

Electromagnetic Wave Absorption Properties of Coatings with Carbonyl-iron Particles coated by Silicon dioxide Nano-powders

^{1*}Xinwei Ji, ²Ming Lu, ³Feng Ye, ⁴Qian 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 – Silicon dioxide nano-powders prepared by gas-phase synthesis method were directly added into the solvent. The carbonyl iron particles (CIP) were partially coated by a thin layer of silicon dioxide in the solvent. Such coated particles were used as absorbent, with Aliphatic Polyurethane (APT) resin as matrix, to prepare the microwave absorbing coating. The morphology, elemental composition and electromagnetic characteristics of silicon dioxide coated and uncoated CIP were studied using SEM, EDS, and a vector network analyzer. Its electromagnetic parameters were measured in the range from 2GHz to 18GHz. The results show that the direct mixing method can effectively change the iron-based particle electromagnetic parameters, achieving the purpose of regulating the impedance matching. The minimum reflection loss of the coating shifts to the high-frequency region with increasing weight concentrations of silicon dioxide. The peak value of -31.1 dB was obtained at 16.5 GHz for the coatings with 50 wt.% CIP and 5wt.% silicon dioxide nano-powders.

Keywords –carbonyl iron particles; silicon dioxide by gas-phase; reflectivity; electromagnetic wave absorbing coating

1. Introduction

Radar Absorbing Materials are compounds that absorb incidental electromagnetic radiation in tuned frequencies and dissipate it as heat. Its preparation involves the adequate processing of polymeric matrices filled with compounds that act as radar absorbing centers in the microwave range[1]. The transition metals and their oxides have a wide variety of applications. The carbonyl iron particles (CIP) and its compounds are good candidates for the electromagnetic interface shielding[2]. An ideal absorber might comprise material with numerically equal values of complex permeability and permittivity and high loss tangents over a wide range of frequencies. The former ensures a perfect impedance match with air, thus enabling incident signals to enter the material without front-face reflection, and the latter promotes rapid attenuation afterwards. In general, the dielectric constant of absorbent is far greater than the permeability value. Various methods of reducing the dielectric constant

have been used to decrease the differences between absorbent dielectric constant and permeability. [3]prepared silica-coated Fe nanocomposites by the DC arc plasma in a mixed atmosphere of hydrogen and argon. The study of relative complex permittivity and permeability reveals that the excellent microwave absorption properties are the consequence of a proper EM match in microstructure. [4]used the sol-gel method to fabricate Fe crystalline powders coated with silicon dioxide, and giving the effect of the amount of the coating material silicon dioxide on electromagnetic parameters. [5]coated the carbonyl iron micro-particles by silicon dioxide nanoshell, using a sol-gel route with polyvinyl pyrrolidone as the surfactant. The results show that increasing the thickness of silicon dioxide nanoshell, the anti-oxidation property of the Fe/silicon dioxide core-shell composite particles is obviously improved while microwave permittivity decreased monotonously. [6]investigated the synthesis of core-shell particles covered with silica. The coating procedure was described and explained by calculating the potential energies of interaction between the temperature-

sensitive ferrite and silicon dioxide nanoparticles, according to Derjaguin-Landau-Verwey-Overbeck (DLVO) theory. [7] coated the magnetic metal fibers with silicon dioxide for surface modification by the sol-gel process to avoid forming an electrical conductive network due to surface connections. The electromagnetic parameters and absorption properties of silicon dioxide-coated metal fibers were improved greatly due to optimal impedance matching and the electric conductivity decreased, compared to those of uncoated materials.

However, these preparation methods are very difficult to large-scale application, this work attempts to mixed silicon dioxide by gas-phase synthesis method (fumed silicon dioxide) and carbonyl iron particles in the solvent mixture to adjust the electromagnetic parameters of the absorbent, which affect the reflectivity of fabricated coatings.

2. Experimental work

2.1 Materials and fabricating of specimens

CIP and fumed silicon coated CIP filled in aliphatic polyurethane resin were used to fabricate microwave-absorbing coatings. Commercial-grade fumed silicon used in this work was brought from Degussa Company. The CIP are spherical particles of around 2~5 μm in diameter. The specimens of YXE, YXE1 and YXE2 contained concentration of consistent CIP and 0 wt%, 2.5 wt%, 5 wt% fumed silicon dioxide, respectively. The fumed silicon was dispersed in the acetone solution by an ultrasonic bath at room temperature for 15 min. Subsequently, CIP were mixed together and the mixtures were stirred for 20 min at 500 rpm. Then the toluene, titanate coupling agent and dispersant had been added into the mixtures and stirred for 10 min at 1000 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 70°C for complete drying. The spraying and drying procedures were repeated for four times. The thickness of specimens prepared maintained to approximately 1.2 mm.

