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Electromagnetic and Microwave Absorption Properties of Carbonyl-Iron/$Fe_{91}Si_9$ Composites in Gigahertz Range

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ABSTRACT

Carbonyl-iron/$Fe_{91}Si_9$ composites for thin microwave absorbers were firstly prepared by a simple blending technique. The patterns of carbonyl-iron and $Fe_{91}Si_9$ were characterized by scanning electron microscope (SEM). The complex permittivity, permeability and microwave absorption properties of the composites were studied in the frequency range of 2 - 7GHz by a HP8720B vector network analyzer. Complex permittivity and permeability decrease gradually with increasing weight percentage of $Fe_{91}Si_9$ in the composites, the variation of permittivity was very large but the variation of permeability was very small. The composites exhibited excellent microwave absorption properties with increasing $Fe_{91}Si_9$ content. The reflection loss (RL) values less than –20 dB were obtained in the 3.7 - 6.7 GHz frequency range for the paraffin matrix composites with 80 wt% carbonyl-iron/$Fe_{91}Si_9$ powders (weight ratio of carbonyl-iron to $Fe_{91}Si_9$ was 1:1), with thickness of 4.0 - 2.4 mm, respectively. The optimal RL of –45 dB was observed at 5.2 GHz with a matching thickness (dm) of 3.0 mm. The excellent microwave absorption properties were attributed to a better electromagnetic impedance match and a higher electric resistivity.

Keywords: *Carbonyl-iron, $Fe_{91}Si_9$, Electromagnetic Properties, Microwave Absorption*

1. Introduction

In recent years, electromagnetic (EM) interference is worsening with the rapid development of wireless communications and circuit devices. EM-wave absorbers with wide absorbing band in gigahertz range have been developed to eliminate EM interference [1-4]. It is well known that the reflection and attenuation characteristics of EM-wave absorbers are associated with complex permeability ($\mu = \mu' - j\mu''$), permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) and EM impedance match [5]. Compared with ferrites, the metallic magnetic particles are more suitable as EM-wave absorbers because of their high saturation magnetization and high relative permeability at radar wave frequency. However, each kind of metallic magnetic particles has its intrinsic properties and usually can be used only in a special frequency range at given thickness. Since the single-component thin EM-wave absorbers cannot own simultaneously a combination of broad-band and strong microwave absorption in gigahertz range [6]. Multi-component absorbers are employed to obtain effective EM-wave absorbers. Several groups have reported good microwave absorption properties of α-Fe/Y_2O_3 [7], Fe/ZnO [8], Ni/polyaniline [9], Co/Al_2O_3 [10], Ni/C [11], Fe/SiO_2 [12], Co/C [13], etc. These composites all show better microwave absorption properties than corresponding single-component metallic magnetic absorbers. However, the permeability of these composites decreases due to the addition of non-magnetic components, which limits their microwave absorption properties [14].

$Fe_{91}Si_9$ as a kind of magnetic metallic material has relatively high resonance frequency, permeability and electric resistivity [15]. Meanwhile, carbonyl-iron as a cheap and popular magnetic metallic material has superior electric conductivity and large saturation magnetization but weak frequency dependence of complex permeability due to the eddy current loss induced by electromagnetic wave [16]. Therefore, merging the characteristics of these two kinds of materials could be attractive for microwave absorption materials which can produce the best possible results of the merging materials for the furthest attenuation microwave. In this paper, the composites employing commercial carbonyl-iron powders (CIP) and magnetic $Fe_{91}Si_9$ powders (FSP) with different weight ratios were firstly prepared by a simple blending technique. The complex permittivity, permeability and microwave absorption properties of the composites were discussed in the frequency range of 2 - 7 GHz.

2. Experimental

2.1. Materials

In our experiment, CIP was prepared via a conventional thermal decomposition process of iron pentacarbonyl and purchased from Shanxi Xinghua chemistry Co., Ltd. The main characteristics taken from product information are: the particles are spherically shaped with mean size of 4.8 μm and an apparent density of 2.08 g/cm^3. FSP was prepared by gas-atomization method and purchased from Changsha Hualiu Metallurgical Powder Co., Ltd. with the average diameter of 6.2 μm and an apparent density of 3.35 g/cm^3.

