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Simulation and Experiment for Microwave Absorption of Carbon-coated Nickel Nanoparticles Composites

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Abstract: The carbon-coated nickel Ni(C) nanoparticles exhibit excellent microwave absorbing performances, due to the core/shell microstructure. The microwave absorbing property of paraffin-Ni(C) composites was simulated in this study. And the microwave relative permittivity and permeability of Ni(C)/paraffin composites and nano-carbon/paraffin composites were measured, respectively. The results revealed that Ni(C) nanoparticles have better magnetic loss than pure nano-carbon. Ni(C) nanoparticles exhibit main magnetic loss in low frequencies and main dielectric loss in high frequencies which were caused by carbon shells and Ni nano-cores, respectively. Due to the dielectric shells and ferromagnetic cores, the Ni(C) nanoparticles can establish a better electromagnetic match which can obtain a strong microwave absorption. And an excellent microwave absorption property for Ni(C) nanoparticles was obtained. Based on the datum of relative complex permittivity and permeability measured, the reflection loss according to the transmit-line theory was simulated. The reflection loss of 2 mm double-layer coating consisting of Ni(C)/epoxy resin composites was measured. The results indicated that the maximum reflection loss reached -26.73 dB at 12.7 GHz and the less than-10 dB band width was about 4 GHz which basically agreed with the calculational results.

Key words: Carbon-coated nickel nanoparticles, microwave absorption, reflection loss, the relative permittivity, the relative permeability

INTRODUCTION

With the development of wireless communications, microwave absorbers are becoming increasingly important for applications in special fields such as silent rooms, radar systems and military applications (Jung *et al.*, 2006; Ramasubramaniam *et al.*, 2003; Zhang *et al.*, 2007a,b). From the very beginning of the development, nano-materials as a new type of microwave absorber, have been paid more and more attention to by many scientists due to their special properties, such as surface effect, quantum size effect, small size effect and so on. Up to now, microwave nano-absorbers researched all over the world include nano-metals, nano-alloys, nano-oxides, nano-conductive polymers, the composites of nano-metals and insulation mediums etc., (Cao *et al.*, 2007; Chen *et al.*, 2004, 2009; Liao *et al.*, 2010; Qiu *et al.*, 2010; Zuo *et al.*, 2008). The materials with nanowire microstructure would be potential microwave absorbers, the maximum absorption of 8.37 dB was reported when the concentration of the ZnO nanowires is 6% and enhanced to be 12.28 dB when the concentration increased to 7% (Chen *et al.*, 2004). Cao *et al.* (2007) prepared a cage-like material of ZnO/SiO₂ and studied its microwave absorption properties. It was found that the maximum

absorption of the cage like ZnO/SiO₂ was up to 10.68 dB at the frequency of 12.79 GHz and the absorption range under -6.0 dB was from 10-18 GHz.

In recent years, many researchers focus on nano-materials with a special microstructure. Carbon nanotubes (CNTs), due to their special microstructures and fancy physical phenomena, have been highly concerned by many researchers in the field of microwave absorption (Lin *et al.*, 2007; Qing *et al.*, 2010; Tong *et al.*, 2011; Ye *et al.*, 2010; Zhang *et al.*, 2009, 2011; Zhao *et al.*, 2010; Zou *et al.*, 2010). Wu and Kong (2004) prepared multiwall CNT-epoxy resin composites and (Che *et al.*, 2004) reported the microwave absorption properties of CNT-epoxy nanocomposites with Fe filler embedded into CNTs which have high microwave absorption properties. Zou *et al.* (2010) has reported that multi-walled carbon nanotubes filled with Ni nanowire had good microwave absorbing properties.

The core/shell microstructure, due to their interfaces of different materials between the cores and the shells, may be an excellent structure for microwave absorbing. Zhang *et al.* (2006) have theoretically researched the microwave absorption property of carbon coated nickel (Ni(C)) nanoparticles. Their maximum theoretical absorption which was calculated by computer, reaches

32 dB at 13 GHz with 2 mm thickness. But at present reports about their experimental absorption are rarely seen. In this study, the experimental absorption, the relative complex permittivity and permeability of Ni(C)-paraffin wax composites were measured. The possible absorbing mechanisms were discussed as well.

