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Microwave Absorption Properties of Flake Graphite and Carbonyl-iron Particles filled Aliphatic Polyurethane Resin

[1*]Xinwei Ji, [2]Ming Lu, [3]Feng Ye, [4]Qian Zhou

[1*] College of Field Engineering, PLA University of Science and Technology.
3 shi , mail-box 2862#, No.11 AnNing Zhuang Road, Haidian District ,Beijing, 100095, China
[2] College of Field Engineering, PLA University of Science and Technology,
1 Hai Fu Xiang Street , Nanjing, 210007, China
[3] College of Field Engineering, PLA University of Science and Technology,
1 Hai Fu Xiang Street , Nanjing, 210007, China
[4] College of Field Engineering, PLA University of Science and Technology,
1 Hai Fu Xiang Street , Nanjing, 210007, China

[1*]Email: jxw0127@gmail.com Tel: 86-18701564183

Abstract – Microwave absorbing coatings composed of carbonyl iron particles (CIP) and flake graphite (FG) as absorbent and Aliphatic Polyurethane (APT)Resin as matrix were prepared with the method of spraying. The electromagnetic properties of the CIP/FG composites as a function of the frequency and the mass fraction of FG were studied. Their electromagnetic and microwave absorbing properties could be tuned by changing the weight fraction of FG at 4–18 GHz. The electromagnetic parameters increase with increasing the FG contents in the composites. Meanwhile, the peak value of reflectivity moves to low frequency and the minimum reflection loss (RL) value reached −36 dB at 6.0 GHz. The absorption range with RL lower than −10 dB was obtained at 5.23–8.32 GHz for absorber filled with 50 wt.% CIP and 7.5 wt.% FG at 1.2 mm. The surface density of the fabricated coatings is approximately 2.7 kg/m^2 . It is an effective method to fabricate absorbing materials by mixing magnetic absorber CIP and FG as conductive absorber filled insulating APT, meeting the requirements of RAMs.

Keywords – Flake graphite, carbonyl iron particles, reflectivity, electromagnetic wave absorbing coating

1. Introduction

Currently a wide range of materials are used for the design and development of RADAR absorbing materials (RAMs). [1]prepared microwave absorbing materials composed of ordered mesoporous carbon (OMC) as absorbent and paraffin as matrix. The minimum reflection loss (RL) value reached −9.3 dB at 8.0 GHz and the absorption range with RL lower than −5 dB was obtained at 5.8–14.4 GHz for a single-layer absorber filled with 1.98 wt.% OMC at 3.0 mm. [2]measured the complex permittivity spectra and conductivity of graphite nanoplatelet/epoxy composites to assess the application potential of these materials as conductive coatings and/or microwave absorbers with high structural integrity. [3] prepared carbon fiber filled low-density polyethylene matrix composites by means of the Hakke driver. The dielectric properties of the FG/LDPE composites as a function of the frequency and the volume fraction of FG were studied. The percolation theory and the simple concept of polarization in the capacitors were employed to explain their experimental results. [4]investigated the reflection loss properties for the flake-shaped carbonyl-iron particles and paraffin composite. [5]presents the processing and characterization of electromagnetic radiation absorbing paints and sheets based on magnetic and dielectric materials dispersed in polymeric matrices.

The microwave has two components, electric and magnetic which are acting perpendicular to each other[6]. It is required to cancel out both of these components to make microwave invisible. Generally this has been achieved by incorporating magnetic and electrically conducting fillers into a matrix. In the case of a magnetic material, losses are produced by changes in the alignment and rotation of the magnetization spin[5]. In the case of a dielectric material, attenuation is promoted by dielectric particles involving ohmic losses[7]. This work introduced CIP as magnetic absorber and FG as conductive absorber filled insulating APT resin coatings.

