Magnetic Carbon Nanotubes/Graphene Oxide Nanocomposites: One-Step Controllable Production, and Their Excellent Microwave Absorption Capabilities

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In this research, three-dimensional (3D) hierarchical networks of magnetic carbon nanotubes/graphene oxide (CNTs/GO) nanocomposites (NCs) were synthesized by a one-step hydrothermal method with the idea of constructing more interface contacts, a double loss mechanism and synergy effect. The obtained results showed that the content of NCs gradually transitioned from CoCO$_3$ to Co$_x$Fe$_{3-x}$O$_4$. Further, CoCO$_3$-Co$_x$Fe$_{3-x}$O$_4$/CNTs/GO quaternary NCs and Co$_x$Fe$_{3-x}$O$_4$/CNTs/GO ternary NCs could be selectively synthesized when the Co:Fe molar ratio decreased from 1:1 to 1:3. The as-prepared samples displayed excellent microwave absorption properties (MAPs) in terms of the minimum reflection loss ($R_{\text{min}}$) and the corresponding absorption bandwidth (FB$_{\text{cor}}$) with reflection loss values lower than $-10$ dB. The $R_{\text{min}}$ and large FB$_{\text{cor}}$ values for the as-prepared samples were $-54.69$ dB and 2.36 GHz, $-63.99$ dB and 2.48 GHz, $-63.36$ dB and 2.68 GHz, respectively. The designed 3D hierarchical networked structure of magnetic GO/CNTs NCs displayed outstanding comprehensive MAPs, which could deliver a practical and effective strategy to develop high-performance microwave absorbers (MAs), considering their low cost, lightweight, and high stability.

Keywords: magnetic GO/CNTs, 3D hierarchical networks structure, interfacial polarizations, dielectric and magnetic loss mechanisms, microwave absorption properties

INTRODUCTION

In the past decades, microwave absorbers (MAs) have drawn significant attention owing to the expanded electromagnetic (EM) radiation and interference problems (Novoselov et al., 2004; Zhang et al., 2014; Cao et al., 2019). It is well acknowledged that MAs with characteristics such as strong absorption ability, large absorption bandwidth, lightweight, low cost, and remarkable chemical stability are highly desirable because of their promising military and civil application prospects.
(Zhang et al., 2015; Fang et al., 2016). According to the EM energy conversion principle, the incident EM wave can be attenuated by dielectric and magnetic losses (Wei et al., 2017). Therefore, nanocomposites (NCs) consisting of dielectric loss materials and magnetic nanoparticles (NPs) are considered an effective way to achieve high-performance MAs (Liu et al., 2015, 2018; Quan et al., 2017). For example, CoNi@Air@TiO$_2$ materials and magnetic nanoparticles (NPs) are considered an effective way to achieve high-performance MAs (Liu et al., 2016), Co$_{0.8}$Fe$_{2.4}$O$_4$@MoS$_2$ (Long et al., 2019), Ni@SnO$_2$ (Zhao et al., 2016), Co$_x$Fe$_{3-x}$@SiO$_2$ (Chen et al., 2017) have been demonstrated to display outstanding microwave absorption properties (MAPs). Generally, owing to their excellent synergistic effect, the development of high-performance MAs mainly focused on core@shell structure magnetic NCs, in which dielectric substances are utilized as a shell and magnetic NPs as a core. Among them, different dielectric substances such as ZnO, TiO$_2$, and BaTiO$_3$ were often used as a shell (Liu et al., 2008; Jiang et al., 2014; Xu et al., 2018). However, the biggest drawback of these NCs is the high density, which hugely restricts their practical application.