2.2. Preparation

CIP and FSP were uniformly mixed according to a given proportion. Subsequently, they were added into the molten paraffin. The mixtures were sufficiently mixed by constant stirring at 343 K for 30 min, and then poured into a coaxial cylindrical mold with 3.04 mm in inner diameter, 7.00 mm in outer diameter. After cooling to room temperature, the mixtures were cut into the sample with 2.00 mm in thickness. Three samples were prepared by varying the CIP and FSP ratios, the weight ratios of CIP to FSP were 9:1, 2.3:1, and 1:1, which were marked as CF1, CF2, and CF3, respectively. The content of CIP/FSP in the paraffin matrix were 80 wt% for all the samples. In addition, two pure CIP and FSP samples were prepared.

2.3. Measurement

The morphologies of the CIP and FSP were observed using scanning electron microscope (SEM). The EM parameters (complex permittivity and permeability) of the samples were measured by a HP8720B vector network analyzer in the 2 - 7 GHz range. According to transmit line theory [17], the reflection loss (RL) curves were calculated by the measured EM parameters and the thickness of absorbers.

3. Results and Discussion

3.1. Morphology

SEM images of raw commercial FSP and CIP are shown in **Figures 1(a)** and **(b)**.

The particles of FSP show perfect spherical shape with diameter varying from 1 to 20 μm. The particles of CIP are spherical shape like an onion bulb, and the diameters are in the range of 1 - 10 μm, all the particles show the low degree of agglomeration.

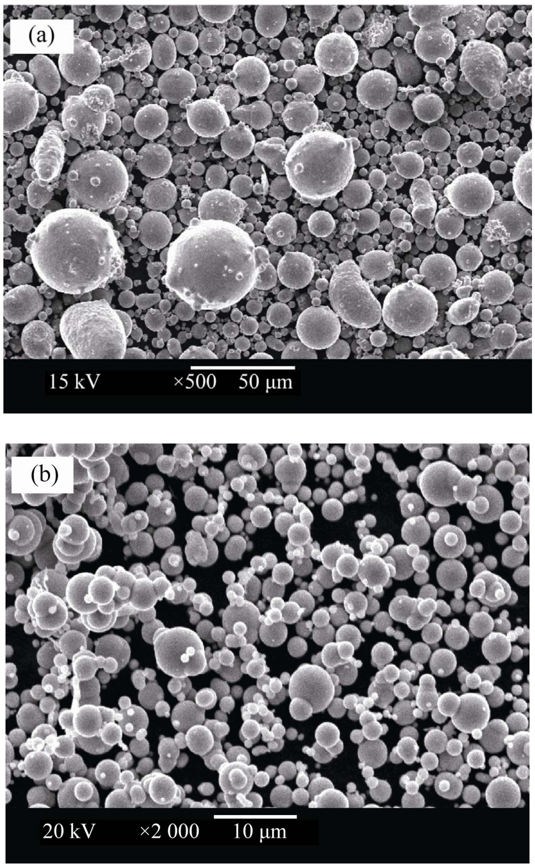


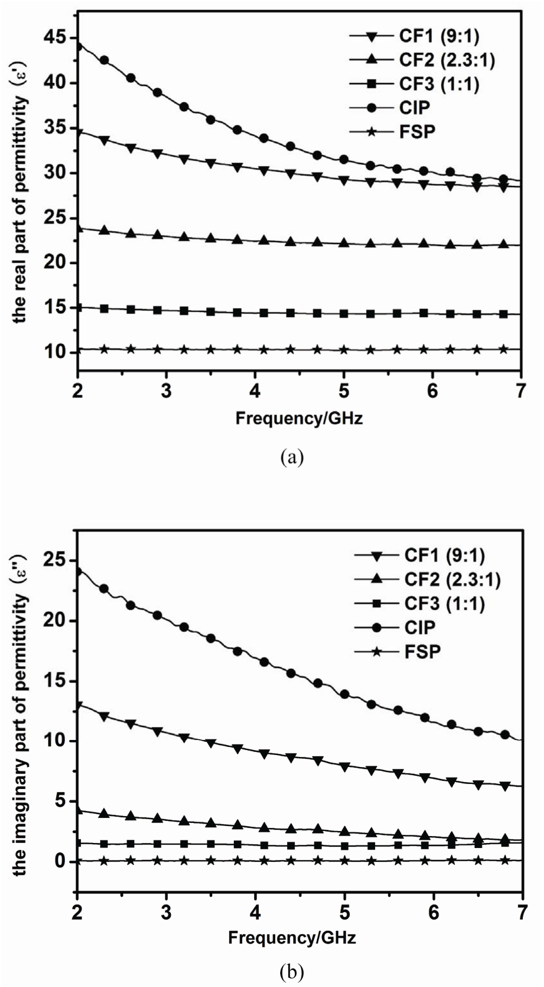
Figure 1. SEM images of raw commercial $Fe_{91}Si_9$ (a) and carbonyl-iron (b).