2.2 Characterization

The morphologies of CIP and fumed silicon coated CIP 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 60 wt% the absorbents 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 samples, reflection/absorption measurements in the frequency range of 2-18 GHz were carried out using the Naval Research Laboratory (NRL) arch method.

3. Result and discussion

3.1 Morphological Study

The microstructures of the fumed silicon coated/uncoated CIP were revealed by SEM images as shown in Fig.1.

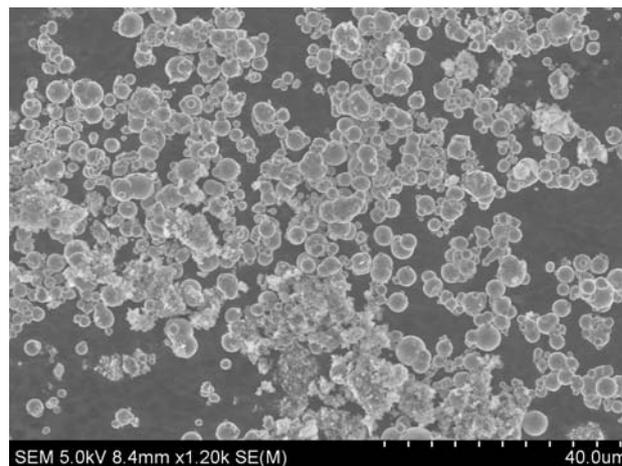


Figure 1. SEM images of fumed silicon coated and uncoated CIP

It can be seen that the CIP are spheroidal structure, and some of the iron particles were coated with silica thin layer. In fact the absorbent for the coating was a kind of mixture containing silicon-coated CIP and uncoated ones. The energy spectrum analysis results of the silicon coated CIP was shown in Table 1. The weight ratio of Si element is 11.97% and that of Fe is 44.40%.

Table 1. Chemical composition of silicon coated CIP

Chemical composition	wt.%	at.%
C	16.32	30.99
O	28.21	40.23
Si	11.97	9.73
Fe	44.40	19.05

In addition to the silicon layer on the surface of iron particles, there was dissociative fumed silicon in the mixture. As a result of fumed silica surface with large silicon hydroxyl, hydrogen bonds between these hydroxyl groups were formatted among the fumed silicon oxidation, when they were fully dispersed in a liquid system, then forming the silica network structure. The modification of fumed silica surface by titanate coupling agent can improve the binding force between polyurethane resin and fumed silica interface, which increase dispersion stability and compatibility of fumed silica in polyurethane resin. The polyurethane resin composite have more comprehensive better performance.

3.2 Complex permittivity and permeability

Complex permittivity and magnetic permeability 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. The real part and imaginary part of the complex permittivity of silicon coated and uncoated CIP used in the specimens are showed in Fig. 2. The real part of permittivity (ϵ') decreased with the increasing of frequency in the range 4GHz~18GHz. And with the silicon content increasing, the value decreases from around 20 to 17. The real part of permittivity of YXE2 almost kept constant with the frequency increasing, and the values of YXE1 decreased slightly with increasing frequency. The imaginary part of permittivity (ϵ'') fluctuates in the range of 4~18GHz.

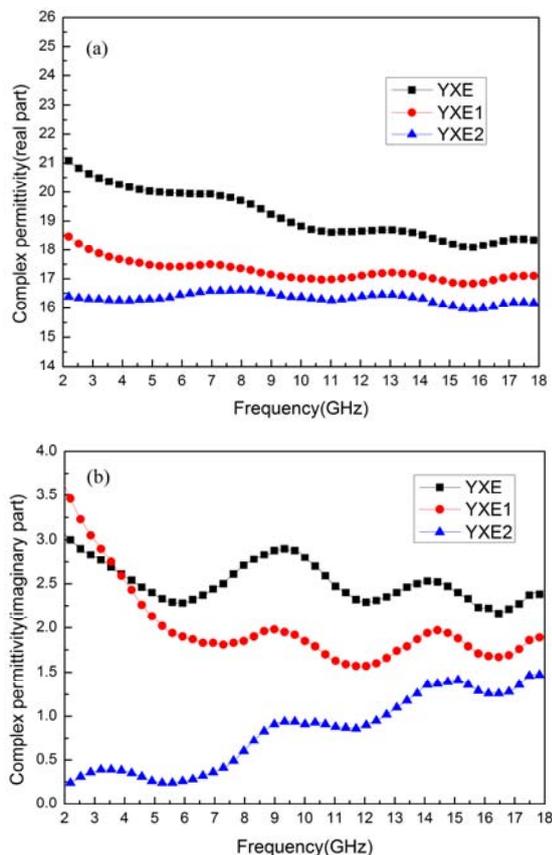


Figure 2. The real (a) and imaginary part (b) of complex permittivity of fumed silicon coated/uncoated CIP