In recent years, owing to the lightweight, unique tube structure and excellent dielectric loss, carbon nanotubes (CNTs) were used as substrate to construct the CNTs-based for designing the ideal MAs (Chen et al., 2015; Wang et al., 2018). Examples include Fe/CNTs (Che et al., 2004), ZnFe$_2$O$_4$/CNTs (Khabouri et al., 2015), and Fe/Co/Ni/CNTs (Wen et al., 2011), which were demonstrated to display very outstanding MAPs. Generally, the previously designed core@shell structure CNTs-based NCs were mainly composed of CNTs as a shell and different categories of magnetic NPs as a core (Lv et al., 2008; Weissker et al., 2010). However, the magnetic properties and magnetic loss abilities for these kinds of CNTs-based NCs are hugely limited and cannot be well modulated due to the magnetic NPs acting as the core, and the low encapsulation of magnetic NPs (Pawar et al., 2019; Long et al., 2020). Therefore, constructing the reverse core@shell structure magnetic CNTs-based NCs, in which magnetic NPs serve as the shell and CNTs as the core, is an effective way to overcome these problems (Wang et al., 2012; Li et al., 2017).

Furthermore, the recent results indicated that the outstanding properties of graphene (G) and the designed G-based could provide many more interfaces, which were conducive to improve MAPs (Qu et al., 2016; Wang et al., 2019). Considering these aspects, in this article, we designed and synthesized magnetic CNTs/graphene oxide (GO) NCs by a one-step hydrothermal method. By changing the Co:Fe molar ratio, different compositions of magnetic NPs/CNTs/GO NCs could be selectively produced. The obtained results showed that the as-prepared magnetic CNTs/GO NCs displayed very excellent MAPs.

**EXPERIMENTAL**

**Synthesis of Magnetic CNTs/GO NCs**

Similarly to the previously reported experimental method shown in Figure 1 (Wu et al., 2020), 80 mg of GO was prepared by a modified Hummers method (Hummers and Offeman, 1958). The prepared GO was added in 40 mL deionized water and ultrasonicated for 30 min at room temperature (RT). Next, 40 mg of hydroxylated CNTs were ultrasonically dispersed into the GO solution for another 30 min to obtain the GO/CNTs substrate solution. Then, the solution of Co$^{2+}$ and Fe$^{3+}$ was obtained by ultrasonic dissolving C$_6$H$_5$CoO$_2$•$n$H$_2$O (1 mmol) and FeCl$_3$•6H$_2$O (1 mmol) into a 30 mL ethylene glycol solution. Subsequently, the resulting Co$^{2+}$ and Fe$^{3+}$ solution was dissolved into GO/CNTs substrate solution by stirred for ca. 60 min. Finally, urea (0.9 g) were thoroughly dispersed into the mixed solution for another 5 min. The final solution was transferred into a 100 mL Teflon-lined stainless steel autoclave and heated at 200°C for 24 h and left to cool to RT. The synthesized magnetic CNTs/GO NCs were washed several times and magnetically centrifuged. The obtained sample was left to dry. The experimental conditions were kept the same, with the exception of 2 and 3 mmol of FeCl$_3$•6H$_2$O, which were used as the Fe source in the synthesis of magnetic NPs/CNTs/GO NCs to control compositions of the sample. The samples were denoted as S1, S2, and S3 for easy description, respectively.

**Characterization and Measurement**

The phases and microstructures of the as-prepared magnetic CNTs/GO NCs were characterized by an X-ray powder diffractometer (XRD) (model Smart Lab, Rigaku), Raman spectroscopy (Jobin-Yvon Labram HR800), Thermal gravity analysis (TGA) (Netzsch Sta 449F3), transmission electron microscope (TEM) (model Tecnai-G20), and X-ray photoelectron spectroscopy (XPS) (model Escalab 250Xi, Thermo Fisher Scientific). 30 wt% of the obtained samples were mixed with wax and pressed into toroidal shaped samples (R$_{out}$: 7.0 mm, R$_{in}$: 3.0 mm), respectively, to investigate the EM properties and MAPs. The as-prepared composites were measured by a vector network analyzer (Agilent E8363B) in the frequency region of 2–18 GHz.