3.2. EM Characteristics

The frequency dependence on the relative permittivity of the CIP, FSP and composite samples CF1, CF2, and CF3 are shown in **Figure 2**.

It is found that the values of the complex permittivity are sensitive to the content of FSP addition. Both the values of ε' and ε'' decrease gradually with increasing weight percentage of FSP in the composites.

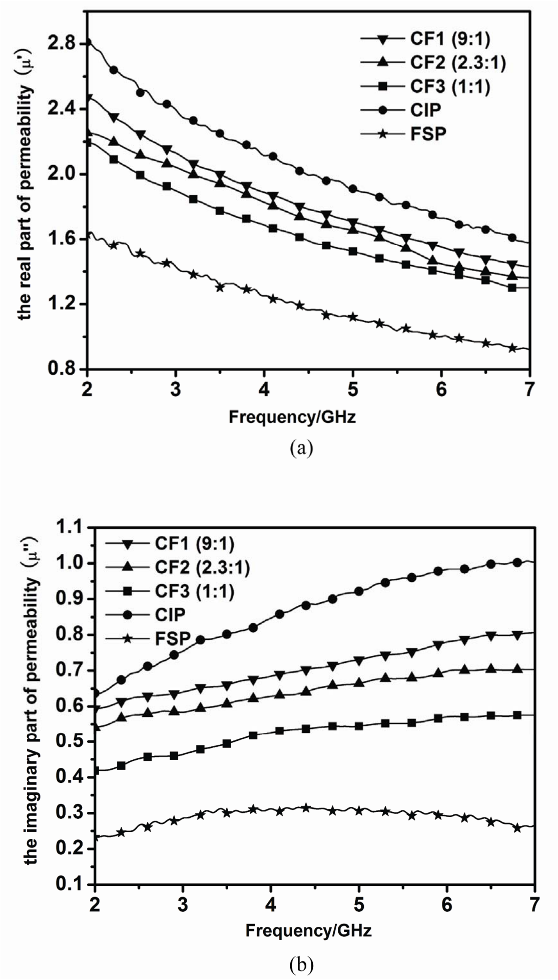
For the CF1 composites, the values of ε' and ε'' decline from 34.6, 13.1 to 28.5, 6.2 gradually with increasing frequency in the 2 - 7 GHz. For the CF2 composites, the values of ε' show less variation (ε' = ~23) and the values of ε'' decline from 4.3 to 1.7 between 2 and 7 GHz. The values of ε' and ε'' show almost constant over the whole measured frequency in the composites of CF3 (ε' = ~14.5 and ε'' = ~1.4). According to free-electron theory [18], $\varepsilon'' = \sigma/(2\pi f \varepsilon_0)$, where σ is the conductivity. It can be found clearly that the sample



(a)

(b)

Figure 2. Frequency dependence on the relative permittivity, the real part ε' (a) and the imaginary part ε'' (b) of the carbonyl-iron, $Fe_{91}Si_9$ and carbonyl-iron/$Fe_{91}Si_9$ paraffin matrix composites with different weight ratios of carbonyl-iron to $Fe_{91}Si_9$.



(a)

(b)

Figure 3. Frequency dependence on the relative permeability, the real part μ' (a) and the imaginary part μ'' (b) of the carbonyl-iron, $Fe_{91}Si_9$ and carbonyl-iron/$Fe_{91}Si_9$ paraffin matrix composites with different weight ratios of carbonyl-iron to $Fe_{91}Si_9$.