MATERIALS AND METHODS

Pretreating and dispersing of Ni(C) nanoparticles: Ni(C) nanoparticles and Sodium Dodecyl Benzene Sulfonate (SDBS) which were mixed according to certain proportion, were added into a certain amount of deionized water. The mixed solution was refluxed for 30 min at room temperature, then the precipitates which were Ni(C) nanoparticles mixed with SDBS, were separated out of the solution. The slurry of Ni(C) nanoparticles were prepared by adding these pretreated Ni(C) nanoparticles into the anhydrous ethanol with the addition of some dispersants then dispersing these Ni(C) nanoparticles by methods of electric stirring for 10 min and ultrasonic dispersion for 10 min successively.

Preparation of coaxial samples: The coaxial samples of paraffin-Ni(C) and nano-carbon/paraffin composites were prepared by uniformly dispersing the Ni(C) nanoparticles in paraffin matrix which were toroidal shaped samples with 7 mm outer diameter and 3 mm inner diameter, respectively. The electromagnetic parameters of the coaxial samples were measured by using the system of AV3618 vector network analyzer.

Preparing of the coating samples: With Ni(C) nanoparticles pretreated as filler, epoxy resin as the main component and anhydrous ethanol as dispersion medium, the Ni(C) nanoparticles/epoxy resin coating were prepared by dispersing these slurry of Ni(C) nanoparticles into epoxy resin. These coatings were painted onto the 180 mm H180 mm aluminum plate, then the reflection samples of microwave absorbing were obtained. The reflection losses were measured by AV3618 vector network analyzer.

RESULTS AND DISCUSSION

TEM analysis: Figure 1 shows the TEM images of the carbon-coated nickel nanoparticles. As shown in Fig. 1, the carbon-coated ferromagnetic metal nanoparticles are about 20-80 nm in diameter and their average diameter is about 40 nm.

Electromagnetic properties: Figure 2 shows the relative complex permittivity and permeability of the paraffin-Ni(C)

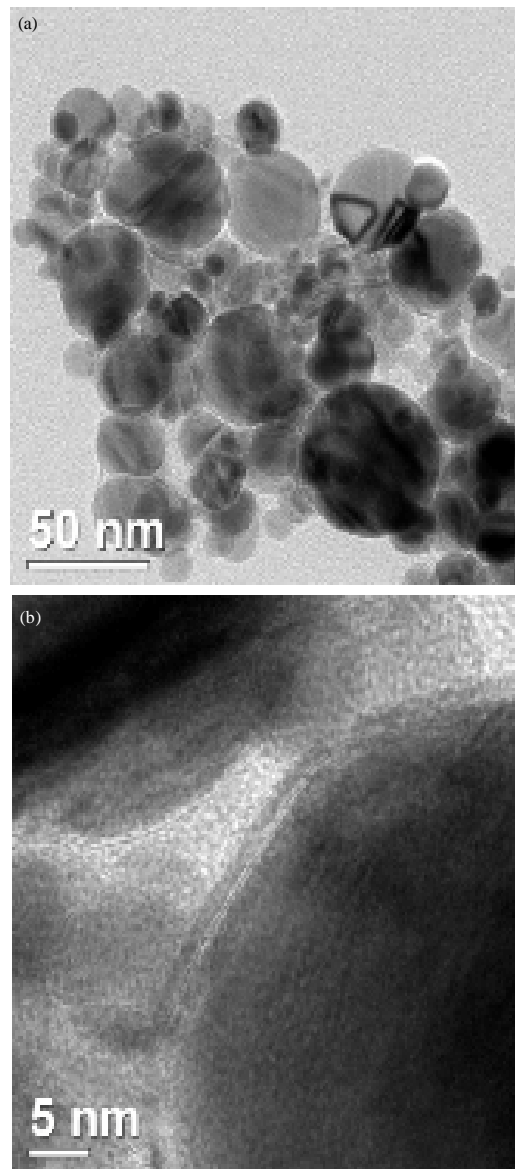


Fig. 1(a-b): TEM images of the carbon-coated nickel nanoparticles

composites with 50 wt.% Ni(C) nanoparticles measured over 2-18 GHz. It can be found that the values of real part of relative complex permittivity (ϵ') decline sharply from 12.8-4.9 in a frequency range from 2-18 GHz and the maximum value is 12.8 at 2 GHz. The maximum value of imagine part of relative complex permittivity (ϵ'') is 7.1 at 14.8 GHz which implies an obvious dielectric loss in high frequency. The dielectric loss may be attributed to the relaxation of polarization between the core/shell interfaces as the frequency varies. The values of real part of relative complex permeability (μ') decline from 1.5-1.0 in the