The results show that the dielectric constant of spherical iron particles can decrease after the surface modification by fumed silicon. But the trend was different. The real part of permittivity decreased with the increasing of fumed silicon content. And they almost keep consistent with the increasing of frequency. When it comes to the imaginary part of the permittivity, with the addition of fumed silicon, the permittivity of YXE1 increased at the low frequency and then decreased with the frequency

increasing. This may be due to adding a small amount of silica plays a major role of uniformly dispersed about CIP, the opportunity of electromagnetic wave scattering in the mixture increases, resulting in dielectric loss increased at lower frequency.

When the content of fumed silicon continues to increase to 5wt.%, the imaginary part of permittivity of YXE2 decreased sharply at low frequency, then increased and fluctuated with the frequency increasing. With the increasing content of fumed silicon coated CIP, the formation possibility of conducting network decreased, resulting in reducing the conductivity of the mixture, thereby leading to the imaginary part of the dielectric constant decreased.

Silicon dioxide surface modified spherical iron particles can be used for the preparation of thin, lightweight microwave absorbing materials.

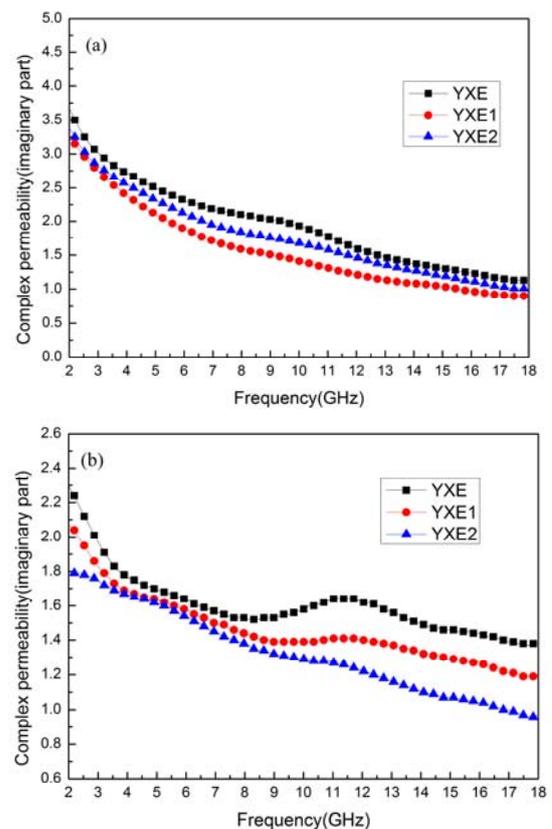


Figure 3. The real (a) and imaginary (b) part of complex permeability of fumed silicon uncoated/coated CIP

In general, compare to the unmodified sample, the real part of permittivity dropped by an average of about 3, and the imaginary part dropped by an average of about 2, while the corresponding complex permeability changes a little. By comparing with the decreasing of the complex permittivity, the complex permeability of the samples decreased slightly with the fumed silicon content increased.

Fig. 4 shows the dissipation factors represented by the dielectric ($tg\delta_e = \epsilon''/\epsilon'$) and magnetic loss ($tg\delta_m = \mu''/\mu'$) of the uncoated and coated CIP (adding different amounts of fumed silicon). The dielectric loss of the coated CIP is smaller than the uncoated ones. Furthermore, $tg\delta_e$ decreased largely and $tg\delta_m$ decreased

slightly with the increase of fumed silicon content. In particular, $tg\delta_e$ about YXE1 decreased and fluctuated in the range of 4-18GHz. While $tg\delta_e$ of YXE2 increased and fluctuated reaching maximum of 0.08 at the frequency 18GHz. These results indicate that with the increasing of silicon content in the mixture, the dielectric loss tangent decreased. But with the increasing of frequency, the trend of YXE1 and YXE2 were on the contrary. The dielectric loss tangent of YXE1 increased firstly and then decreased, fluctuating with the increasing of frequency. And that of YXE2 decreased sharply at the low frequency and then increased with the frequency increasing. The trend was the same as those of the imaginary of permittivity. At the same time, the magnetic loss tangent changes slightly with the increasing of fumed silicon content. It means that the fumed silicon has minor effect to the magnetic properties of CIP.