**RESULTS AND DISCUSSION**

Figure 2 presents the XRD patterns and Raman spectra of the as-prepared magnetic CNTs/GO NCs. Figure 2A shows that all the as-prepared samples display similar XRD patterns, pointing to the same category of sample. The characteristic peaks at 30.08°, 35.44°, 43.06°, 53.44°, 56.97° and 62.59° corresponding to (220), (311), (400), (422), (511), and (440) crystal planes are consistent with the standard XRD patterns of spinel CoFe$_2$O$_4$ (JCPDS: 22-1086). In comparison, one peak at 32.6° was observed clearly over sample S1 and S2, assigned to (104) crystal planes of CoCO$_3$ (JCPDS: 78-0209). However, this peak cannot be seen over sample S3. The comparison results reveal that the obtained samples evolve from CoCO$_3$ to Co$_x$Fe$_{3-x}$O$_y$ when the amount of Fe source increases from 1 mmol to 3 mmol. In confirmation of the results, the experimental conditions were kept the same, with the exception of 5 mmol FeCl$_3$•6H$_2$O, which was used as the Fe source to produce the sample, S4 for easy description. As shown in Figure 2A, the XRD pattern indicates that the phase of CoCO$_3$ cannot be observed over the as-prepared S4, which is consistent with our previous reported result.
FIGURE 1 | Schematic illustration for the synthesis process of the as-prepared magnetic CNTs/GO NCs.

FIGURE 2 | (A) XRD patterns, and (B) Raman spectra of the as-prepared samples.

(Wu et al., 2020). Moreover, consistent with the GO/CNT-Fe₃O₄ NCs reported elsewhere (Wang et al., 2014), the characteristic diffraction peaks for GO and CNTs cannot be seen, which should be attributed to the uniform dispersion of CoₓFe₃₋ₓO₄ NPs on the surface of CNTs/GO. The existence of carbon materials can be confirmed by Raman spectra. As shown in Figure 2B, the obtained magnetic CNTs/GO NCs show three evident peaks located at 1347 cm⁻¹, 1586 cm⁻¹, and 2694 cm⁻¹, respectively. These peaks can correspond to the D band, G band and 2D band of carbon material, respectively. It is well-known that the D band originates from the sp³ defect/disorder in the graphitic structure, the G band is indicative of a high crystallinity graphitic layer, and the 2D band is excited by a double-resonant Raman process (Ritter et al., 2006; Popov and Lambin, 2013). Based on the obtained results, we can confirm the existence of GO and CNTs in the obtained NCs.

Figure 3 shows the typical TGA curves and XPS spectra of as-prepared samples. As shown in Figure 3A, it can be seen that the obtained S2 and S4 samples present the evident weight loss under air atmosphere in the temperature range of 240–620°C. Based on the XRD results, the mass loss can mainly be ascribed to the thermal decomposition of GO and CNTs, and the residual substances should be assigned to CoₓFe₃₋ₓO₄ NPs. The TGA results indicate that the CoₓFe₃₋ₓO₄ content in the obtained S2 and S4 samples is 18.4% and 30.4%, respectively. To confirm the composition of the obtained samples, Figure 3B provides their XPS spectra. It can be deduced that all the obtained NCs have four peaks located at 284, 530, 712, and 781 eV, respectively. Further, these peaks can be assigned to C 1s, O 1s, Fe 2p, and Co 2p.

To obtain the chemical valence states of Fe and Co, Figure 4A provides the high-resolution XPS spectra of Fe 2p. Four peaks, which are at ca. 711.2, 719.3, 724.6, and 731.4 eV, respectively,
can be observed. Among these peaks, 711.2 and 724.6 eV can be assigned to the Fe 2p$_{3/2}$ and Fe 2p$_{1/2}$, while the other peaks correspond to the satellite peaks of Fe 2p$_{3/2}$ and Fe 2p$_{1/2}$. Similarly to the previous result (Yamashita and Hayes, 2008), the obtained result suggests the presence of Fe$^{3+}$ in the obtained samples. As indicated in Figure 4B, the Co 2p high-resolution spectra also display four evident peaks. The peaks located at 780.9 and 796.6 eV originate from Co 2p$_{3/2}$ and Co 2p$_{1/2}$, and the peaks at 786.8 and 803.3 eV are the satellite peaks of Co 2p$_{3/2}$ and Co 2p$_{1/2}$, which indicate the existence of Co$^{2+}$ ions.
FIGURE 5 | TEM images of (a,b) S1, and (c,d) S2, respectively.