CF3 has higher resistivity than the samples CF1 and CF2.

The real part μ' and imaginary part μ'' of relative permeability of the CIP, FSP and composite samples CF1, CF2, and CF3 are plotted as a function of frequency in **Figure 3**.

It is obvious that μ' decreases gradually with increasing frequency, but μ'' shows an increasing trend with increasing frequency. It is found that μ' and μ'' exhibit a linear variation as a function of the increasing addition of FSP. The changes in μ' and μ'' with the frequency for the composites obey the Lichitenecker's logarithm mixed law [2],

$$\log\mu = \delta_1\log\mu_1 + \delta_2\log\mu_2 \quad (1)$$

where δ_1 and δ_2 are the normalized volume ratios of CIP and FSP, respectively. μ, μ_1 and μ_2 are the complex permeability for the composite samples, CIP and FSP, respectively. Compared with CIP, FSP has lower μ' and μ'', so the addition of FSP will result in the decrease of relative permeability.

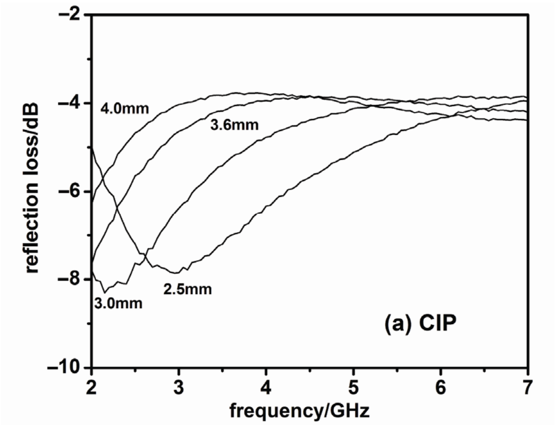
3.3. Microwave Absorption Properties

The RL of electromagnetic radiation under normal incidence of the electromagnetic filed on the surface of a single-layer material backed with a perfect conductor can be defined by [19]:

$$RL = 20\lg\left|\frac{Z-1}{Z+1}\right| \quad (2)$$

where, Z is the normalized impedance between input impedance Z_{in} of the single-layered absorber and the impedance of free space Z_0, and is expressed as $Z = \frac{Z_{in}}{Z_0} = \sqrt{\frac{\mu}{\varepsilon}}\tanh\left(j\frac{2\pi d}{\lambda_0}\cdot\sqrt{\mu\varepsilon}\right)$ here, μ and ε represent the relative complex permittivity and the permeability of the composite medium, respectively, d is the thickness of an absorber, and λ_0 is the wavelength of the incident wave in free space. Thus, the RL of an absorber is a function of six characteristic parameters, viz., ε', ε'', μ', μ'', f and d. The microwave absorption properties of the composites have been calculated from a computer simulation using (2) for various measured values of ε', ε'', μ', μ'' previously obtained. The calculated RL as a function of frequency for the samples with different thickness are shown in **Figure 4**.

Figure 4 shows a typical relationship between RL and frequency for the paraffin matrix composites with 80 wt% CIP/FSP. Generally, "RL < –20 dB" is considered as adequate microwave absorption, as the RL value of –20 dB is comparable to 99% of microwave absorption according to (2). It is seen that the RL of carbonyl-iron and $Fe_{91}Si_9$ is very low and the peak values more than –20 dB in the frequency range of 2 - 7 GHz. The CF1 sample exhibits poor microwave absorption performance with the RL values more than –20 dB from 2 to 7 GHz, which may be due to the low EM impedance match between the high permittivity and relative low permeability. However, with increasing FSP content, microwave absorption is evidently improved. The RL values of the CF2 sample are less than –20 dB in the range of 2.0 - 3.5 GHz over absorbers thickness of 5.2 - 3.0 mm, and the optimal RL value is –33 dB at the frequency of 3 GHz with a matching thickness (d_m) of 3.6 mm. The CF3 sample provides the best microwave absorption performances. The RL values of the CF3 sample less than –20 dB are obtained in the 3.7 - 6.7 GHz frequency range, with thickness of 4.0 - 2.4 mm, respectively. In particular, the optimal RL value of –45 dB is observed at 5.2 GHz with d_m of 3.0 mm. In order to make the results more clearly, the elec-



