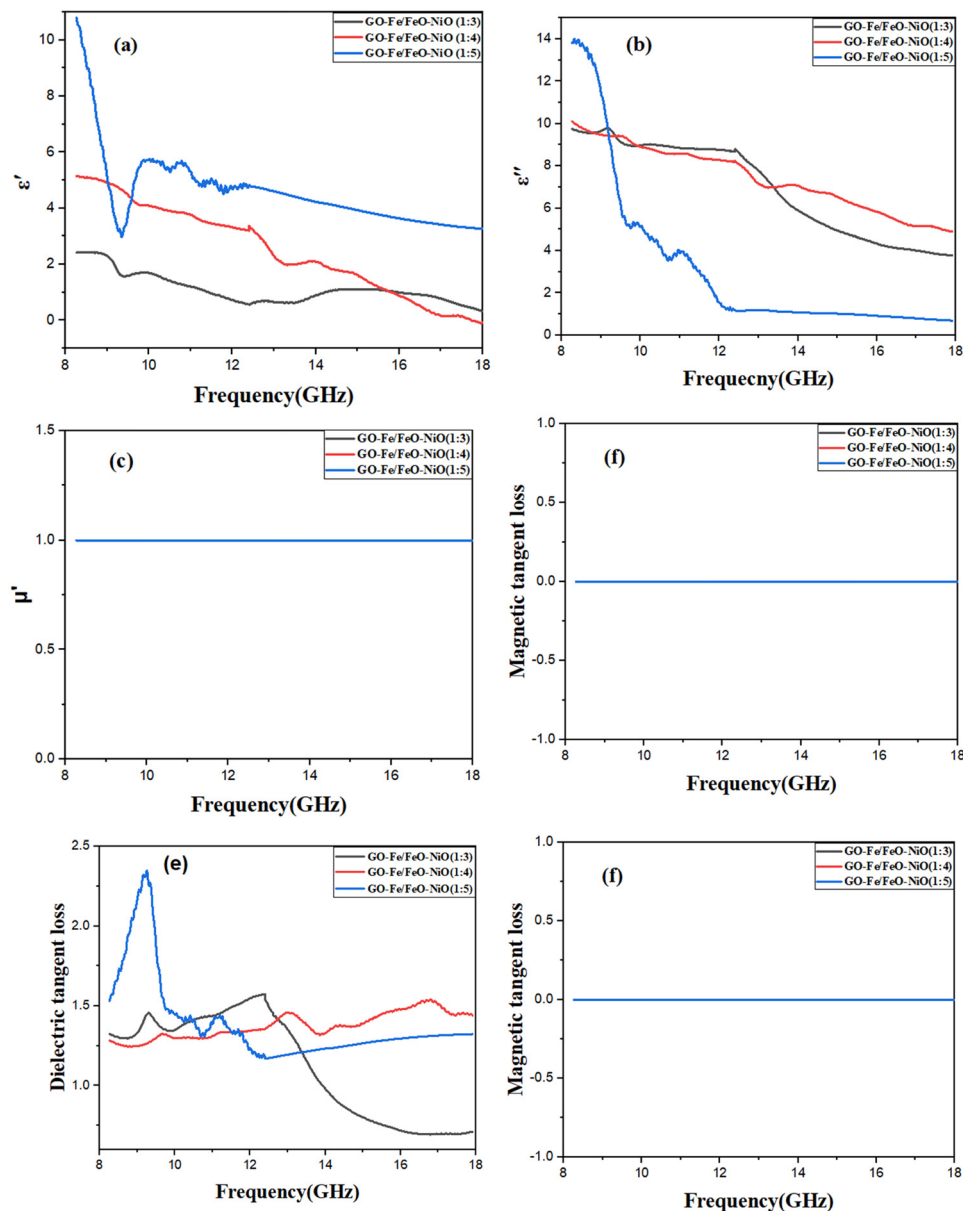


Fig. 8 Electromagnetic parameters, real and imaginary parts of permittivity (a and b) and permeability (c and d), and dielectric ($\tan \sigma_e$) and magnetic ($\tan \sigma_\mu$) tangent losses (e and f) of GO-Fe/FeO-NiO HNF composites.

increasing the weight ratio of Fe/FeO-NiO HNFs, the ϵ' values are increased, originating from the large number of dipoles created in Fe/FeO-NiO HNFs, and increasing the area of interfaces between Fe/FeO-NiO HNFs and GO sheets leads to the higher amount of charge accumulation at these interfaces, which enhances the polarized dipoles. Hence, the ϵ' values rise with the increase in Fe/FeO-NiO HNF weight ratio.