2. Experimental work

FG and CIP filled aliphatic polyurethane resin were used to fabricate microwave-absorbing coatings. FG and commercial-grade CIP used in this work were brought from Qing Dao HuaTai and Jiang Su TianYi company, respectively. The CIP are spherical particles of around 2~5 μm in diameter. The FG particles are flake with the thickness about 500 nm. The specimens of 1#, 2#, 3#, 4# and 5# contained concentration of 50 wt% CIP particles, and 0 wt%, 2.5 wt%, 5 wt%, 7.5 wt% and 10 wt% FG. The FG particles and CIP were individually dispersed in the acetone solution by an ultrasonic bath at room temperature for 5 min. Subsequently, the solution of FG particles and CIP were mixed together and stirred for 10 min at 1500 rpm. Then the toluene, titanate coupling agent and dispersant had been added into the mixtures and stirred for 10 min at 1500 rpm. After aliphatic polyurethane resin had been added into the mixtures, the mixtures were uniformly mixed by stirring at 1000 rpm for 30 min at room temperature. After well dispersion of the fillers, the mixtures were sprayed on a flat aluminum plate (300*300*2 mm) by spray gun and kept in oven at 80°C for complete drying. The spraying and drying procedures were repeated for four times. The thickness of specimens prepared was maintained to approximately 1.2 mm.

The morphologies of the specimens with CIP and (or) FG were observed by a field-emission scanning electron microscope (FE-SEM, Hitachi S-4800), operated at 15.0 kV. The cylindrical toroidal samples with 3.04 mm in inner diameter, 7.0mm in outer diameter and 2.0 mm in thickness were fabricated by uniformly mixing wax with 40 wt% and then pressed into cylindrical compacts. The transmission/reflection method was applied to determine the relative complex permeability and permittivity of the sample-wax composites through an HP-8722ES vector network analyzer system. For the electromagnetic characterization of the specimens, reflection/absorption measurements in the frequency range of 4-18 GHz were carried out using the Naval Research Laboratory (NRL) arch method[8].

3. Result and discussion

3.1 Morphological Study

The microstructures of the 4# coating were revealed by SEM images as shown in Fig.1. It can be seen that the CI particles and FG were uniformly separated in the APU resin matrix. The microwave-absorbing coatings can be successfully fabricated through the uniform dispersion of two different fillers. Furthermore, with the concentration of FG increased, some FG might be cracked into smaller flake due to a strong mixing shear force during the mixing process.

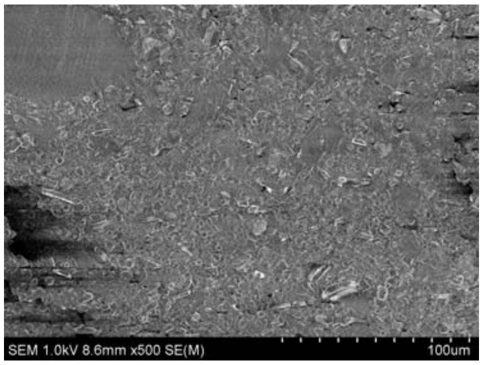
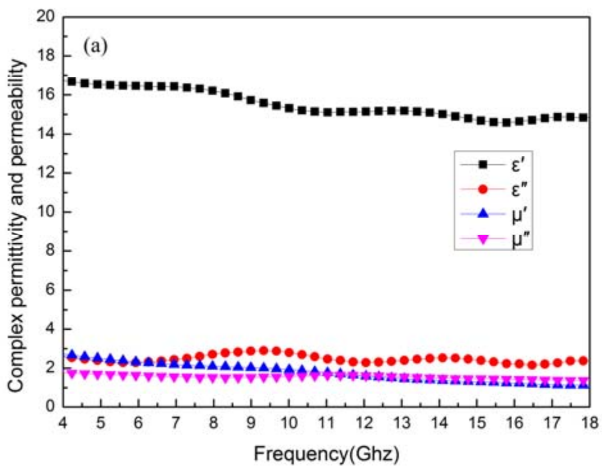


