

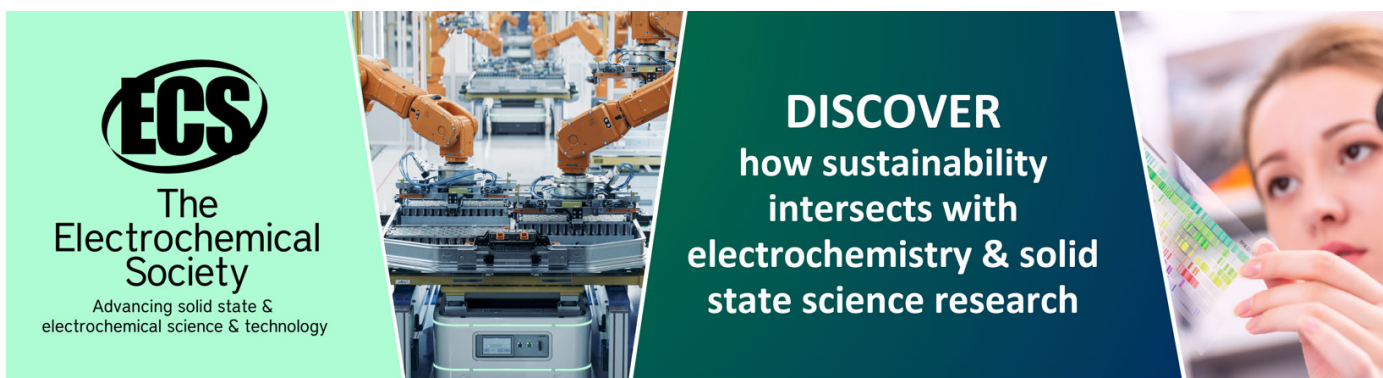
Complex permittivity, permeability and electromagnetic wave absorption of α -Fe/C(amorphous) and Fe₂B/C(amorphous) nanocomposites

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Complex permittivity, permeability and electromagnetic wave absorption of α -Fe/C(amorphous) and Fe₂B/C(amorphous) nanocomposites

Jiu Rong Liu¹, Masahiro Itoh¹, Takashi Horikawa¹,
Masaru Itakura², Noriyuki Kuwano² and Ken-ichi Machida¹

¹ Collaborative Research Center for Advanced Science and Technology, Osaka University,
2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

² Department of Materials Science and Technology, Graduate School of Engineering
Sciences, Kyushu University, 6-1 Kasuga-Kohen, Kasuga, Fukuoka 816, Japan

E-mail: machida@rcast.osaka-u.ac.jp

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Abstract

The complex permittivity, permeability and electromagnetic (EM) wave absorption properties have been investigated for resin compacts containing 75 wt% composite powders of α -Fe/C(a) and Fe₂B/C(a). The real and imaginary parts (ϵ'_r and ϵ''_r) of relative permittivity for the resin composites remained almost constant in the 0.05–20.05 GHz range. The imaginary part of the relative permeability (μ''_r) exhibited a wide peak in the 1–9 GHz range for α -Fe/C(a) and in the 2–18 GHz range for Fe₂B/C(a). Both resin composites exhibited good EM wave absorption properties (RL < –20 dB) in the 4.4–8.3 GHz range with an absorber thickness of 1.9–3.4 mm, and in the 7.5–16 GHz range for an absorber thickness of 1.2–2.2 mm, respectively.