The ϵ'' values of GO-Fe/FeO-NiO HNFs (1:3), (1:4), and (1:5) composites are shown in Fig. 8b. The ϵ'' values of GO-Fe/FeO-NiO HNFs (1:3) and (1:4) are almost the same (10–6) but the further increment in the Fe/FeO-NiO HNFs ratio in the sample (1:5) leads to a sudden drop in its ϵ'' value to 1. This result referred to an increment in the agglomeration and clustering of Fe/FeO-NiO HNFs by increasing the weight ratio of Fe/FeO-NiO HNFs from 3 to 5, due

to strong electrostatic interaction among these flakes, as shown in Fig. 6b–d. The clustering and accumulation of Fe/FeO-NiO HNFs decrease the interface between Fe/FeO-NiO flakes and GO sheets, which leads to an increment in interfacial polarization. So, the amounts of Fe/FeO-NiO HNFs that were loaded on the GO substrates must be optimized. Fig. 8b also shows that the ϵ'' values decrease slowly with increasing frequency due to the effect of frequency dispersion.²⁸ As shown in Fig. 8c and d, μ' and μ'' have a constant value of 1 and 0, respectively, because non-magnetic mode was used to calculate EM parameters.

The minimum reflection loss ($RL_{\min}$) of GO-Fe/FeO-NiO HNFs (1:3), (1:4), and (1:5) composites *versus* frequency is plotted in Fig. 9a–c (2D) and Fig. 9a'–c' (3D). Fig. 9 shows that GO-Fe/FeO-NiO HNFs (1:3) presents an $RL_{\min}$ of -75.22 dB at

9.2 GHz with a matching thickness of 4.4 mm with a narrow absorption bandwidth of 0.75 GHz (8.6–9.4 GHz) for 90% absorption ($RL \leq -10$ dB). The $RL_{\min}$ for GO-Fe/FeO-NiO HNFs (1:4) is -53 dB with a bandwidth of 3 GHz (8.2–11.2 GHz) at 4.4 mm thickness. The $RL_{\min}$ for GO-Fe/FeO-NiO (1:5) is -18 dB with 3.2 GHz absorption bandwidth (14.8–18 GHz). Remarkably, the $RL_{\min}$ corresponding to the maximum microwave absorption could be located at various frequencies by modulating the matching thicknesses of absorbers.²⁹

The reflection loss of composites obviously depends on the contents of Fe/FeO-NiO HNFs in the composite; in a way, the reflection loss decreased with increasing the weight ratio of Fe/FeO-NiO HNFs to 5. But the absorption bandwidth is

increased. The results represented in Fig. 8 and 9 are interpreted in the following section (absorption mechanisms).

3.8 Absorption mechanisms

The modifying absorption properties of the composites can be explained by the following causes: The synergistic effects between GO and Fe/FeO-NiO HNFs were because of Debye and interfacial (Maxwell-Wagner) relaxations. The GO sheets and the empty spaces existing between Fe/FeO-NiO in hexagonal flakes lead to a relatively large specific surface area, thereby providing more active sites for electromagnetic wave reflection and scattering, and can effectively cut off electromagnetic wave propagation and produce loss.^{30–35}