Figure 1. SEM images of the 4# coatings with CIP 50wt.% and FG 7.5wt.%

3.2 Complex permittivity and permeability

Complex permittivity (ε) and magnetic permeability (μ) are parameters related to a material's dielectric and magnetic properties. They are among the most important characteristics of absorbing materials, and are directly associated with their absorbing properties[5]. In order to investigate the intrinsic reasons for microwave absorption of the coatings, complex permittivity and permeability of the CIP and FG particles were measured using the T/R coaxial line method. The complex permittivity in microwave frequency band is simply expressed by $\varepsilon = \varepsilon' - j\varepsilon''$, and the complex permeability is expressed by $\mu = \mu' - j\mu''$. The real part of complex permittivity (ε') represents the energy storage capacity. And the imaginary part of the complex permittivity (ε'') and permeability (μ'') account for the energy loss dissipative mechanisms in the materials[9]. Fig.2 shows the relative values of permittivity and permeability of CIP and FG with paraffin. With the increasing of frequency, both the permittivity and permeability of CIP decreased slowly in the rage of 4-18GHz. It can be seen from Fig.2(b) that the relative permittivity of FG also decreased with the increasing of frequency, meanwhile, the permeability of FG almost keep constant. The real part is about 1, and the imaginary part is approximately 0, which means FG has no contribution to magnetic loss.



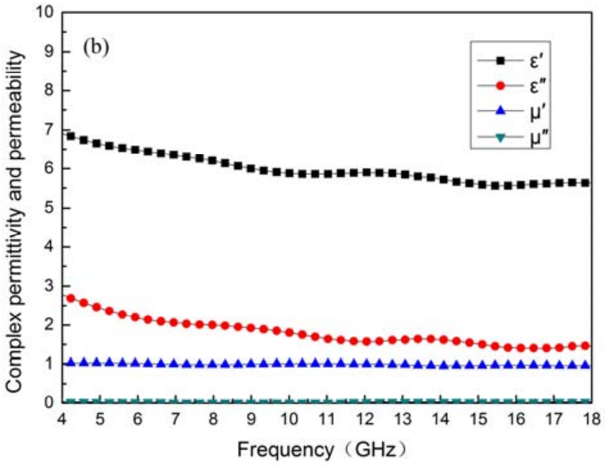
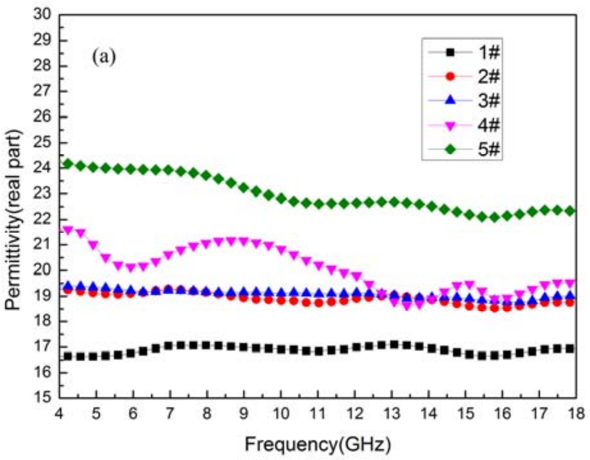


Figure 2. The relative permittivity and permeability of (a) CIP with 50wt.% paraffin and (b) FG with 80wt.%

The samples of constant CIP and different FG with paraffin were prepared to measure the electromagnetic parameters of hybrid as absorbent in the coatings. Fig.3. represents the real part (a) and imaginary part (b) of the complex permittivity about the composites with different FG content. It is visible that, with the increasing of FG content, the real part increases from about 17 to 23, and the imaginary part increases from about 2.5 to 10. The dielectric constants decrease slowly with increasing the frequency and rise gradually with increasing the FG contents in the composites. As the mass fraction of the FG is up to 10% (5#), an abrupt increase in the dielectric constant occurs. In general, the dielectric constant of composites depends on the configuration and internal fractal structure, which is proportional to the quantity of charge stored on the surface when the composites are under an applied electric field[9]. From this point of view, the higher dielectric constant can obtain in the coatings with the higher FG content.