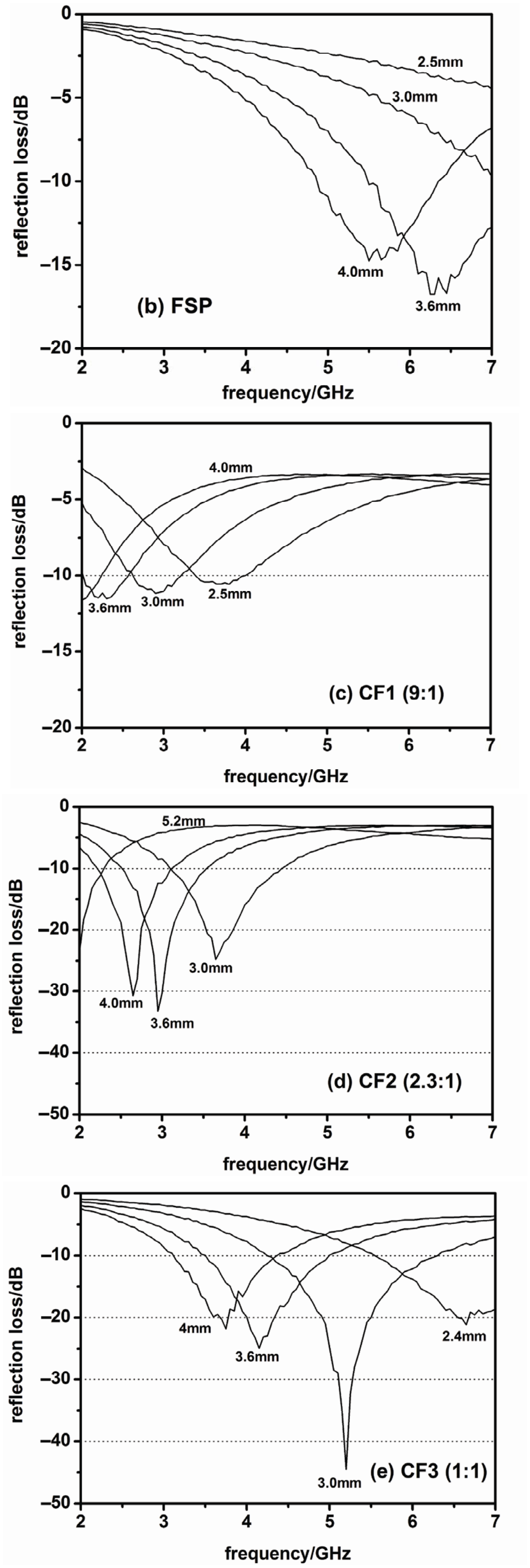


Figure 4. Frequency dependences of reflection loss values for the carbonyl-iron, $Fe_{91}Si_9$ and carbonyl-iron/$Fe_{91}Si_9$ paraffin matrix composites with different weight ratios of carbonyl-iron to $Fe_{91}Si_9$.

Table 1. Electromagnetic wave absorption properties of the carbonyl-iron/Fe$_{91}$Si$_9$ paraffin matrix composites with different weight ratios of carbonyl-iron to Fe$_{91}$Si$_9$.

Sample (80 wt%)	Frequency (GHz) (RL < −20 dB)	Thickness (mm) (RL < −20 dB)	Optimal RL value (dB)	fm (GHz) optimal RL	dm (mm) optimal RL
CF1（9:1）	……	……	12	2	4
CF2（2.3:1）	2.0 - 3.5	3.0 - 5.2	33	3	3.6
CF3（1:1）	3.7 - 6.7	2.4 - 4.0	45	5.2	3

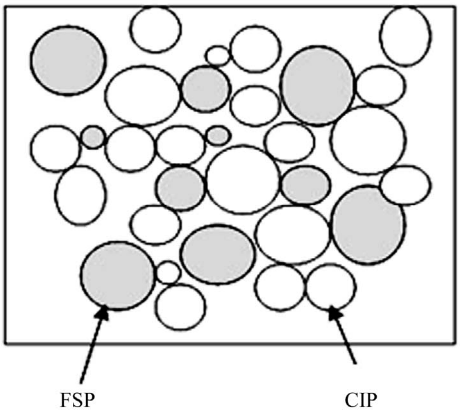


Figure 5. Distribution of Fe$_{91}$Si$_9$ and carbonyl-iron powders in paraffin matrix.

tromagnetic wave absorption properties of the composite samples CF1, CF2, and CF3 prepared under the optimized conditions are summarized in **Table 1**.

