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Microwave Absorption Properties of Carbonyl Iron Particles Filled in Polymer Composites

Wongyu JANG · Shanigaram MALLESH · Ki Hyeon KIM*
Department of Physics, Yeungnam University, Gyeongsan 38541, Republic of Korea

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We report the electromagnetic wave absorption performance of carbonyl iron-particles (CIP) magnetic fillers in a polydimethylsiloxane (PDMS) matrix prepared by using a doctor blade method. The X-ray diffraction and the field-emission scanning electron microscopy studies of the CIPs revealed a BCC crystal structure and an average diameter of 3 μm. For the CIPs, the saturation magnetization, M_S, was 205 emu/g, and the coercivity, H_C was around 12 Oe. The microwave absorption properties were measured using a vector network analyzer in the frequency range from 0.1 to 18 GHz. We observed a systematic increase in the minimum reflection loss (RL) with increasing weight fraction of CIP fillers (46 wt% to 72 wt%) in the PDMS matrix. The minimum RL was -27.5 dB at 14.6 GHz with a thickness of only 1.5 mm and an effective absorption bandwidth of up to 6.8 GHz (RL @ -10 dB). Therefore, the present study suggests that CIP magnetic fillers in a PDMS matrix are a good candidate for thin, broadband absorbing fillers.

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I. INTRODUCTION

With the rapid development of electronic devices and wireless communication technologies, electromagnetic interference and radiation pollution have become a severe problem for human health. Therefore, it is essential to design the electromagnetic (EM) wave absorption materials with a thin thickness, lightweight, strong absorption ability with broad bandwidth, and chemical stability, which are required for practical applications [1–7]. The complex permittivity and permeability values of the absorber determine the EM wave absorption performance depending on impedance matching conditions [6,7].

Recently, the EM wave absorption properties of carbonyl iron particles (CIP) gained remarkable interest owing to its relatively low electrical conductivity, high saturation magnetization, Curie temperature, and permeability compared with other metallic particles [8–10]. Nevertheless, the high density, poor stability, and large loading content constrain their practical applications. Several reports on EM wave absorption properties of CIP and its composites have been investigated in the frequency range from 2 – 18 GHz. Yu *et al.* reported the minimum value of reflection loss (RL) for CIP is – 24.3 dB at 11.8 GHz with a thickness of 3 mm [11]. Yang *et al.* demonstrated that the minimum RL is reached to -23 dB at 5.5 GHz with a thickness of 3 mm for flake-shaped CIP [12]. The minimum RL value was -25.1 dB at 14.2 GHz with a thickness of 6 mm is observed for CIP coated on graphite composites [13]. Zhang *et al.* presented the minimum RL value in CIP/milliable polyurethane composites was – 28.4 dB at 3.4 GHz at a thickness of 3 mm [14]. However, lightweight and effective broadband microwave absorption at small thickness is still challenging in CIP and its composites.

In this report, the EM wave absorption sheets of CIP fillers in the PDMS matrix fabricated using a doctor blade method to study the performance of EM wave absorption. The microstructure and magnetic properties of CIP particles were investigated.

*E-mail: kee1@ynu.ac.kr

II. EXPERIMENTS

Carbonyl iron particles (CIP, BASF, EW) magnetic fillers and polydimethylsiloxane (PDMS, Dow Corning) polymer resin matrix used to prepare a flexible EM wave absorbing sheet. The PDMS is mechanically flexible, wear-resistance, and high transparency. The advantage of PDMS is a uniform dispersion of magnetic fillers within a polymer matrix can be obtained [15]. The CIP mixed with soft PDMS using a paste mixer, then after, hard PDMS is added to the solution continued mixing of the solution in a paste mixture. The obtained mixture solution was used to cast the sheet on a release film tape using the doctor blade method. The sheet was dried in an air oven at 60 °C for 12h and separated from release film tape. The EM wave absorbing sheets with various weight fractions of CIP (46 wt%, 58 wt%, 66 wt% and 72 wt%) in the PDMS matrix were fabricated to appraise the EM wave absorption properties.

The phase purity and crystal structure of the CIP was obtained by using X-ray diffraction (XRD) data (PANalytical, X'pert PRO) with CuKα (λ=1.54059 Å) radiation. Field emission scanning electron microscopy (FESEM, S-4800, Hitachi) was used to analyze the morphology of the CIP. A vibrating sample magnetometer (VSM, 7404-S, LakeShore) was used to measure the isothermal magnetization at room temperature. The as-prepared CIP in PDMS flexible sheets were punched to make toroid-shaped samples with an outer diameter of 7.00 mm and an inner diameter of 3.04 mm for EM wave absorption property measurements. The frequency-dependent complex permeability and permittivity were measured by (Keysight) Vector Network Analyzer (VNA, N5222B) with a 40 mm coaxial line.