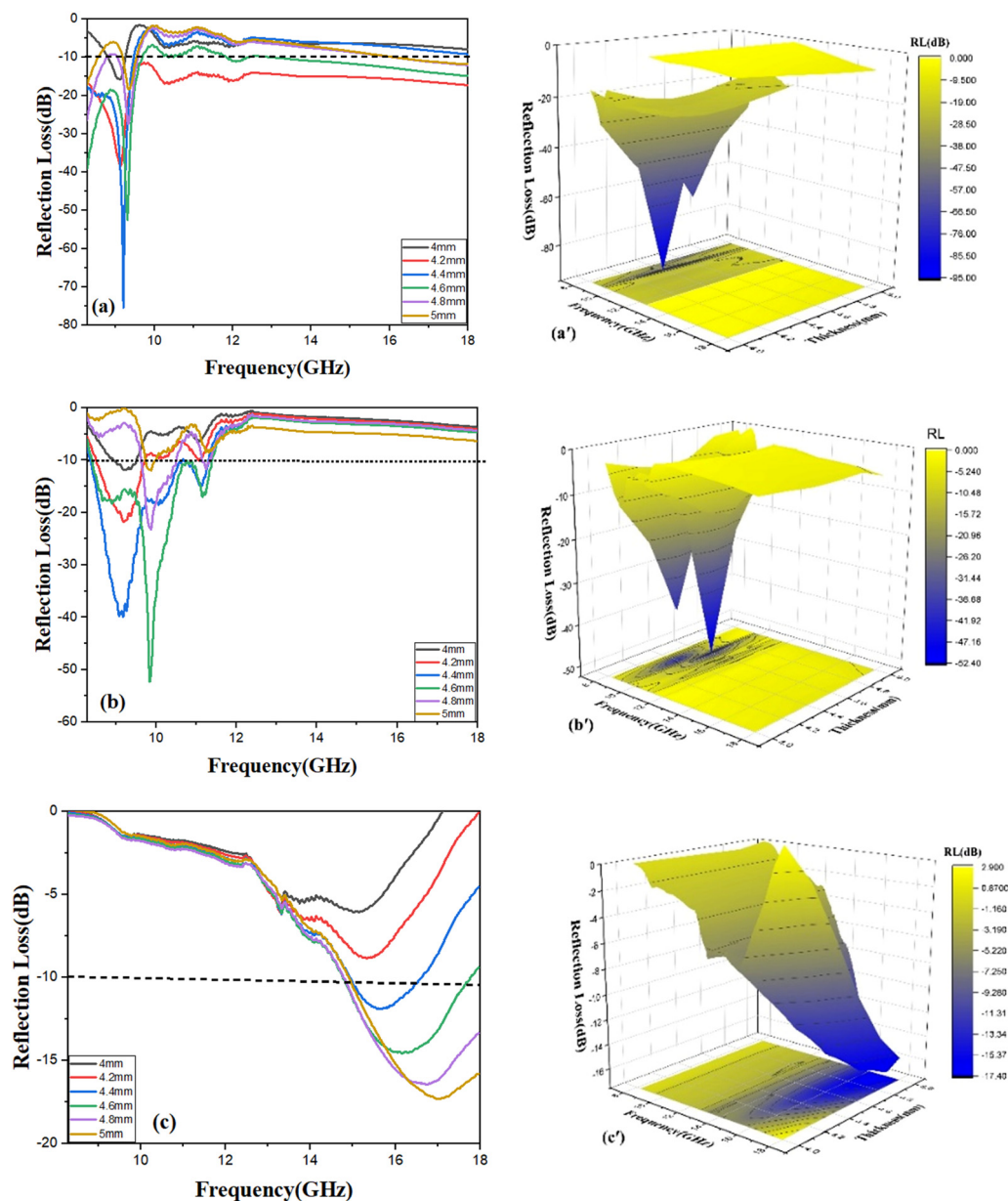


Fig. 9 2D (a–c) and 3D (a'–c') reflection loss dependence on frequency for flake composites: GO-Fe/FeO-NiO HNFs (1:3), (1:4) and (1:5) as (a–c), respectively with six different thicknesses of absorbers.

Possible EM wave absorption mechanisms of the GO-Fe/FeO-NiO HNF composite are shown in Fig. 10. Brilliant EM wave absorption capacity of the GO-Fe/FeO-NiO HNF composite results from the joint action of several absorption mechanisms. The layered structure of GO sheets and Fe/FeO-NiO HNFs emanates enhanced scattering and multiple reflections between these planes. In the unique morphology of the GO-Fe/FeO-NiO HNF composite, the electromagnetic wave propagation path can be extended to reduce the electromagnetic wave.^{35,36}

Furthermore, specifications of high charge mobility and electrical conductance of graphene originate from the sp^2 hybridization of carbon atoms, and the higher electrical conductance and specific surface lead to an easier combination of conductance networks. Free electrons, which follow the theory of free electrons, when they pass through a conductive channel under the influence of an electromagnetic field, lead to a current and supply the absorber with high conduction loss.^{37,38} In the end, the accumulated defects and functional groups were formed from dipoles that existed at Fe/FeO-NiO HNFs, GO and at their interface, disconnected and reoriented after attracting energy under the EM field. The dipole changes cannot keep up with the changes and increase in the frequency of the electric field, which leads to the phenomenon of hysteresis, resulting in polarization and relaxation.³⁹⁻⁴¹