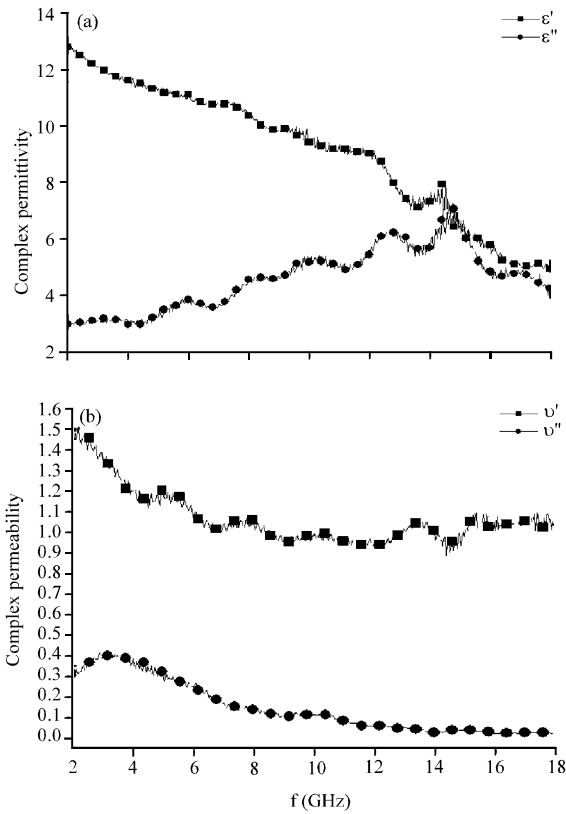


Fig. 2(a-b): Relative complex (a) Permittivity and (b) Permeability of the paraffin-Ni(C) composites with 50 wt.% Ni(C) nanoparticles measured over 2-18 GHz

frequency range of 2-18 GHz, meanwhile, the maximum value of imaginary part of relative complex permeability (ν'') is 0.4 at about 4 GHz, suggesting a natural resonance happened at that frequency. It is worthy of note that the values of ν'' are between 0.02 and 0.4 in the whole measured frequency range which implies some magnetic losses occurred in the paraffin-Ni(C) composites. It is reasonable that the magnetic loss is attributed to magnetic Ni cores.

Figure 3 shows the relative complex permittivity and permeability of the nano-carbon/paraffin composites with 15 wt.% carbons nanoparticles measured over 2-18 GHz. The values of ϵ' decline from 24.0-10.9 and the maximum value is 24.0 at 2 GHz, while the maximum value of imaginary part of relative complex permittivity (ϵ'') is 16.3 at 2 GHz. Both are higher than the maximum values of paraffin-Ni(C) composites. The permeability of nano-carbon/paraffin composites is lower than that of paraffin-Ni(C) composites. These results show nano-carbons are of dielectric microwave absorption properties and Ni(C) nanoparticles are of both dielectric and magnetic

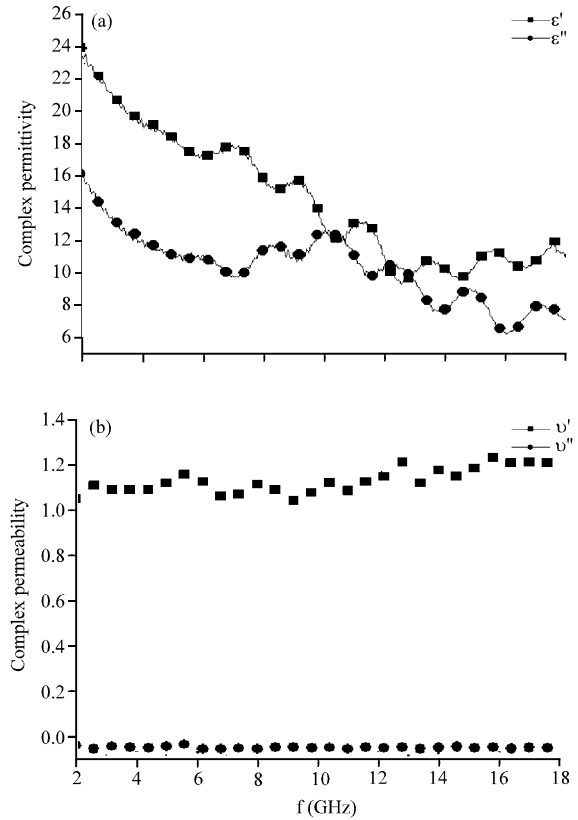


Fig. 3(a-b): Relative complex (a) Permittivity and (b) Permeability of nano-carbon/paraffin composites with 15 wt.% carbons nanoparticles measured over 2-18 GHz

