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Electromagnetic wave absorption properties of nanocomposite powders derived from intermetallic compounds and amorphous carbon

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Abstract

Nanocomposite materials consisting of Y_2Fe_{17} and amorphous carbon (a-C) were prepared by a mechanical grinding method. Mean crystalline size of the Y_2Fe_{17} particles in the nanocomposite materials was evaluated to be 20 nm from the line broadening of diffraction peaks. Complex permittivity, permeability and electromagnetic wave absorption properties of resin compact containing 75 wt.% Y_2Fe_{17}/a-C powders showed the good electromagnetic wave absorption properties of the minimum reflection loss (RL) value of – 48 dB at 17 GHz with a matching thickness of a 1.3 mm because the eddy current loss was effectively suppressed by increasing the electric resistance for Y_2Fe_{17} particle with hybridization with a-C as an insulator.

Keywords: Electromagnetic wave absorption; GHz range; Magnetic anisotropy; Nanocomposite; Mechanical grinding

1. Introduction

With the development of communication devices using the electromagnetic waves of GHz range, such as mobile phones (0.8–1.5 GHz), LAN systems (2.45, 5.0, 19.0, 22.0, 60.0 GHz) and satellite broadcast systems (11.7–12.0 GHz), the serious electromagnetic interference problems have been emerged. The electromagnetic wave absorbers have been widely investigated to eliminate the above problems. The electromagnetic absorption materials can be classified into three types by their absorption mechanism: dielectric loss, conductive loss and magnetic loss. Among them, the magnetic-type absorbers provide a kind of interesting features according to their complex permeability (μ'–$j\mu''$) and permittivity (ε'–$j\varepsilon''$). Metal oxides, e.g. NiZn- and MnZn-ferrites are used as the electromagnetic absorption media in the absorbers against MHz ranges [1,2]. Such magnetic materials are not able to apply in the GHz range of electromagnetic waves according to the sharp decrease of magnetic loss (μ'') due to their Snoek's limit. On the other hand, the metallic magnetic materials, such as Fe and Co, are one of the suitable candidates for using thin electromagnetic wave absorbers for GHz range, as they still possess the high magnetization values even in a high frequency range and their Snoek's exist at a high frequency side [3–5]. However, some defects are remained in the metallic magnetic materials for using the electromagnetic absorber. As the electric resistance for these materials is low, it causes the decrease of magnetization due to the eddy current loss induced by electromagnetic wave irradiation. For this problem, Sugimoto and co-workers prepared the Fe/SmO nanocomposite material by the hydrogenation disproportionation and the subsequent oxidation treatments of Sm_2Fe_{17} and reported the good electromagnetic wave absorption properties on it [6,7]. In this nanocomposite, the fine Fe particles were isolated from one another by the insulating SmO particles to effectively reduce the eddy current loss. In our previous works, the Fe/Y_2O_3 and (Fe, Fe_3B)/Y_2O_3 nanocomposite materials were prepared from the respective rapid quenched Y_2Fe_{17} and $Y_2Fe_{77.5}B_{17.5}$ ribbons, which possessed the small and homogeneous metal tissue, by the similar disproportionation process, and showed the excellent

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electromagnetic wave absorption properties with the several GHz range [8,9].

In this study, the nanocomposite materials consisting of Y_2Fe_{17} and amorphous carbon (a-C) were prepared by a simple mechanical grinding method. The a-C is one of the suitable candidates instead of metal oxides as the insulator because of its lightness and good electric resistivity of $\sim 10^4\ \Omega$ m [10]. The electromagnetic wave absorption properties on the nanocomposite materials of Y_2Fe_{17}/a-C were studied in the GHz frequency range.