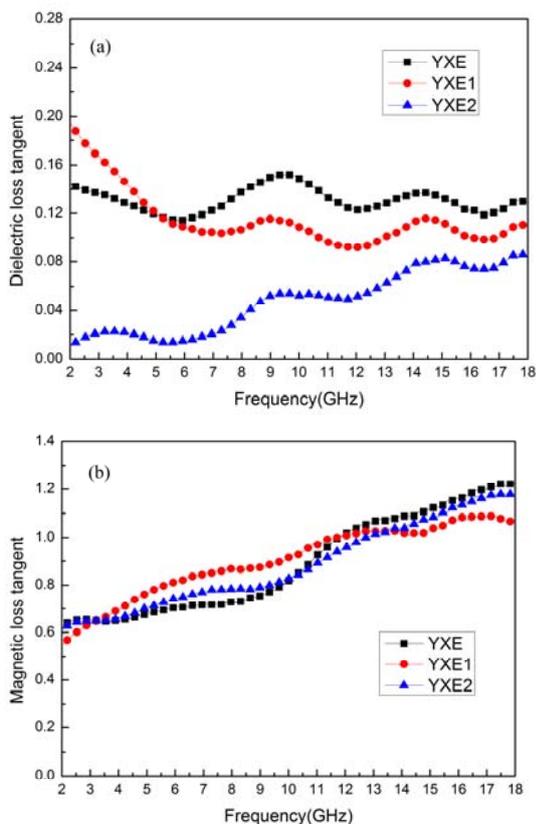
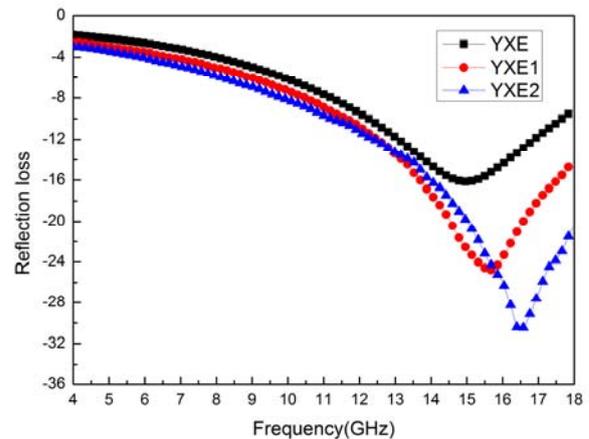


Figure 4. The dielectric (a) and magnetic (b) loss tangent of fumed silicon coated CIP

3.3 Microwave Absorbing Properties

The RL of the specimens were measured with NRL arch method and the results were shown in Fig.5. It expresses the relationship between reflection loss and frequency for the coatings with silicon coated and uncoated CIP. The specimens of YXE1 and YXE2 contained concentration of 2.5 wt% and 5 wt% fumed silicon powders, have RL values below -10dB in the range of 10.52~18GHz and 9.83~18GHz, respectively. The minimum RL value of specimen YXE2 reaches -30.5dB at the frequency 16.5GHz, and that of YXE with uncoated CIP reaches

16.14dB at 14.96GHz. The effective bandwidths (RL<-10dB) of YXE is 5.74GHz.



The microwave absorbing properties enhanced with increasing the addition of fumed silicon oxidation and a maximum reflection loss of -31.1 dB was obtained at 16.5 GHz with the thickness 1.2 mm. The result is due to the improving of the surface impedance matching, resulting from the decreasing of dielectric constant while keeping the permeability vary slightly at the same time, which is beneficial to the electromagnetic wave into the coatings.

The modification by fumed silicon improved the CIP electromagnetic parameters, adjusting the impedance matching. A variety of multiple loss mechanism of composite was obtained to get stronger absorption ability, due to the network structure between the fumed silicon in the polymer matrix material with micron CIP together. The more complicated composites may cause more electromagnetic scattering and multiple loss mechanisms, resulting in the higher absorbing ability about the absorbent.

4. Conclusion

Microwave absorbing coatings composed of CIP and fumed silicon coated CIP as absorbent and APT Resin as matrix were prepared with the method of spraying, with thickness been approximate 1.2 mm and the surface density been about 2.7 kg/m^2 . The electromagnetic and microwave absorbing properties of the coatings as function of frequency were investigated. The minimum reflection loss of the specimens with silicon coated CIP is -31.1 dB at 16.5 GHz.

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