1. Introduction

Recently, there has been an increase in the number of applications of electromagnetic (EM) waves in the G-band (5.6–8.2 GHz) and X-band (8.2–12.4 GHz) for synthetic aperture radar and satellite broadcast systems and consequently much attention has been devoted to the study of EM wave absorbing materials in anti-EM interference coatings and self-concealing technology. Among the candidates for EM wave absorbers, soft metallic magnets are particularly interesting. Since metallic magnetic materials have large saturation magnetization values and their Snoek's limit is at a high frequency level, the values of the relative complex permeability (μ_r) can still remain high in the high frequency range [1–3]. Therefore, it is possible to make thin absorbers from metallic magnets. However, there is a serious problem when metallic magnetic materials are used as absorbers. The electrical

conductivity of these materials is high and the high frequency permeability decreases due to the eddy current loss induced by the EM wave. It is necessary to use particles smaller than the skin-depth for suppressing the eddy current loss in order to produce EM wave absorbers from such materials. Fine powders of sendust or carbonyl iron are now available as commercial products, but they can only be used as EM wave absorbers in the low GHz range [3–5].

Sugimoto *et al* [6, 7] have found that the α -Fe/SmO composites derived from arc-melted Sm₂Fe₁₇ ingots by hydrogen disproportionation and subsequent oxidation treatment exhibit good EM wave absorption properties in the 0.73–1.3 GHz range. We have also reported that the nanocomposites of α -Fe/Y₂O₃, α -Fe/Fe₃B/Y₂O₃, ϵ -Fe₃N/Y₂O₃, etc as prepared by the melt-spinning technique show good EM wave absorption properties in different frequency ranges [8–10]. However, there are several hurdles

in employing the above method for fabricating EM wave absorption materials: the preparation process is complicated, expensive rare-earth metals are used, and this method cannot be used for other magnets as EM wave absorbers because of the complicated hydrogen disproportionation and oxidation treatments. For these reasons, novel composites prepared using an alternative mechanical grinding process are taken to be the EM wave absorption materials in this study [11–13]. In the novel composites, amorphous carbon C(a) derived from charcoal powders was used to replace rare-earth oxides as the separator for suppressing the eddy current loss in the mechanical grinding technique. The reasons for selecting C(a) as a separator are as follows: the measured electric resistivity of C(a) is about $10^4 \Omega \text{ m}$ as reported by Mochidzuki *et al* [14], which is larger than that of metallic magnets ($\sim 10^{-6}$ – $10^{-8} \Omega \text{ m}$) [15]. Although this value is smaller than those of insulators, it is expected to be effective in suppressing the eddy current loss because the electrical resistivity of α -Fe/SmO is about $100 \Omega \text{ m}$ [6]; carbon is very light. Therefore, the novel composites can still retain a high magnetic material content, which results in a large μ_r'' value. Thin absorbers could be produced from the novel composite materials. Here, the EM wave absorption properties of α -Fe/C(a) and $\text{Fe}_2\text{B}/\text{C(a)}$ composite materials are investigated in the G-band and X-band.

2. Experimental procedure

Ingots of the interstitial-type compound Fe_2B were first prepared from Fe and B (>99.9% in purity) by means of induction melting in Ar. Fe_2B ribbons of $50 \mu\text{m}$ thickness were produced from the ingots of Fe_2B using a melt-spun apparatus. After dry ball milling in Ar, Fe_2B powder with particle sizes of $2\text{--}4 \mu\text{m}$ was obtained. α -Fe/C(a) and $\text{Fe}_2\text{B}/\text{C(a)}$ composites were prepared by ball-milling the mixture of α -Fe ($3\text{--}10 \mu\text{m}$) and Fe_2B with 6 wt% C(a) powder at a rotation speed 3.3 r s^{-1} for 30 h in Ar, respectively. The resultant powders were characterized by x-ray diffraction (XRD), and the microstructures were analysed using scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Epoxy resin composites were prepared by homogeneously mixing epoxy resin with 75 wt% composite powder and pressing into cylindrical compacts. These compacts were cured by heating at 453 K for 30 min, and then they were cut into toroidal samples of 7.00 mm outer diameter and 3.04 mm inner diameter. The scattering parameters (S_{11} , S_{21}) of the toroidal samples were measured using a Hewlett-packard 8720E network analyser. The relative permeability (μ_r) and permittivity (ϵ_r) values were determined from the scattering parameters as measured in the $0.05\text{--}20.05 \text{ GHz}$ range. The reflection loss (RL) curves were calculated from the relative permeability and permittivity at the given frequency and absorber thickness, according to the following equations:

$$Z_{\text{in}} = Z_0 \left(\frac{\mu_r}{\epsilon_r} \right)^{1/2} \tanh \left\{ j \left(\frac{2\pi f d}{c} \right) (\mu_r \epsilon_r)^{1/2} \right\}, \quad (1)$$

$$\text{RL} = 20 \log \left| \frac{Z_{\text{in}} - Z_0}{Z_{\text{in}} + Z_0} \right|, \quad (2)$$

where f is the frequency of the EM wave, d the thickness of the absorber, c the velocity of light, Z_0 the impedance of air and Z_{in} is the input impedance of the absorber. The attenuation of the EM wave and reflectance of the absorber at the surface were determined from the frequency dependence of the RL.

3. Results and discussion

Figure 1 shows the typical XRD patterns measured on the α -Fe/C(a) and $\text{Fe}_2\text{B}/\text{C(a)}$ powders. From figure 1(b), it was found that a single phase of the Fe_2B compound is formed using the melt-spun technique, as suggested by Coene *et al* [16]. The $\text{Fe}_2\text{B}/\text{C(a)}$ composites were prepared after milling with C(a) powder and Fe_2B exhibited a wider line broadening of the XRD peaks; furthermore, there was no shift in the diffraction angles for the XRD peaks of Fe_2B . The mean crystalline sizes of the Fe_2B particles were evaluated to be about 60 nm (as-obtained) and 30 nm (after ball-milling) from the line broadening of the XRD peaks, using Scherrer's formula. The composite particles were found to be of roughly spherical shape in the SEM and TEM photographs of $\text{Fe}_2\text{B}/\text{C(a)}$ powders and had a wide size distribution from a hundred to several hundreds of nanometres (figure 2(c)), but the grain size of Fe_2B varied from 20 to 50 nm (see figure 2(d)). From the above results, we can see that the original particles of Fe_2B were substantially dispersed into finer ones after ball-milling with C(a) powder; C(a) was only embedded among the Fe_2B grains, but not incorporated into the crystal lattice of Fe_2B during the ball-milling process. The same results as above were also obtained on the α -Fe/C(a) nanocomposites, as shown in figure 1(a). As observed on the

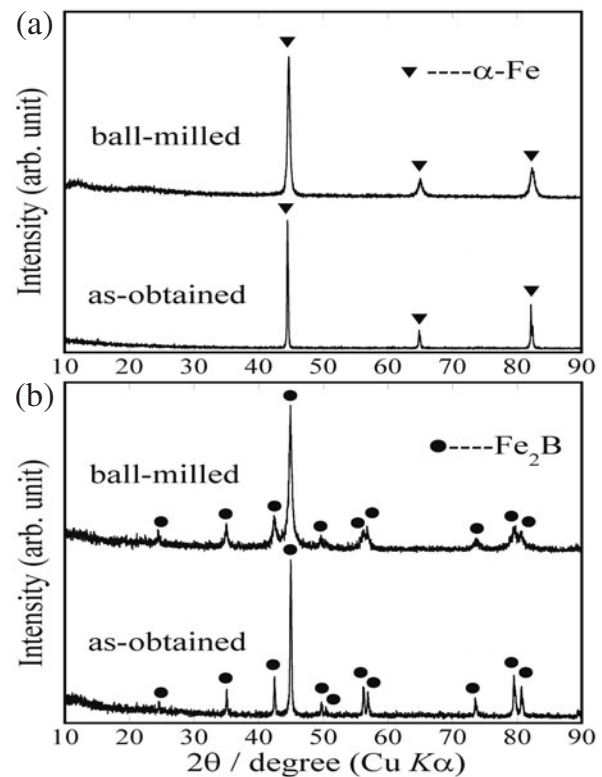


Figure 1. XRD patterns of the α -Fe (a) and Fe_2B (b) composites after ball-milling with amorphous carbon powder at a rotation speed 3.3 r s^{-1} for 30 h, together with as-obtained ones.