III. RESULTS AND DISCUSSION

Figure 1(a,b) illustrates the FESEM images of CIP obtained at different magnifications. It is evident from the figure that the morphology of CIP displays a spherical shape with an average particle size of 3 μm. The phase purity and crystal structure of the CIP sample obtained from XRD data as shown in Fig. 2(a). The diffraction peaks of (110), (200) and (211) correspond

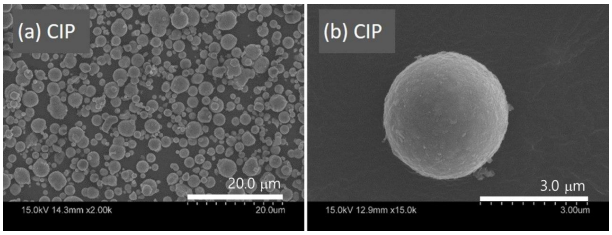

Fig. 1. (Color online) (a,b) FESEM images of CIP at different magnifications.

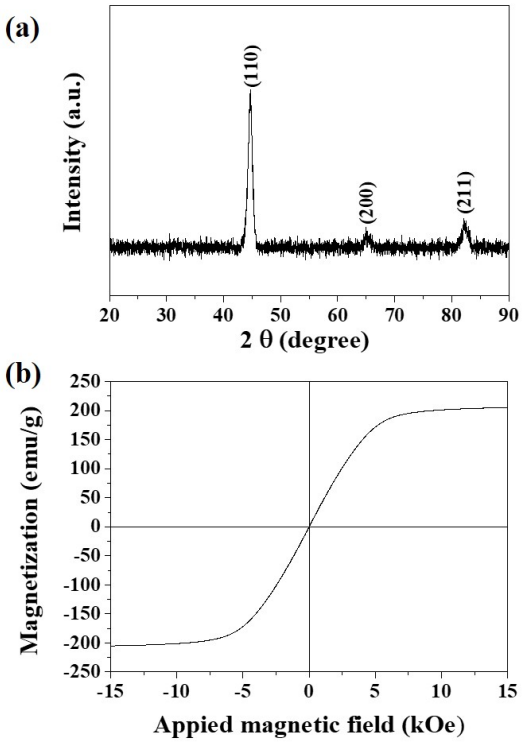

Fig. 2. (a,b) XRD patterns and M-H curve of CIP samples.

to body-centered cubic (BCC) α-Fe (JCPDS 06-0696). This observation is in good agreement with previous reports [8,13]. The average crystallite size is estimated to be 17.6 nm by employing Scherrer's formula [16]. Figure 2(b) describes the magnetization measured as a function of the magnetic field at room temperature reveals the saturation magnetization, (M_S) 205 emu/g and coercivity (H_C) 12 Oe for CIP sample.

It is well established from the literature; the EM wave absorption properties can be calculated using complex permittivity and permeability. In order to assess the EM wave absorption properties of CIP filled PDMS composites (42 wt% to 72 wt%), we measured the complex

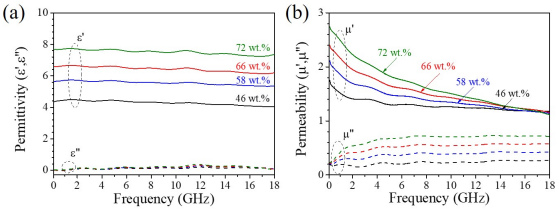

Fig. 3. (Color online) (a) The relative complex permittivity real part (ε') and imaginary part (ε''). (b) The relative complex permeability real part (μ') and imaginary part (μ'') of CIP filler PDMS composites.

permittivity and permeability of the composites in the frequency range from 0.1 to 18 GHz. Figure 3 (a) shows the frequency-dependent real and imaginary parts of the permittivity of CIP films. The real part of permittivity (4.4 for 46 wt%, 7.7 for 72 wt%) shows a linear increment with increasing the weight fraction of CIP in the PDMS matrix. The imaginary part of permittivity exhibits negative values at low frequency and then, almost remains constant (< 1) for the complete measured frequency range. The real part of permeability increases from 1.74 to 2.73 with increasing weight fraction of CIP from 46 wt%, to 72 wt% at low frequency ($<$ 1GHz), further, it decreased to 1.2 on increasing frequency to 18 GHz as shown in Fig. 3(b). On the other hand, the imaginary part of permeability is minimum at low frequency ($<$ 1GHz) and then increases with frequency ($<$ 5GHz) and remains the same with the increase of frequency.