(Bennet et al., 2016). In general, the obtained XPS results are consistent with their XRD patterns.

Figure 5 provides the morphologies of the as-prepared S1 and S2. As shown in Figure 5a,b it can be observed that larger quantities of Co$_{x}$Fe$_{3-x}$O$_{4}$ and CoCO$_{3}$ NPs attached on the surface of GO and CNTs, similarly to the reported GO/CNT-Fe$_{3}$O$_{4}$ NCs (Wang et al., 2014). A closer TEM observation indicates that the NPs, CNTs, and GO nanosheets are well connected with each other to form a good three dimensional (3D) hierarchical networks structure. In the same manner as S1, as displayed in Figures 5c,d, the obtained S2 consists of large numbers of NPs, CNTs and paper-like GO, which presents a typical structure of 3D hierarchical networks. Combined with the XRD and XPS results, the conclusion suggests that the as-prepared S1 and S2 are 3D hierarchical networks structure of CoCO$_{3}$-Co$_{x}$Fe$_{3-x}$O$_{4}$/CNTs/GO quaternary NCs. Equally, the TEM investigation of S3 and S4 (as presented in Figure 6) shows that they are composed of Co$_{x}$Fe$_{3-x}$O$_{4}$ NPs with a relatively uniform size, CNTs and GO nanosheets, which bind together to establish the structure of 3D hierarchical networks. Unlike S1 and S2, the XRD, XPS and TEM results demonstrate that the as-prepared S3 and S4 are 3D hierarchical networks structure of Co$_{x}$Fe$_{3-x}$O$_{4}$/CNTs/GO ternary NCs. Generally, with the formation of Co$_{x}$Fe$_{3-x}$O$_{4}$ with large quantities, the obtained S3 has a larger Co$_{x}$Fe$_{3-x}$O$_{4}$ content compared to S2. It is well known that the value of minimum reflection loss ($R_{\text{min}}$) and its corresponding frequency bandwidth ($\text{FB}_{\text{cor}}$), with the reflection loss (RL) values lower than $-10$ dB (90% EM wave absorption) are the important characteristic for measuring high performance MAs (Ye et al., 2018). Further, it is very desirable that MAs can simultaneously have a low $R_{\text{min}}$ and large $\text{FB}_{\text{cor}}$ values. To investigate their MAPs, the values of RL were calculated based on the measured EM parameters and transmission line theory (Yusoff et al., 2002):

\[
Z_{\text{in}} = Z_{0} \sqrt{\frac{\mu}{\varepsilon}} \tanh \left( j \frac{2\pi f d \sqrt{\mu \varepsilon}}{c} \right) \tag{1}
\]
 Where $f$ is the frequency of the EM wave, $d$ is the thickness of absorber, $c$ is the velocity of light, $Z_0$ is the impedance of air, and $Z_{in}$ is the input impedance of absorber. Figure 7 provides the RL curves of S1, S2, and S3. As shown in Figures 7A,B, the RL$_{min}$ and large FB$_{cor}$ values for S1 are $-54.69$ dB at 11.08 GHz with the matching thickness ($d_m$) value of 8.57 mm and 2.36 GHz. The as-prepared S2 presents the RL$_{min}$ value of $-63.99$ dB at 14.96 GHz with the $d_m$ value of 6.60 mm (Figure 7C), and the FB$_{cor}$ value of 2.48 GHz (Figure 7D), respectively. Equally, as shown in Figures 7E,F, one can find that the RL$_{min}$ value for S3 is ca. $-63.36$ dB at 13.12 GHz with the $d_m$ value of 7.95 mm, and its FB$_{cor}$ value is 2.68 GHz. Moreover, the corresponding maximum absorption bandwidths ($AB_{max}$) with RL $< -10$ dB for S1, S2, and S3 (as provided in Figure 8) are 2.60 GHz with the $d_m$ value of 7.69 mm, 2.72 GHz with the $d_m$ value of 6.00 mm, and 3.08 GHz with the $d_m$ value of 6.33 mm, respectively. In general, the designed magnetic CNTs/GO NCs display very outstanding MAPs. The as-prepared S3 exhibits the superior comprehensive MAPs compared to S1 and S2. As summarized in Figure 9, the proposed magnetic CNTs/GO NCs present excellent comprehensive MAPs compared to the representative related NCs reported previously (Li et al., 2015; Yang et al., 2016; Zhang et al., 2016; Zhang K.C. et al., 2017; Zhang S. et al., 2017; Ren et al., 2018; Shu et al., 2018a,b; Hu et al., 2019; Lu et al., 2019; Xu et al., 2019).