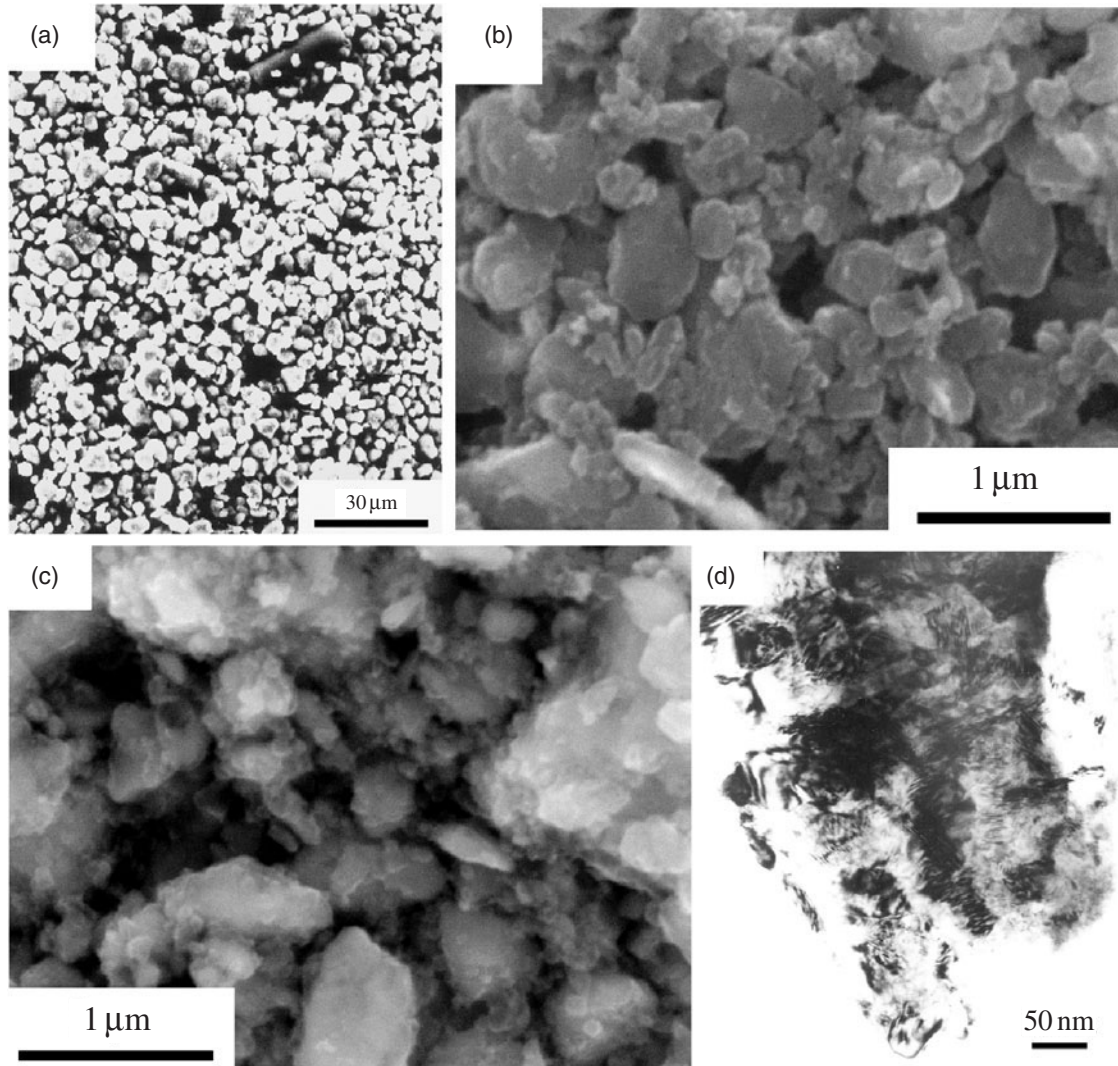


Figure 2. SEM images of (a) α -Fe, (b) α -Fe/C(a), (c) $\text{Fe}_2\text{B/C(a)}$ powders and (d) TEM micrograph of $\text{Fe}_2\text{B/C(a)}$ nanocomposites.