The Debye relaxation process can be represented with the Cole-Cole semicircles in Fig. 11 (ϵ'' against ϵ'), which are calculated by the following equation:⁴²

$$\left(\epsilon' - \frac{\epsilon_s + \epsilon_\infty}{2}\right)^2 + (\epsilon'')^2 = \left(\frac{\epsilon_s + \epsilon_\infty}{2}\right)^2 \quad (4)$$

where ϵ_s represents the static dielectric constant and ϵ_∞ is the dielectric constant at infinite frequency. One of the important mechanisms in the absorption of electromagnetic waves, the Debye dipole relaxation mechanism is for dielectric dissipation materials. Each semicircle in the $\epsilon' - \epsilon''$ curve shows the Debye relaxation process. The conductive paths are developed *via* the graphene sheets, simplifying electronic transportation leads to an increasing permittivity. Besides, the polarization at the interfaces and defects can contribute to ϵ'' .⁴³ The Cole-Cole curves simultaneously show long tails and semicircles that are somewhat distorted. This indicates that the conductive loss also dominates in the dielectric loss. The $\epsilon' - \epsilon''$ curves for the samples GO-Fe/FeO-NiO HNFs (1:3), and GO-Fe/FeO-NiO HNFs (1:5) composites exhibit at least two semicircles, which indicate the existence of multiple Debye dipolar relaxation processes. One semicircle is observed for the GO-Fe/FeO-NiO HNFs (1:4) composite.

Furthermore, for all samples it can be seen that semicircles are distorted to some degree; this proposes that the Debye relaxation is not the only mechanism for the dielectric loss but other mechanisms including conduction loss and interfacial polarization could be responsible for microwave absorption.⁴⁴

According to the transmission line theory, the attenuation constant α estimated through the equation reflects the absorption ability of the EM wave by the absorbers.⁴⁵

$$\alpha = \frac{\sqrt{2}\pi f}{c} \sqrt{(\mu''\epsilon'' - \mu'\epsilon') + \sqrt{(\mu''\epsilon'' - \mu'\epsilon')^2 + (\epsilon'\mu'' + \epsilon''\mu')^2}} \quad (5)$$

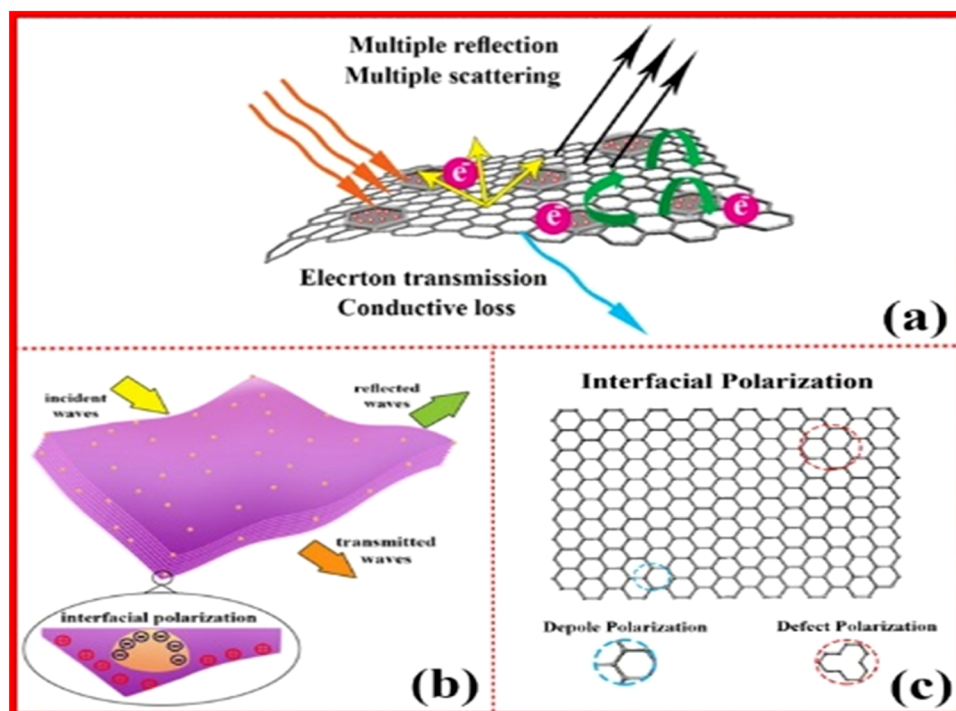


Fig. 10 Schematic representation of microwave absorption mechanisms for GO-Fe/FeO-NiO HNF composites.