To effectively understand their MAPs, Figure 10 gives their complex permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) and complex permeability ($\mu = \mu' - j\mu''$). As shown in Figures 10A,B, besides the shift of peak at ca. 13 GHz, the as-prepared samples present the similar $\varepsilon'$ and $\varepsilon''$ variation tendency in the frequency region owing to the same categories of samples. The complex permittivity exhibits several resonant peaks in the whole frequency range, indicating the multiple relaxation processes (Huang et al., 2019). Furthermore, the $\varepsilon'$ and $\varepsilon''$ values of samples present the decreased tendency with the frequency increasing from 2 to 18 GHz, which can be explained by the Debye theory (Wang et al., 2015). Overall, the obtained S2 has larger values of $\varepsilon'$ and $\varepsilon''$ compared to S1 and S3, which can be ascribed to the highest content of CNTs/GO in the as-prepared S2. According to the previous result (Li et al., 2017), the $\varepsilon'$ and $\varepsilon''$ values could be effectively enhanced by increasing the content of CNTs. The obtained S3 presents the lowest values of $\varepsilon'$ and $\varepsilon''$, which should be ascribed to the gradual formation of Co$_x$Fe$_{3-x}$O$_4$ with large quantities in NCs resulting in the decreasing content of CNTs/GO. Figures 10C,D present the $\mu'$ and $\mu''$ values of the obtained samples. Generally, it can be observed that the obtained samples exhibit the gradually growing $\mu'$ and $\mu''$ values with increased addition of Fe sources, which can be related to the enhanced content of Co$_x$Fe$_{3-x}$O$_4$ NPs in the NCs. Figure 11 provides the dielectric loss (tan $\delta_E = \varepsilon''/\varepsilon'$) and magnetic capabilities (tan $\delta_M = \mu''/\mu'$). Overall, the as-prepared magnetic CNTs/GO NCs display much higher tan $\delta_E$ values than those of tan $\delta_M$, which indicates that the attenuation of wave EM mainly originates from dielectric loss (Feng et al., 2020; Li et al., 2020). Moreover, the obtained S2 displays larger values of tan $\delta_E$ compared to S1 and S3. Further, the large
tan$\delta_m$ values of S3 are higher than those of S1 and S2. The difference in dielectric loss and magnetic loss abilities should be related to the different CNTs/GO and magnetic Co$_x$Fe$_{3-x}$O$_4$ NPs contents.

Based on the obtained EM parameters, RL curves and the previously reported mechanisms for the interpretation of the good MAPs (Liu et al., 2012, 2020), the excellent MAPs of the as-prepared magnetic CNTs/GO NCs can be explained by the geometrical effect. For this model, the values of $d_m$ and peak frequency ($f_m$) should satisfy the 1/4 wavelength equation (Liu et al., 2020):

$$d_m = \frac{nc}{4f_m\sqrt{\left|\mu\right|\left|\varepsilon\right|}} \quad (n = 1, 3, 5 \cdots)$$  \hspace{1cm} (3)