SEM photographs of α -Fe and α -Fe/C(a) powders (figures 2(a) and (b)), the α -Fe particle size varied from 3 to 10 μm , but the α -Fe/C(a) composite particles showed a wide size distribution from 100 nm to 1 μm . This observation shows that the particles of α -Fe were substantially dispersed into finer ones after ball-milling with C(a) powder.

Figure 3(a) shows that the real part ϵ'_r and imaginary part ϵ''_r of the relative permittivity for the resin composites with 75 wt% α -Fe/C(a) powder were almost constant between 2 and 14 GHz, for which the relative permittivity ($\epsilon_r = \epsilon'_r - j\epsilon''_r$) showed less variation ($\epsilon'_r = \sim 12$ and $\epsilon''_r = \sim 0.4$). For the $\text{Fe}_2\text{B/C(a)}$ resin composites, the ϵ'_r and ϵ''_r values were almost constant in the 2–16 GHz range ($\epsilon'_r = \sim 12.2$ and $\epsilon''_r = \sim 0.7$), but ϵ'_r increased slightly for frequencies below 2 and above 16 GHz. This could be because some oscillation occurred in the ϵ'_r curve measurement at low and high frequencies close to the limits of the equipment. The electric resistivity of α -Fe/C(a) and $\text{Fe}_2\text{B/C(a)}$ composites was measured to be about 100 Ωm . The high resistivity is attributed to the microstructures of α -Fe/C(a) and $\text{Fe}_2\text{B/C(a)}$ nanocomposites, in which C(a) plays the role of a separator embedded among α -Fe and Fe_2B grains.

The real part of the relative permeability (μ'_r) declined from 3.8 to 0.80 with frequency for α -Fe/C(a) resin composites, as shown in figure 3(b); furthermore, the imaginary part of the relative permeability (μ''_r) increased from 0.2 to 0.98 in the 0.05–6.0 GHz range, and then decreased in the higher frequency range. For the $\text{Fe}_2\text{B/C(a)}$ resin composites, μ'_r increased from 0.1 to 0.63 over a range of 0.5–12 GHz, and then decreased. Consequently, the μ''_r curve exhibited a peak in a broad frequency range (2–18 GHz). Compared with α -Fe/C(a), the $\text{Fe}_2\text{B/C(a)}$ composites showed lower maximum values for both μ'_r and μ''_r as shown in figure 3(b). These lowered values are due to the smaller magnetization of Fe_2B compared to α -Fe. However, the maximum point of the μ''_r curve for the $\text{Fe}_2\text{B/C(a)}$ composites shifted to the higher frequency (~ 12 GHz).

Figure 4(a) shows the typical relationship between RL and frequency for the resin composites with 75 wt% α -Fe/C(a) powder. RL values of resin composites less than -20 dB were obtained in the 4.4–8.3 GHz range with an absorber thickness of 1.9–3.4 mm, and a minimum RL value of -58 dB was observed at 5.9 GHz with a thickness (d_m) of 2.5 mm. For the resin composites with 75 wt% $\text{Fe}_2\text{B/C(a)}$ powder, the