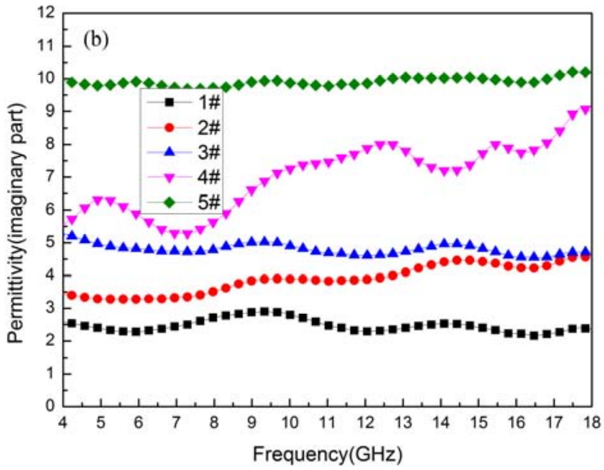


Figure 3. The real part and imaginary part of complex permittivity of specimens with different FG content

As can be seen from Fig.3 (b), the imaginary part of permittivity of the composites increased with the FG content increasing. When the conductive filler content got below the percolation threshold, the main term affecting the ε'' is the interfacial polarization and its associated relaxation, while the conductivity term plays a secondary role[9]. With the FG content increased gradually, composite dielectric constant increases quickly. This is because the FGs are not in direct contact between the particles. The total capacitance and dielectric constant rise due to the formation of a large number of micro capacitance (super capacitor network)[10]. At the same time, dielectric loss of the composite is rising rapidly, caused by a partial flow current, due to the spacing between particles decreased gradually.

In general, in the electric field of low frequency, the polarization process can keep up with the change of the electric field rate, polarization and electric field change synchronously[10]. So all the polarization is contribute to dielectric constant. In high frequency, the electric field changes so fast that partial polarization process can not keep up with change in electric field. The interfacial polarization, the orientation polarization, and the displacement polarization process withdraw from contribution to dielectric constant successively with frequency increasing[10]. The real part of the dielectric constant decreased with the increasing of frequency in the range of 4-18GHz. There is frequency dispersion property, which facilitates the broadband absorption. At the same time, the imaginary part of the permittivity increases with different increment, respectively. With the frequency increasing, a strong vortex is generated due to that polarization can't keep up with the change of electric field under the electromagnetic wave. And the Eddy currents will release a lot of heat in internal flow, so the vortex causes a portion of incident electromagnetic wave energy consumption[11].

Fig. 4 shows the real and imaginary part of complex permeability of the samples as a function of frequency. The real part of the complex permeability of the samples decreased from approximately 2.7 to 1.1, and the imaginary part decreased from about 1.8 to 1.3, with frequency increasing from 4 to 18 GHz.

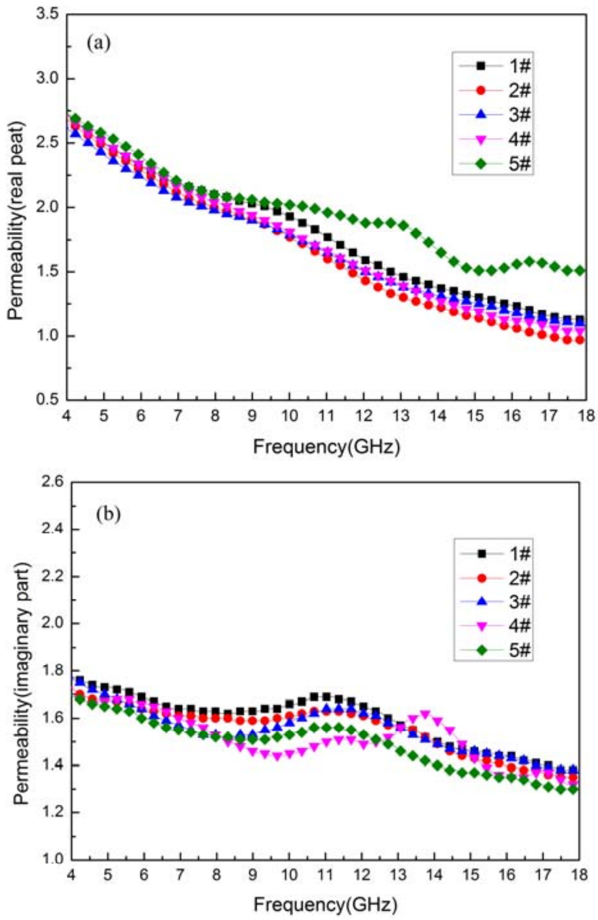


Figure 4. The real part and imaginary part of complex permeability of specimens with different FG content