In general, the value of RL measured below –10 dB (90% absorption) is regarded as efficient microwave absorbers. Based on measured complex permittivity and permeability, the RL values were calculated using the following equations given below [17,18]

$$RL(\text{dB}) = 20\log|(Z_{in} - Z_0)/(Z_{in} + Z_0)| \qquad (1)$$

$$Z_{\text{in}} = Z_0(\mu_r/\varepsilon_r)^{1/2}\tanh\left[j\left(\frac{2\pi fd}{c}\right)(\mu_r\varepsilon_r)^{1/2}\right] \qquad (2)$$

where ε_r and μ_r are the relative complex permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) and complex permeability ($\mu_r = \mu' - j\mu''$), respectively, Z_{in} is the input impedance of absorber, Z_0 (377 Ω) is the free space impedance, f is the frequency, c is the velocity of EM wave in free space, and d is the absorber thickness. To get a minimized RL values, the impedance of absorber should be matched with

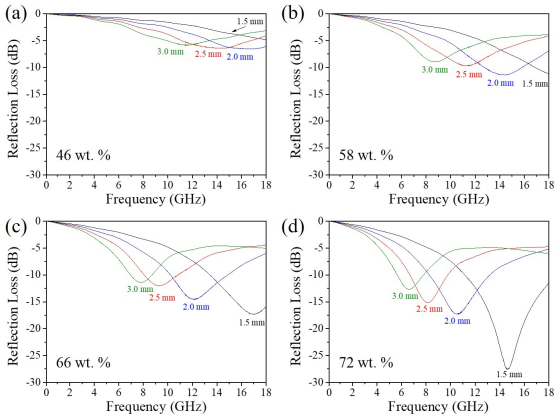

Fig. 4. (Color online) (a-d) The electromagnetic wave reflection loss at various thicknesses of CIP filler PDMS composites.

that of air impedance, which impedance is governed by permittivity and permeability values of each medium. When the weight percentage of micron-sized conductive fillers are increased in a polymer matrix, the permittivity and permeability are generally increased. For matching impedance at a specific frequency, the permittivity, permeability, and thickness related with EM path of absorber should be controlled optimally. When an EM wave is incident to absorber on PEC (perfect electric conductor) backed plate, the incident EM wave makes two kinds of reflection at the interface between air and absorber, absorber and PEC backed plate, respectively. These two reflected EM waves occur the 180° out of phase destructive interference by a higher order of refractive index of interfaces. When the thickness of absorber is 1/4 of the wavelength ($\lambda/4$), the reflected signal should be almost canceled, and then RL value is minimized [19].

Figure 4 demonstrates the RL evaluated using relative permittivity and permeability with an increment of absorber thickness for CIP filled PDMS composites. The minimum value of RL is below – 10 dB in the complete measured frequency range when the weight fraction of CIP is $\geq$ 46%, further it increases with the weight fraction of CIP in the PDMS matrix. The RL peaks were shifted to lower frequency with the increment of the sample thicknesses. The minimum value of RL of CIP composite (72 wt%) is reached to – 27.5 dB at 14.6 GHz with a thickness of only 1.5 mm. The bandwidth measured at – 10 dB (BW@-10 dB) is 6.8 GHz at a thickness

of 1.5 mm. The enhanced microwave absorption properties were attributed to the synergistic effect of dielectric and magnetic loos, good impedance matching, and strong attenuation of the EM wave through the absorber material. The present investigations on CIP filled PDMS composites, useful for broadband microwave absorption application.

IV. CONCLUSIONS

The EM wave absorption films with varying weight fraction of CIP in the PDMS matrix were successfully prepared via a doctor blade method. The structure and morphology and magnetic properties of CIP sample were investigated using XRD, FESEM, and VSM. The complex permittivity and permeability were measured in the frequency range from 0.1 – 18 GHz. The EM wave absorption properties were improved with increasing wt% of CIP in the PDMS matrix. The minimum RL of CIP (72 wt%) is reached up to – 27.5 dB at 14.6 GHz with the effective bandwidth (BW@-10 dB) is 6.8 GHz at a thickness of 1.5 mm. These results specify that the CIP is a good candidate filler for the broadband microwave absorption materials.

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