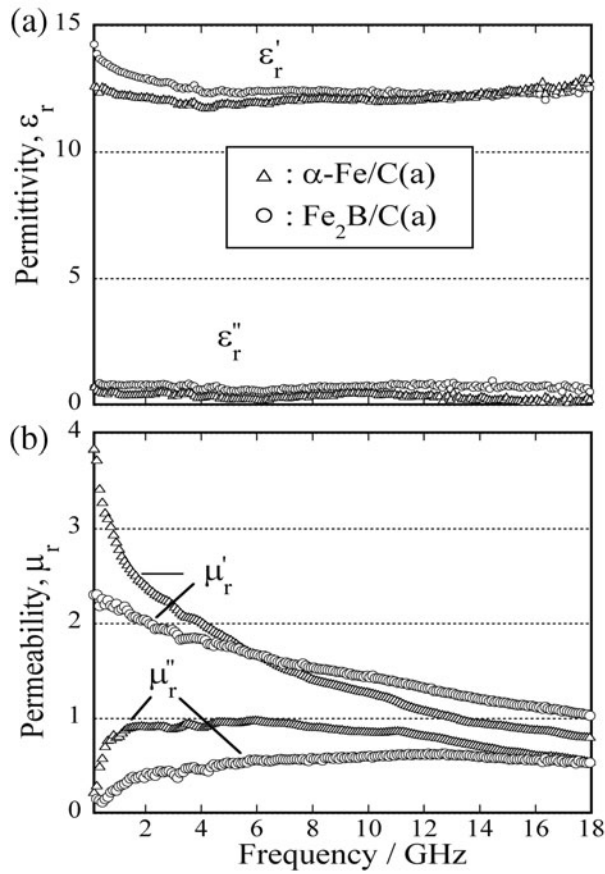


Figure 3. The relative permittivity ϵ_r (a) and permeability μ_r (b) plotted against frequency for α -Fe/C(a) and $\text{Fe}_2\text{B}/\text{C(a)}$ resin composites with 75 wt% powders.

frequency dependence of RL values of less than -20 dB was recorded in the 7.5–16 GHz range with absorber thickness ranging from 1.2 to 2.2 mm. In particular, a minimum RL of -60 dB was obtained at 10.7 GHz as shown in figure 4(b). Compared with ferrites, the thicknesses (d_m) of $\text{Fe}_2\text{B}/\text{C(a)}$ absorbers decreased by about 30–50% in the same frequency region [17–19]. At the same time, we also investigated the EM wave absorption properties of the resin composites with 75% α -Fe powders (3–10 μm) in the 0.05–20.05 GHz range (figure 4(c)). However, the α -Fe resin composites only gave a weak EM absorption compared with α -Fe/C(a) ones. This result indicates that the C(a) addition plays an important role suppressing the eddy current loss when using α -Fe/C(a) as the EM wave absorber.

One criterion for selecting suitable magnetic EM wave absorption materials is the location of its resonance frequency (f_r), which should be matched with the frequency of the EM wave used. The f_r value is generally proportional to the magnetic anisotropy field H_A of the material used according to ferromagnetic resonance theory [20]. Compared with the α -Fe/ Y_2O_3 composites [8], the minimum reflection point of α -Fe/C(a) shifted from 2.6 to 5.9 GHz, and moreover, the maximum value of f_m (RL < -20 dB) was located at 8.3 GHz. These peculiar frequency characteristics can be understood as follows: for α -Fe/ Y_2O_3 nanocomposites, the sphere-like grains of α -Fe (~ 20 nm) were derived from the intermetallic compound Y_2Fe_{17} by hydrogen disproportionation and