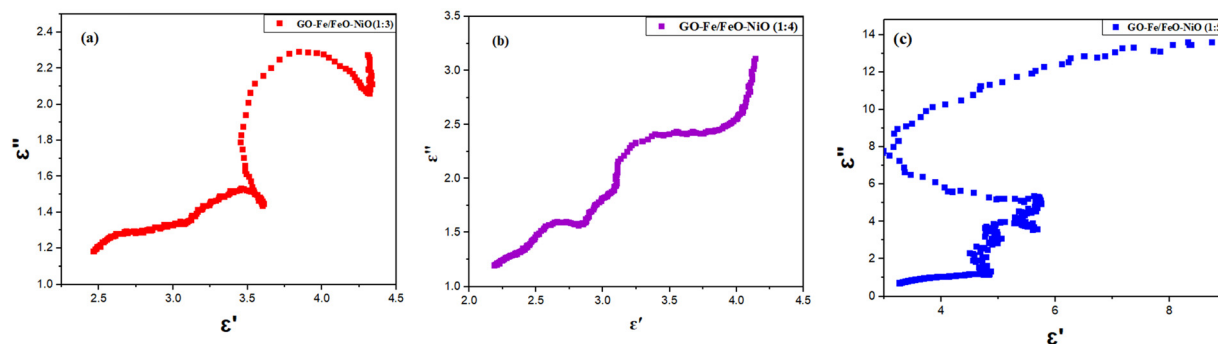


Fig. 11 Typical Cole–Cole semicircle of GO–Fe/FeO–NiO HNF composites.

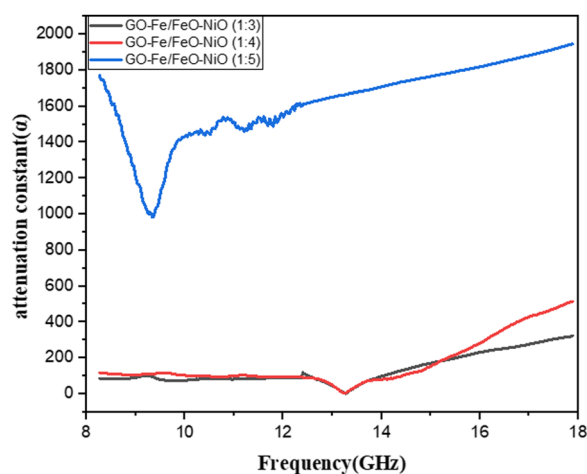


Fig. 12 Attenuation constant (α) for the samples GO–Fe/FeO–NiO HNF composites.

The attenuation constant α (Fig. 12) oscillates by frequency enhancement, up to attaining the maximum value at high frequency. All products exhibit similar attenuation capability; noteworthy, GO–Fe/FeO–NiO (1:4) did not show maximum attenuation ability; must be changed into: GO–Fe/FeO–NiO (1:5) shows maximum attenuation ability, this demonstrates that the illustrious EM wave absorption behavior due impedance adaptation characteristics is considerable.

4 Conclusions

In summary, GO–Fe/FeO–NiO HNF composites with an interesting morphology were synthesized using a two-step approach. First, Fe/FeO–NiO HNFs were prepared with a unique morphology, in which Fe/FeO NPs with a narrow size distribution were uniformly deposited on hexagonal NiO flakes and then Fe/FeO–NiO hexagonal nanoflakes were loaded on the GO sheets in three different weight ratios of 3, 4 and 5 to GO to achieve the optimum weight ratio of Fe/FeO–NiO HNFs to GO to attain the best absorption capability of the GO–Fe/FeO–NiO HNF composite. The TEM results confirmed the exciting 2D/0D/2D morphology of the GO–Fe/FeO–NiO HNF composite. The microwave absorption property of the GO–Fe/FeO–NiO HNF composites,

which was investigated *via* calculation of their EM parameters, shows excellent reflection loss ability of GO–Fe/FeO–NiO HNF composites (1:3), (1:4) and (1:5) (−75, −50, and −18 dB) with the absorption band of 0.75, 3 and 3.2 GHz in 4.4 mm sample thickness.

Conflicts of interest

There are no conflicts to declare.

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