Study the effect of Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ doped electromagnetic and microwave absorbing properties on RGO based composites

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Abstract. Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ is one of the traditional microwave absorbing materials, and it has attract the focus of the whole world in the past years. The composites behave well in absorbing areas due to its coexistence of dielectric property and ferrimagnetism. The graphene is one of the new absorbing materials and it has been used in varies areas due to its excellent electrical conductivity, thermal conductivity and mechanical properties. In the work, the Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles were sufficiently compounded with the reduced graphene (RGO), and the microstructure and microwave absorbing performance of the composites were characterized. Consequently, the results indicated that the as-prepared materials perform well in microwave absorption area.

1. Introduction

The electromagnetic pollution has turned out to be one of the new environmental pollution in the pasted years due to the development of the electronics technique. The existence of the weaponry have faced the great challenge due to the continuous renewal of the radar detection technique. Therefore, the study of the microwave absorbing materials that with excellent microwave absorbing properties is of great importance [1-6]. The magnetic loss, resistance loss and dielectric loss characteristics account for the major role in microwave absorption abilities [7-9], then it is a point to combine the different kinds of microwave absorbing materials together to prepare the composites that is of better absorbing properties.

Reduced Graphene (RGO) have been widely applied in microwave absorption due to its excellent electrical conductivity and mechanical properties, and once it were compounded with other absorbing materials, the microwave absorption performance of the composites may be strengthened [10-12]. For example, once oxidized graphene is doped into the barium ferrite, thus the absorbing frequency band is widened and the absorbing abilities is increased [13-15]. Moreover, ferrite serve as a traditional microwave absorption due to its coexistence of dielectric property and ferrimagnetism. And the ferrite possess excellent microwave absorption characteristics when the Nickel and Zinc were added into the composites. Besides, once the material is prepared in nano-sized, the microwave absorbing ability would be greatly enhanced [16]. For example, the BaFe$_{11}$ (Ti$_{0.5}$Mn$_{0.5}$)$_3$O$_{19}$ nanoparticles have been prepared by coprecipitation method, and the result indicates that the particles were nano-sized, and the qualified absorption bandwidth (<10dB) covered 9.2~12.2GHz. In the work by Chung and his coworkers, hydrazine was applied in the fabrication of Fe$_3$O$_4$/RGO/polyaniline composites and the results suggest that the composites of electrical conductivity up to 2.6 S/cm exhibited absorption shielding ~26 dB when the composite thickness was 2.5 mm.
In this work, Ni-Zn ferrite were compounded with RGO by sufficient ultrasonic agitation method. And the microstructure of the as-prepared samples were characterized by XRD and SEM. Then, the electromagnetic parameters of the composites were tested by VNA. Consequently, the microwave absorption performance was discussed.

2. Experimental

The RGO were prepared by hummers method, the mixture of 1 g NaNO$_3$ and 2 g graphite powders were added into concentrated 120 ml H$_2$SO$_4$ under stirring at 0 ℃ for 0.5 h in an ice bath. Then stirred the solution at 0℃ for 1 h. Subsequently, 6 g KMnO$_4$ powders were slowly added into the mixture. Until stirring for 2h, the mixture was settled at 30 ℃ for 0.5 h and 150 ml deionized water was slowly added. After the mixture was heated and kept at 98 ℃ for 15 min, 50 ml 5% H$_2$O$_2$ was slowly added into the solution. Then, the resulting solution was washed with 5% HCl and deionized water for 5 cycles and the final solution was the GO aqueous solution.

The Ni-Zn ferrite were prepared by sol-gel method, citric acid monohydrate were added to the mixed solution of zinc nitrate and nickel nitrate (quantitative weigh) and regulated the pH to 6 by ammonia solution, kept heated reaction until the solution turn to sol rubber, then keep heating the solution at 80 ℃ until the samples turn into xerogel, finally, the samples were calcined in muffle for two hours under 650 ℃, then would get the Ni-Zn ferrite powder. The RGO and the as-prepared ferrite were sufficient ultrasound for 50 min, and then add the ferrite into the RGO solution. Expecially, in experiment, the mass ratio of the RGO in the composites is 5%. Followed by fully stirred method. Then the mixture were freeze drying, and the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ were gained.