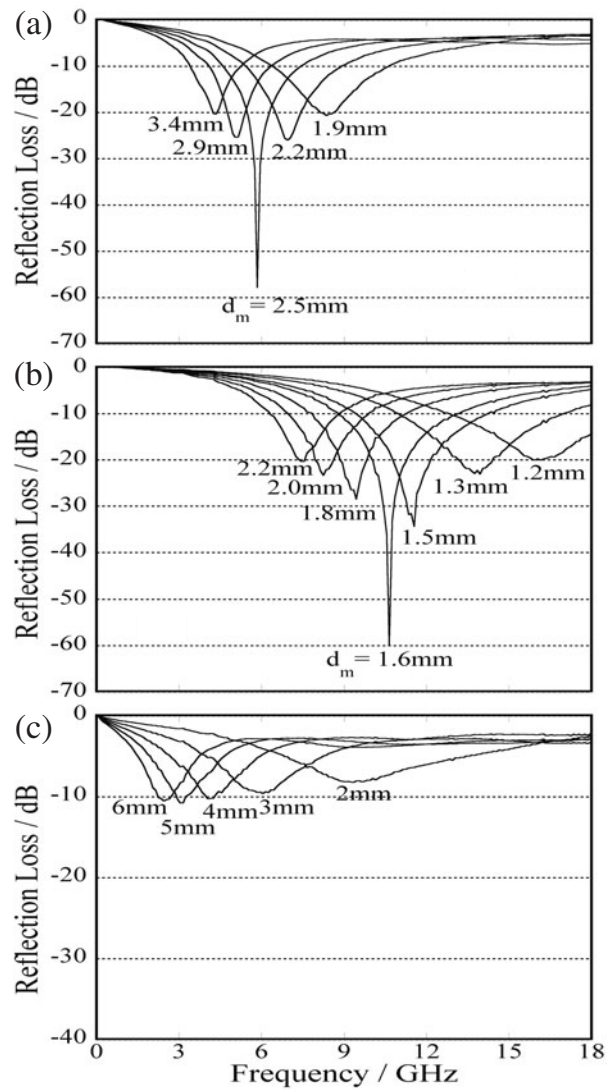


Figure 4. Frequency dependences of the RLs for the resin composites with 75 wt% (a) α -Fe/C(a), (b) $\text{Fe}_2\text{B}/\text{C(a)}$ and (c) α -Fe powders at different thicknesses.

oxidation treatments, and hence the f_r value (~ 1.6 GHz) is ascribed to the relatively small intrinsic anisotropy of α -Fe [6]. In this study, the α -Fe particles (3–10 μm) became successively finer and a regular flake-like morphology was obtained after ball-milling with C(a) powder, as suggested by Rochman *et al* [13] and Deng *et al* [21]. The flake-like α -Fe particles exhibited a large shape anisotropy field, which resulted in the increase in f_r , for the α -Fe/C(a) nanocomposites. A comparison of the sample $\text{Fe}_2\text{B}/\text{C(a)}$ with α -Fe/C(a) showed that the minimum reflection point shifted to a higher frequency—from 5.9 to 10.7 GHz. This shift is attributed to the fact that Fe_2B has a larger f_r value (~ 11.4 GHz) than α -Fe (~ 1.6 GHz), as evaluated from the expression $2\pi f_r = \gamma H_A$, where γ is the gyrometric ratio and H_A is the anisotropic field, because of its high anisotropy field (~ 0.32 MA m^{-1}) [16]. From the μ_r'' curves shown in figure 2(b), we can also observe that the maximum μ_r'' value of $\text{Fe}_2\text{B}/\text{C(a)}$ shifts to the higher frequency (~ 12 GHz). As a result, the effective EM wave absorption (RL < -20 dB)

shifted to a higher frequency range compared to that of α -Fe/C(a) resin composites.

4. Conclusion

The α -Fe/C(a) and Fe₂B/C(a) nanocomposites are prepared by ball-milling the α -Fe and Fe₂B with C(a) powder, respectively, where C(a) plays the double role of the milling medium and the separator for suppressing the eddy current loss. Compared with α -Fe/C(a), the Fe₂B/C(a) resin composites have good EM wave absorption characteristics in the higher frequency range due to the H_A value of Fe₂B being larger than that of α -Fe. This work suggests that the α -Fe/C(a) and Fe₂B/C(a) composites can be used as good EM wave absorption materials in the G band (5.6–8.2 GHz) and X band (8.2–12.4 GHz), respectively.

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