By comparing with the increasing of the complex permittivity, the complex permeability of the samples remains nearly constant with the FG content increased, indicating no magnetic loss contribution from the FG particles.

3.3 Microwave Absorbing Properties

Fig.5 shows how changes in the FG content affect the energy absorption. It can be observed that the resonant absorption peak is displaced with respect to frequency, and its amplitude vary significantly due to the increasing of FG content. The coatings of 1#, 2#, 3#, 4# and 5# contained concentration of 50 wt% CIP and 0wt%, 2.5 wt%, 5 wt%, 7.5 wt% and 10 wt% FG, respectively. Specimens 2# and 3# have RL values below -5dB in the range of 6.14GHz~10.60GHz and 6.0GHz~18GHz. The minimum RL value of specimen 4# reaches -36.22dB at the frequency 6.62GHz. And specimen 5# has RL values less than -5dB in the rage of 4GHz~10.48GHz.

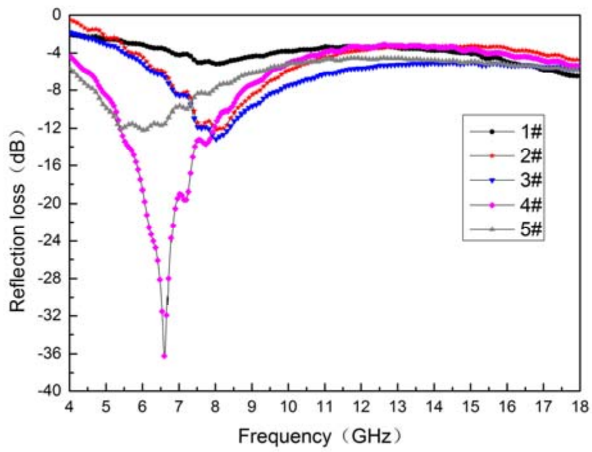


Figure 5. Reflection loss of the coatings with consistent CIP and different content of FG 1# 0 wt%, 2# 2.5 wt%,3# 5 wt%, 4# 7.5 wt% and 5# 10 wt% respectively

The minimum reflection loss increased and moved to lower frequency with the FG content increasing, and the peak value of specimen 4# with CIP 50wt.% and FG 7.5wt.% is −36.22 dB at 6.62 GHz. But when FG content continues to increase, the reflection loss of specimen 5# with 10wt.% FG decreased on the contrary. The lower reflection loss of the 5# specimen can probably be attributed to the coating with too large a complex permittivity, which can cause additional reflective wave on the coating surface[9]. Therefore, when the FG content increased, more reflection can be caused on the coating surface and decrease the microwave absorption.

4. Conclusion

Microwave absorbing coatings composed of CIP and FG as absorbent and APT Resin as matrix were prepared with the method of spraying. The absorption range with RL lower than −10 dB was obtained at 5.23–8.32 GHz for absorber filled with 50 wt.% CIP and 7.5 wt.% FG with coating thickness at about 1.2 mm. The surface density of the fabricated coatings is approximately 2.7 kg/m^2 . The added FG can effectively regulate the electromagnetic parameters of the hybrid, resulting in the increasing of dielectric loss. With the increasing of FG content, the minimum reflection loss increased and moved to lower frequency. But when FG content increases excessively, it will reduce the absorption peak, indicating that the weight of adding FG has an optimum value.

References

[1] HongjingWu, L. Wang, S. Guo, Z. Shen, Double-layer structural design of dielectric ordered mesoporous carbonparaffin composites for microwave absorption, Applied Physics A Materials Science $ Processing, (2012) 439–446.