2. Experimental

The ingot of Y_2Fe_{17} was prepared from the appropriate amounts of Y shots and Fe rods (>99.9% in purity for both metals) by means of arc melting in Ar atmosphere. The Y_2Fe_{17} ribbons were obtained by a single roller melt-spinning apparatus with roll surface velocity of 20 m/s from the above ingot. The resultant ribbons were crushed into powders below the diameter of $\sim$3 μm, and then were mechanically mixed with a-C (weight ratio, Y_2Fe_{17}:a-C = 94:6) in Ar by the planetary-type ball mill for 30 h with the revolution rate of 200 rpm. The powder of a-C was heated in vacuum at 673 K for 3 h to remove the moisture before use. The resultant powders were checked by an X-ray diffractometer using Cu Kα radiation (RIGAKU RINT2200). Epoxy resin composite were prepared by mixing epoxy resin with 75 wt.% composite powders and pressing into cylindrical-shaped compacts. After curing at 453 K for 30 min, they were cut into toroidal shape (ϕ_{out}: 7.00 mm, ϕ_{in}: 3.04 mm). The scattering parameters (S_{11}, S_{21}) were measured on the toroidal-shaped samples by a network analyzer (Agilent Technology 8720E) in the range of 0.05–18 GHz. The relative permeability (μ_r) and permittivity (μ_r) values were determined from the scattering parameters to evaluate the electromagnetic wave absorption properties.

3. Results and discussion

Fig. 1 shows XRD patterns for the Y_2Fe_{17} powders: (a) as-obtained and (b) after mixing with a-C by ball milling. As shown in Fig. 1a, the XRD patterns of Y_2Fe_{17} prepared by melt-spinning could be assigned to the Th_2Ni_{17}-type structure with a space group of $P6_3/mmc$. No peak derived from the Fe precipitation (main peak is appeared at around $2\theta = 44^\circ$) was observed in the X-ray profile, indicating that the single phase of Y_2Fe_{17} could be formed by the melt-spinning method under the present condition. After mechanically grinding with a-C, all of the XRD patterns were indexed as the Y_2Fe_{17} phase and there was no peak corresponded to carbon components due to the amorphous state of carbon. The diffraction peaks, however, became to be broadened as shown in Fig. 1b. Mean primary particle sizes of the Y_2Fe_{17} powders as-obtained and after mechanically grinding with a-C were evaluated from the line broadening of the X-ray diffraction peaks by the following Scherrer's equation:

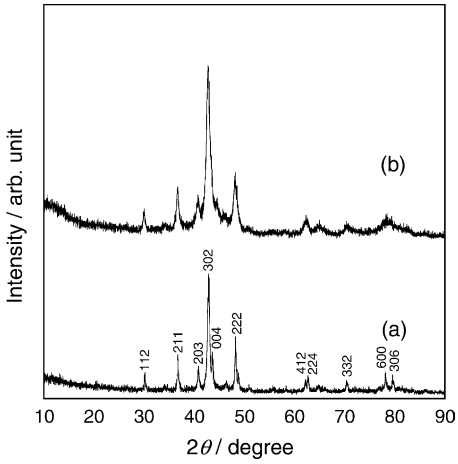

Fig. 1. XRD patterns for the Y_2Fe_{17} powders: (a) as-obtained and (b) after mechanical grinding with amorphous carbon at rotation speed of 200 rpm for 30 h.

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

where D is the primary particle size, λ the wavelengths of the X-rays, θ the diffraction angle and β is the FWHM of the peak. The mean particle sizes of the Y_2Fe_{17} as obtained and mixed Y_2Fe_{17}/a-C powders were calculated to be $\sim$40 and $\sim$20 nm, respectively. It is considered that the small a-C particles collapsed the primary tissues of Y_2Fe_{17} by the force of mechanical grinding and prevented the recombination of fine Y_2Fe_{17} tissues by inserting among the Y_2Fe_{17} metal particles, that is, to decrease the size of Y_2Fe_{17} particles. Moreover, there is no shift in the diffraction angles for the diffraction peaks of Y_2Fe_{17} before and after the mechanical grinding. This result indicates that a-C was not incorporated into the crystal lattice of Y_2Fe_{17} during the mixing process. The results of XRD measurements suggest that the small particles of Y_2Fe_{17} were substantially dispersed in a-C by the mechanical grinding to provide the nanocomposite particles.