Figure 12 provides their comparison results between the experimental value of $d_m^{exp}$ (obtaining directly from their RL curves) and their theoretical curves of $d_m^{theo}$ [plotting by the formula (3)]. One can find that the scattered dots of $d_m^{exp}$ land on
the theoretical curves of $d_{\text{eff}}$. Consistent with the CoNi@PRM-NC, ZnFe$_2$O$_4$@C, and CNTs@MoS$_2$ reported elsewhere (Gao et al., 2019; Yan et al., 2019; Wang et al., 2020), the agreement between the experimental and theoretical values suggests the cancelation effect of reflected EM waves and a strong EM wave attenuation.

Based on the previous models and our obtained results (Wang et al., 2014; Shu et al., 2018a), as depicted in Figure 13, as for our designed magnetic CNTs/GO NCs, the possible paths for the EM wave attenuation are mainly attributed to the following facts: (i) the 3D hierarchical networks structure results in the multiple reflections and/or scattering of the EM wave (Figure 13A), which plays a significant role in the EM wave attenuation; (ii) many more interface contacts among magnetic NPs, CNTs, and GO nanosheets (Figure 13B) induce the formation of interface polarization with large quantities, which can effectively dissipate the energy of the incident EM wave; (iii) the designed NCs consisted of dielectric substances and magnetic NPs (Figure 13C), which can simultaneously provide the dielectric and magnetic loss mechanism. The excellent synergistic effect between them further improves the attenuation of the EM wave; (iv) the high conductivity of GO/CNT and a large number of defects and functional groups existing on CNTs (Figure 13B) and GO (Figure 13D) can generate massive electrons, defect dipoles and interface polarizations, which are also conducive of the dissipation of the EM wave.
**FIGURE 10** | EM parameters for the as-prepared samples: (A,B) complex permittivity, and (C,D) complex permeability.

**FIGURE 11** | Frequency dependence (A) dielectric loss tangent, and (B) magnetic loss tangent of the as-prepared samples.
FIGURE 12 | Comparison results between the calculated and experimental matching thicknesses for (A) S1, (B) S2, and (C) S3.

FIGURE 13 | Schematic illustration of possible EM wave absorption avenues of magnetic CNTs/GO NCs. (A,B) Multiple reflections, (C) dielectric and magnetic loss, and (D) defect dipole and interface polarization.

CONCLUSION

In summary, we proposed a one-step hydrothermal method to selectively synthesize different compositions of magnetic CNTs/GO NCs such as CoCO$_3$-Co$_x$Fe$_{3-x}$O$_4$/CNTs/GO quaternary NCs and Co$_x$Fe$_{3-x}$O$_4$/CNTs/GO ternary NCs. The TEM investigation indicated that the designed NC consisted of CoCO$_3$-Co$_x$Fe$_{3-x}$O$_4$ NPs, CNTs, and GO nanosheets, which were well bound together to form the good 3D hierarchical networks structure. Owing to the construction of many more interface contacts among magnetic NPs, CNTs, and GO nanosheets, an excellent synergistic effect between dielectric and magnetic loss substances, the as-prepared magnetic CNTs/GO NCs presented very extraordinary MAPs. The $RL_{min}$ and
FB\textsubscript{cor} values for the as-prepared samples were ca. −54.69 dB and 2.36 GHz, −63.99 dB and 2.48 GHz, −63.36 dB and 2.68 GHz, respectively. Moreover, owing to their low cost, lightweight and high stability, the proposed 3D hierarchical networks structures of magnetic GO/CNTs NCs exhibit very outstanding comprehensive MAPs, which may be used as promising candidates for high performance MAs and are very much worth further study.

**DATA AVAILABILITY STATEMENT**

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation, to any qualified researcher.

**AUTHOR CONTRIBUTIONS**

MW contributed to the conceptualization, methodology, investigation and provided resources. XQ contributed to the conceptualization, methodology, and the writing, review and editing of the manuscript. RX contributed to the conceptualization, methodology, and the writing, review and editing of the manuscript.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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