microwave absorption properties. The relative complex permeability of nano-carbons reveals that the values of ν' are between 1.0-1.3 and the values of ν'' are almost zero which implies there are no magnetic losses occurred in the nano-carbons. The results show that magnetic Ni cores coated by carbons can improve the magnetic loss of carbon nanoparticles.

On the other hand, according to the free electron theory, $\epsilon'' \approx 1/2\pi\epsilon_0\rho f$, where ρ is the electrical resistivity. The lower ϵ'' indicates a higher electrical resistivity. Therefore, it can be concluded that the paraffin-Ni(C) composites have higher electrical resistivity in comparison with nano-carbons. The higher electrical resistivity may result from few point defects existing in Ni(C) nanoparticles (Watts *et al.*, 2003) than nano-carbons. The protective graphite shells on the surface of Ni nanoparticles can effectively disperse Ni(C) nanoparticles in paraffin (Che *et al.*, 2006; Liu *et al.*, 2005). It is convinced that the core/shell microstructure of the Ni(C) nanoparticles, especially the graphite shells on the surface of nanoparticles, can improve the microwave absorption of nano-carbons particles.

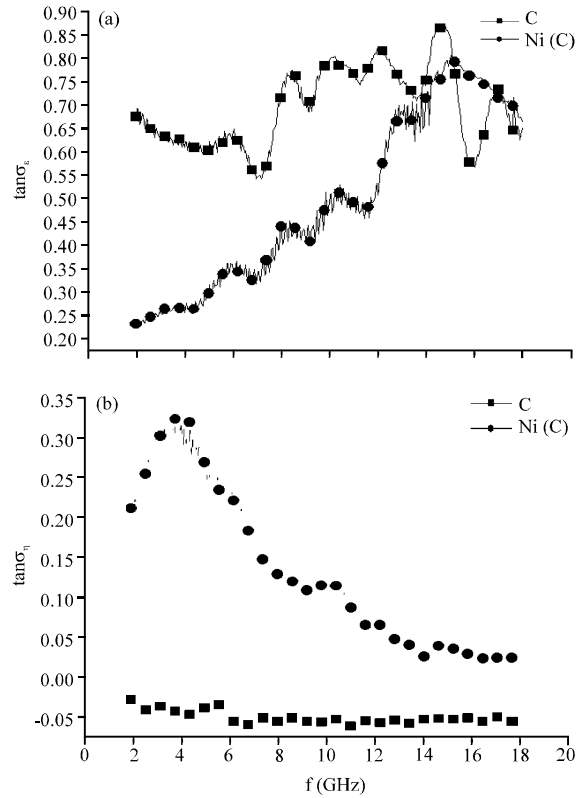


Fig. 4(a-b): Relationship between the loss tangent (a) $\tan\sigma_e$ and (b) $\tan\sigma_i$ frequencies for Ni(C) nanoparticles and nano-carbons

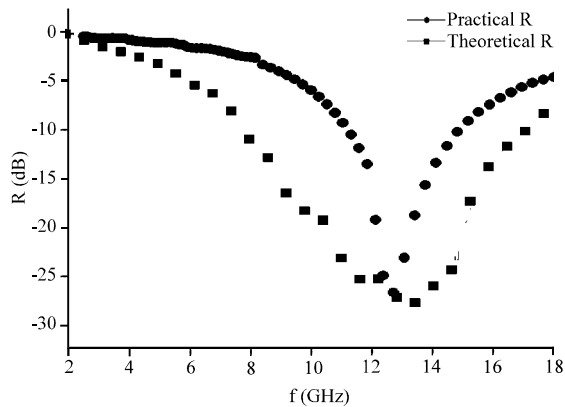


Fig. 5: Simulated and practical reflection loss of the 2 mm double-layer coating of carbon-coated nickel/Epoxy resin