The frequency dependence of relative permittivity ($\varepsilon_r = \varepsilon'_r - j\varepsilon''_r$) for the resin composite included 75 wt.% Y_2Fe_{17}/a-C is shown in Fig. 2. The real and imaginary parts (ε'_r and ε''_r) of relative permittivity were almost constant in the range from 0.05 to 18 GHz, suggesting the high resistivity of Y_2Fe_{17}/a-C composite. In fact, a pellet of Y_2Fe_{17}/a-C composite, which was prepared by pressing without resin, had the high electric resistivity of $\sim$200 Ω m. This value is significantly larger than that reported on $Nd_2Fe_{14}B$ ($1.4 \times 10^{-6}\ \Omega$ m) [11]. The high resistivity observed on the Y_2Fe_{17}/a-C composite is ascribable that the a-C particle insulated the conductive Y_2Fe_{17} particle by embedding among Y_2Fe_{17} grains as a separator.

Fig. 3 shows the real and imaginary parts (μ'_r and μ''_r) of relative permeability as a function of frequency. The real part of relative permeability μ'_r declined from 2.3 to 0.98 with

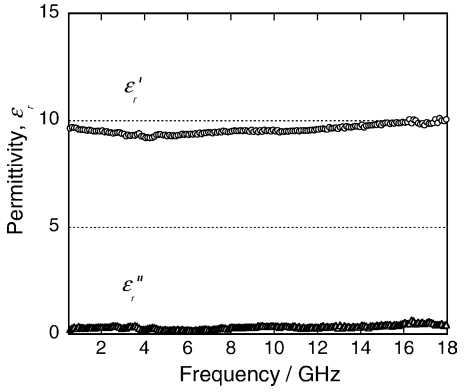

Fig. 2. The ε_r' and ε_r'' curves plotted against frequency for the Y_2Fe_{17}/a-carbon resin composites with 75 wt.% powders.

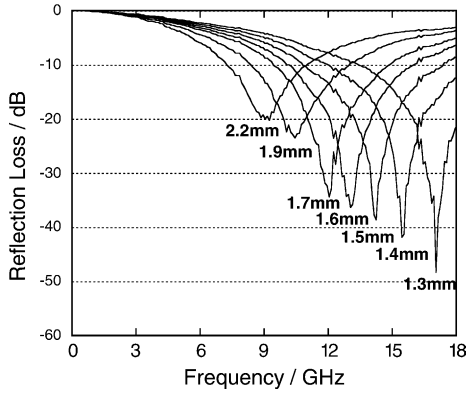

Fig. 4. Frequency dependence of the reflection loss (RL) for the Y_2Fe_{17}/a-carbon resin composites with 75 wt.% powders with various matching thickness.

increasing the frequency, whereas the μ_r'' values gave a broad peak in the range of 2–18 GHz, and kept still the high value in the above GHz region. From the results of measurements for relative permittivity and permeability, it can be expected that the Y_2Fe_{17}/a-C composite gives the good electromagnetic absorption properties at high frequency range.

The reflection loss (RL) curves were determined from the relative permeability and permittivity measured directly and variable parameters such as frequency and absorber thickness as following equations:

$$Z_{in} = Z_0 \left(\frac{\mu_r}{\varepsilon_r}\right)^{1/2} \tan h\{j\left(\frac{2\pi fd}{c}\right)(\mu_r \varepsilon r)^{1/2}\} \quad (2)$$

$$RL = 20 \log \left|\frac{Z_{in} - Z_0}{Z_{in} + Z_0}\right| \quad (3)$$

where f is the frequency of the electromagnetic wave, d the thickness of an absorber, c the velocity of light, Z_0 the impedance of air and Z_{in} is the input impedance of absorber.

The variation of reflection loss values with frequency for the resin composite is shown in Fig. 4. The RL values elevated with increasing the frequency and thickness and the values less than −20 dB were obtained in the range from 9 to 18 GHz with the respective thickness from 2.2 to 1.3 mm. The minimum RL value of −48 dB was observed at 17 GHz on a sample with a matching thickness (d_m) of 1.3 mm. As described above, the Y_2Fe_{17}/a-C composite had the high resistivity value derived from its nano-sized structure, so that the good electromagnetic absorption properties could be obtained due to the decrease of eddy current loss. To compare with ferrites, the matching thickness (d_m) for Y_2Fe_{17}/a-C composite absorbers thinned down about 20–40% in the same frequency range [12,13], since the Y_2Fe_{17}/a-C resin composite possess the higher μ_r'' value than ferrites in the above frequency range. Furthermore, the effective absorption (RL < −20 dB) for the Y_2Fe_{17}/a-C resin composite was obtained in the range of 9–18 GHz, which is higher than that for α-Fe/Y_2O_3 reported previously [8]. The natural resonance frequency f_r is related to the magnetic anisotropy field H_A as a below equation:

$$2\pi f_r = \gamma H_A \quad (4)$$

where γ is the gyrometric ratio [14]. The magnetic anisotropy field values and estimated natural resonance frequencies for Y_2Fe_{17} and Fe are listed in Table 1. The above shift in the absorption range is due to the larger anisotropy field H_A of Y_2Fe_{17} than that of Fe because of the difference of crystal structure between anisotropic Y_2Fe_{17} (hexagonal) and isotropic Fe (bcc). Consequently, the Y_2Fe_{17}/a-C composite powder showed the good electromagnetic absorption properties in the several dozen GHz range due to the appropriate

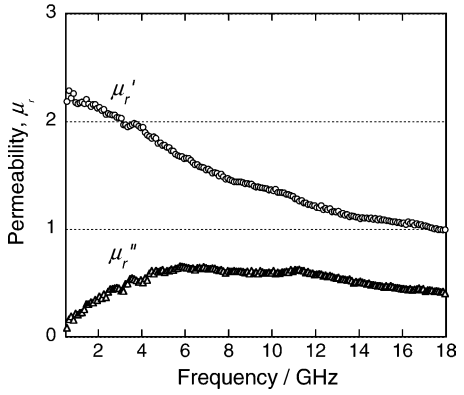

Fig. 3. Variation of μ_r' and μ_r'' as a function of frequency for Y_2Fe_{17}/a-carbon resin composites with 75 wt.% powders.

Table 1
Magnetic anisotropic field values for Y_2Fe_{17} and Fe, together with their calculated natural resonance frequencies and matching frequency range (f_m) observed in this study

	H_A ($\times 10^4$ A/m)	f_r (GHz)	f_m (GHz)[a]
Y_2Fe_{17}	32.5[b]	11.1	9–18
Fe	4.4[b]	1.5	2–3.5

[a] The results observed in the Y_2Fe_{17}/a-carbon and Fe/Y_2O_3 composites. For latter, the details described in ref. [8].
[b] Data from the literature shown in ref. [15,16].

magnetic anisotropy field of Y_2Fe_{17}, and it can be applied for the LAN systems (19.0–22.0 GHz) and satellite broadcast systems (11.7–12.0 GHz).

4. Conclusions

The nanocomposite powders of Y_2Fe_{17}/a-C are prepared by the mechanical grinding method using the ball mill. After grinding with a-C, the crystalline size of Y_2Fe_{17} was decreased from 40 to 20 nm, and no diffusion of carbon into the Y_2Fe_{17} crystal lattice was detected by the XRD measurements. These results suggest that the small particle of a-C cracked the Y_2Fe_{17} crystallites and retarded the further aggregation of Y_2Fe_{17} particles to consequently form the nanostructured Y_2Fe_{17}/a-C composite. The measurement of relative permittivity for the resin composite with 75 wt.% Y_2Fe_{17}/a-C intimates the high resistivity of the resulting composite. The actual resistivity of Y_2Fe_{17}/a-C composite without resin was measured to be 200 Ω m. This result bears out the model for nano-sized structure of Y_2Fe_{17}/a-C composite deduced from the XRD measurements. The RL values for electromagnetic wave on the resin composite were calculated from the results of relative permittivity and permeability measurements. The good electromagnetic wave absorption properties were obtained at the high frequency region on the resin composite (RL value < -20 dB in the range of 9–18 GHz with a matching thickness of a 1.3–2.2 mm). The weight and thickness of absorbers can be decreased using the present composite materials due to the lightness of a-C powders and the high magnetization value of Y_2Fe_{17} in the above frequency range, respectively. As a result, the thin and light electromagnetic wave absorbers against to high frequency range can be obtained from the nanocomposite powders of Y_2Fe_{17}/a-C prepared by the simple mechanical grinding technique.

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