[2] S.-E. Lee, O. Choi, H.T. Hahn, Microwave properties of graphite nanoplatelet epoxy composites, JOURNAL OF APPLIED PHYSICS, 104 (2008).

[3] Z.M. Dang, Y. Shen, L.Z. Fan, N. Cai, C.W. Nan, S.J. Zhao, Dielectric properties of carbon fiber filled low-density polyethylene, Journal of Applied Physics, 93 (2003) 5543-5545.
[4] R.-B. Yang, W.-F. Liang, Microwave properties of high-aspect-ratio carbonyl iron epoxy absorbers, JOURNAL OF APPLIED PHYSICS 109 (2011) 07A311.
[5] L.d.C. Folgueras, M.A. Alves, M.C. Rezende, Microwave absorbing paints and sheets based on carbonyl iron and polyaniline measurement and simulation of their properties, Journal of Aerospace Technology and Management, 2 (2010) 63-70.
[6] P. Bhattacharya, C.K. Das, S.S. Kalra, Graphene and MWCNT Potential Candidate for Microwave Absorbing Materials, Journal of Materials Science Research, 1 (2012) 126-132.
[7] L.d.C. Folgueras, E.L. Nohar, R. Faez, M.C. Rezende, Dielectric microwave absorbing material processed by impregnation of carbon fiber fabric with polyaniline, Materials Research, 10 (2007) 95-99.
[8] Skolnik, M.I., Radar Handbook, New York: McGraw-Hill Book Company. (1970).
[9] Y.C. Qing, W.C. Zhou, S. Jia, F. Luo, D.M. Zhu, Electromagnetic and microwave absorption properties of carbonyl iron and carbon fiber filled epoxy silicone resin coatings, Appl Phys A Materials Science & Processing, 100 (2010) 1177–1181.
[10] M. Han-bing, C. Zhang, Z. Xue-quan, Dielectric Properties of Epoxy/Graphite Composite, Guangzhou Chemical Industry, 38 (2010) 49-51.
[11] W. Li-xi, Z. Jing, H. Xiao-gu, W. Hong-zhou, Z. Qi-tu, Microwave Absorbing Properties of Ferrite/Graphite Composite Coatings, Journal of Materials Science and Engineering, 29 (2011) 688～691.
[12] S.E. Bouazzaoui, M.E. Achour, C. Brosseau, Microwave effective permittivity of carbon black filled polymers Comparison of mixing law and effective medium equation predictions, JOURNAL OF APPLIED PHYSICS, 074105 (2011).
[13] B. WEN, J. ZHAO, Y. DUAN, Numerical analysis of effects of thickness on electromagnetic wave absorption properties of CPCCB paraffin wax composite, Applied Physics B Lasers and Optics, 83 (2006) 629-633.
[14] G.W. Flynn, Perspective The dawning of the age of graphene, THE JOURNAL OF CHEMICAL PHYSICS, 135 (2011).
[15] XIEWei, C. HaiFeng, C. ZengYong, C. ZhaoHui, Z.Y. iang, Radar Absorbing Properties of Light Radar AbsorbingMaterials Based on Hollow_porous Carbon Fibers, Journal of InorganicMaterials, 24 (2009) 320-324.
[16] M.E. Mezeme, S.E. Bouazzaoui, M.E. Achour, C. Brosseau, Uncovering the intrinsic permittivity of the carbonaceous phase in carbon black filled polymers from broadband dielectric relaxation, JOURNAL OF APPLIED PHYSICS, 109 (2011).
[17] V.A. Labunov, A.L. Danilyuk, A.L. Prudnikava, I. Komissarov, B.G. Shulitski, Microwave absorption in nanocomposite material of magnetically functionalized carbon nanotubes, JOURNAL OF APPLIED PHYSICS 112 (2012).
[18] Z. Han, D. Li, X.W. Wang, Z.D. Zhang, Microwave response of FeCo carbon nanotubes composites, JOURNAL OF APPLIED PHYSICS, 109 (2011).