It is clear that the frequency band of RL < –20 dB gets broader and the thickness gets thinner with increasing FSP content in the frequency range of 2 - 7 GHz. The improvement in microwave absorption of the composites with the addition of FSP is suggested to originate from the efficient combination of CIP and FSP. Generally, excellent EM-wave absorption results from efficient complementarities between the relative permittivity and permeability in materials. Either only the magnetic loss or only the dielectric loss may result in weak EM-wave absorption properties due to the imbalance of the EM impedance match [11]. The introduction of FSP to CIP weakens the dielectric loss, but has not weakened the magnetic loss too much. Thus, a better EM impedance match could be established due to the combination of the reduced dielectric loss and nearly invariable magnetic loss, resulting in the enhanced microwave absorption [6].

Figure 5 shows the distribution of FSP and CIP in the paraffin matrix. the FSP acts not only as a magnetic material, increasing the permeability of the composites powder, but also as an insulating matrix distributed among the gaps between carbonyl-iron particles, which could reduce the eddy current loss through increasing electric resistivity as an important reason bringing about the excellent microwave absorption [20].

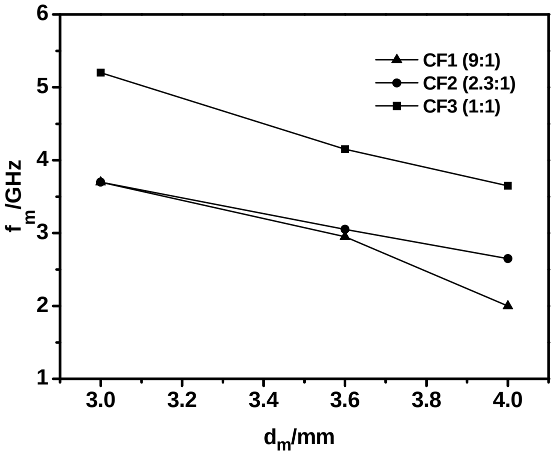


Figure 6. Relationship between matching frequency and matching thickness for carbonyl-iron/Fe$_{91}$Si$_9$ paraffin matrix composites with different weight ratios of carbonyl-iron to Fe$_{91}$Si$_9$.

The d_m and matching frequency (f_m) for minimum reflection with thickness of 3.0 mm, 3.6 mm, and 4.0 mm, respectively, are indentified in case of the composite samples CF1, CF2, CF3 and their relationship is shown in **Figure 6**.

It is obviously that the matching frequency decreases with the increase of absorber thickness, but increases with increasing FSP content. This phenomenon is in consistent with (3) [21],

$$d_m = \frac{c}{2\pi f_m \mu''} \qquad (3)$$

where c is the velocity of light. The d_m become thinner quickly as a result of the phenomenon that μ'' shows an increasing trend with increasing frequency. It is significantly that the composite samples are very propitious to use in the 2 - 7 GHz frequency as one thin absorber.

4. Conclusions

In conclusion, the addition of FSP has remarkable effect on the complex permittivity, permeability and microwave absorption properties of the composites. Complex permittivity decreases quickly but permeability decreases slowly with increasing weight percentage of FSP in the composites. As a result of a better EM impedance match and a higher electric resistivity of FSP, the paraffin matrix composites with 80 wt% CIP/FSP exhibit excellent microwave absorption properties in the frequency range from 2 to 7 GHz. The RL values of the CF3 sample less than –20 dB are obtained in the 3.7 - 6.7GHz frequency range, with thickness of 4.0 - 2.4 mm, respectively. The optimal reflection loss reaches –45 dB is observed at 5.2 GHz with d_m of 3.0 mm. Therefore, the sample of CF3 is promising microwave absorber.

5. Acknowledgements

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