Generally, with the interaction between dielectric loss, magnetic loss and the electric resistivity, microwave absorbers may appear an absorbing peak at certain frequency. The electromagnetism loss mechanism of the materials can be explained by the characteristic change of

loss tangent (loss factor). Figure 4 shows the relationship between the loss tangent ((a) $\tan\sigma_e$ and (b) $\tan\sigma_i$) and frequencies for Ni(C) nanoparticles and nano-carbons. It is revealed that the values of $\tan\sigma_e$ (real part of the loss tangent) for Ni(C) nanoparticles are comparatively smaller at lower frequencies (below 15 GHz) than those for nano-carbons, but bigger at higher frequencies (above 15 GHz). The values of $\tan\sigma_e$ for Ni(C) nanoparticles in higher frequencies are greater than in low frequencies and the maximum value is 0.80 at 15 GHz. It means that Ni(C) nanoparticles exhibit main magnetic loss at low frequencies and main dielectric loss at high frequencies. The maximum values of $\tan\sigma_i$ (imaginary part of the loss tangent) is 0.32 at 4 GHz for Ni(C) nanoparticles and is zero for nano-carbons. In comparison with nano-carbons, the magnetic loss of Ni(C) nanoparticles caused by Ni nano-cores is greater and may improve further their microwave absorbing properties.

Theoretical simulation and practical measure of reflection loss:

Generally, the excellent microwave absorbers result from the efficient complementarity between the relative complex permittivity and permeability of materials. The existence of graphite shells and magnetic Ni cores for Ni(C) nanoparticles can be favourable factors to set up a better electromagnetic match (Zhang *et al.*, 2006). As a result, to further research the microwave absorbing property of paraffin-Ni(C) composites, a reflection loss simulation has been carried out. Based on the data of relative complex permeability and permittivity measured above, double-layer microwave absorbing coating (2 mm in thickness) filled with paraffin-Ni(C) composites was prepared. And the theoretical reflection loss was calculated according to the transmit-line theory (Yusoff *et al.*, 2002). Figure 5 shows the simulated and practical reflection loss of the 2 mm double-layer coating of carbon-coated nickel/Epoxy resin. As shown in Fig. 5, the maximum calculational reflection loss reached -29.00 dB at 13.1 GHz and the absorption frequency range under -10 dB was over 7.6-17.4 GHz. The carbon coated nickel, due to its particular structure, can improve the electromagnetic match and obtain strong microwave absorbing.

To further prove theoretical simulation of the excellent microwave absorbing property of Ni(C) nanoparticles, carbon-coated nickel/epoxy resin microwave absorbing coating was prepared by dispersing the Ni(C) nanoparticles in epoxy resin and painting them onto the aluminum plate. The coating is double-layered and its total thickness is 2 mm. Its inner layer is 1 mm thickness and is composed of carbon-coated nickel/epoxy resin composites with 30 wt.% Ni(C). And its outer layer

is also 1 mm thickness, but consists of carbon-coated nickel/epoxy resin composites with 50 wt.% Ni(C). The practical reflection loss, as shown in Fig. 5, was measured by AV3618 Vector Network Measurement System. It is shown that the maximum practical reflection loss reaches -26.73 dB at 12.7 GHz and the absorption range under -10 dB is from 11.2-14.8 GHz. The absorbing peak measured is close to that of simulation which shows that the Ni(C) nanoparticles indeed have an excellent microwave absorbing property.

CONCLUSION

In conclusion, the composites including Ni(C) nanoparticles exhibit excellent microwave absorbing performances which is attributed to the core/shell microstructure of Ni(C) nanoparticles. The microwave absorbing mechanism of carbon-coated nickel nanoparticles, with dielectric shells and ferromagnetic cores, are a combination of dielectric loss and magnetic loss. The theory simulating results indicate that the maximum theoretical reflection loss reaches -29.00 dB at 13.1 GHz and the absorption frequency range under -10 dB is over 7.6-17.4 GHz. The experimental shows that the maximum reflection loss is 26.73 dB at 12.7 GHz and the absorption bandwidth less than -10 dB is 4 GHz with 2 mm double-layer coating. A more excellent microwave absorber can be prepared by choosing an optimum of layer number and the ratio of Ni(C) nanoparticles in the composites.

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