The microstructure of the as-prepared samples were investigated by X-ray diffraction (XRD, Rigaku Ultima IV, Cu-Ka) in the range of 5°~90°, with a scan speed of 3 s and a step size of 0.02° in 2θ. And the electromagnetic properties of the samples were tested by the vector network analysis (VNA, KEYSIGHT E5071C) via coaxial method.

3. Results and discussion

The XRD patterns of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles were shown in figure 1. Apparently, the wide peak between 8° and 20° indicated the existence of RGO in the nanoparticles, and all the peaks that were shown between 20° and 8° indicated the existence of Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$. Therefore, the results indicate the Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles were successfully grown with the RGO, and the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles were gained.

![Figure 1. XRD patterns of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles.](image)

As is shown in figure 2, it is obvious that the size of the Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ is within 100 nm, indicating the nano-sized Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ particles were gained. Moreover, the RGO is apparently to
see in the picture, and the Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles is well grown on the surface of RGO, indicating the RGO and Ni-Zn ferrite are well growed together by sufficient ultrasonic agitation method.

![SEM image of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles.](image)

**Figure 2.** SEM image of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles.

In the preparation of the samples, paraffin was used as the matrices, and the mass ratio of the composites in the samples were 90%. The inner diameter of the specimens is 3 mm, and the external diameter is 6.9 mm. Consequently, the permittivity and permeability of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles were gained in the range of 2~12.4 GHz (figure 3). As is shown in figure 3a, the real permittivity of the composites is around 4.8, and the imaginary permittivity of the composite is around 0.1. Besides, the real permeability of the composites is around 1.2, and the imaginary permeability of the composites is around 0.2.

![Permittivity and permeability of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles](image)

**Figure 3.** Permittivity and permeability of the RGO/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles (a: permittivity; b: permeability).

The reflection loss ($R_L$), which could be a reflection of the ability of the microwave absorption performance of the absorbing layers on the ideal metal plane, was achieved by the relation

$$R_L = 20 \log \left( \left| \frac{Z_{\text{in}}}{Z_m} \right| \right).$$

(1)

Moreover, the normalized input impedance ($Z_{\text{in}}$) of absorption layer is given as

$$Z_{\text{in}} = (\mu/\epsilon)^{1/2} \tanh \left[ 2 \pi d (\mu/\epsilon)^{1/2}/c \right].$$

(2)
where \( c \) is the light velocity, \( f \) the microwave frequency, \( d \) the thickness of the absorber, \( \mu_r \) and \( \varepsilon_r \) the complex permittivity and permeability of the specimens, respectively [17]. Therefore, the microwave absorption performance of the as-prepared samples were characterized.

Figure 4 exhibits the microwave absorption performance of the RGO/Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles. The microwave absorbing abilities at different thickness are shown in figure 4a, it is obvious that the composites possess best microwave absorption performance at the thickness of 6 mm. And the absorption intensity reaches -19.5 dB at 5.68 GHz, and the qualified bandwidth of the sample reaches 1.6 GHz. Besides, the three-dimensional microwave absorption performance is shown in figure 4b.

![Figure 4. Microwave absorption performance of the RGO/Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles.](image)

According to the discussion of the electromagnetic property and microwave absorbing ability of the as-prepared RGO/Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles, the results indicate that it is a potential method to compound the RGO with the Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) composite to synthesis a new microwave absorption material. The SEM results indicate that the Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles are well compounded with the RGO. Therefore, the as-synthesised RGO/Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles might serve as a potential material in microwave absorbing area.

4. Conclusion

The sufficient ultrasounded method were conducted to synthesis the RGO/Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles. The microstructure of the as-prepared composites were characterized. The results indicate that the Ni\(_{0.5}\)Zn\(_{0.5}\)Fe\(_2\)O\(_4\) is well compounded with the RGO, and the as-prepared samples possess well electromagnetic properties and microwave absorption performance. The composites show best wave absorption performance at the thickness of 6 mm, and the qualified bandwidth of the bandwith reaches 1.6 GHz. Moreover, the absorption intensity of the composites reaches -19.5 dB at 5.68 GHz. Thus the study provide a guide for preparing the composites that possess excellent microwave absorption performance.

5. References

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