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ENHANCED MICROWAVE ABSORPTION OF CNT COMPOSITES MIXING WITH Fe_3O_4 AND CARBONYL IRON

We fabricated two different kinds of composite materials for absorbing microwave in a frequency range of 2 to 18 GHz using coaxial airline and thru-reflect-line (TRL) method. The composite materials having carbon nanotube (CNT) with carbonyl iron (CI) or iron oxide (Fe_3O_4) were fabricated by mixing each components. Magnetic properties were measured by SQUID equipment. Complex permittivity and complex permeability were also obtained by measuring S-parameters of the toroidal specimen dispersing CI/CNT and Fe_3O_4/CNT into the 50 weight percent (wt%) epoxy resin. The real permittivity was improved by mixing the CNT however, the real permeability was same as pure magnetic powders. The CI/CNT had a maximum value of real permittivity and real permeability, 11 and 1.4 at 10 GHz, respectively. The CNT composites can be adapted to the radar absorbing materials, band width 8-12 GHz.

Keywords: Magnetic powders, Complex permittivity, Complex permeability, Composite materials, Microwave absorption

1. Introduction

In the recent years, microwave absorbing materials are focused on shielding of electromagnetic pollution from many kinds of electronic devices and magnetic applicants [1-2]. Also, it is very important issue in defense system for radar absorption [3-5]. Many researchers have concentrated on developing efficient absorbers having low density, low thickness, broad band microwave absorbing characteristic and high durability in extreme conditions [6]. Furthermore, characteristics of microwave absorbing materials are decided by two parameters, complex permittivity ($\varepsilon^* = \varepsilon' - j\varepsilon''$) and permeability ($\mu^* = \mu' - j\mu''$) that are optimized by controlling of magnetic and conductive concentrations [7]. In this study, we used carbonyl iron (CI) and iron oxide (Fe_3O_4) as a magnetic component and carbon nanotube (CNT) as a conductive one. It is generally known that CI has been used for electromagnetic wave absorbing materials because of its low price, high specific saturated magnetization. In addition, Fe_3O_4 has similar advantages such as low price, relatively strong magnetic properties and good magnetization durability at high temperature condition [8]. CNT have been utilized for electromagnetic interference (EMI), microwave absorption and electrode. One of the biggest benefit of CNT is low density that can reduce density and total weight of composite materials.

Herein, we simply mixed CNT with CI or Fe_3O_4 using sonication process in a nonpolar solvent for dispersion and optimization of electromagnetic properties of the composite materials. The sonication process was appropriate to the mass production system and the production cost could be reduced. We carried out structural analysis using SEM and XRD and measured magnetic property using the SQUID. Also, we calculated complex permittivity and complex permeability of composite materials such as CI, CI/CNT, Fe_3O_4 and Fe_3O_4/CNT from measured S-parameter. The pure magnetic powders and composite materials were fabricated with toroidal shape by mixing them into the 50 wt% epoxy resin for measuring complex permittivity and complex permeability. The complex permittivity and complex permeability of the toroidal specimen was characterized by using vector network analyzer (VNA, N5222A, Agilent Technologies) and coaxial airline. We acquired the values of magnetization, complex permittivity and complex permeability. Based on the measured complex permittivity and permeability, the CNT composites can be adapted to the microwave absorption (8-12GHz).

2. Experimental

We prepared pure CI and Fe_3O_4 powders and mixed with CNT by sonication process. All of the chemicals used in this research were purchased from Sigma Aldrich and mixed without further purifications. At first, a triethylene glycol was poured in 60 ml glass vials and then the CNT was put into the triethylene glycol. The solution was mixed with glass stick with sonication for well dispersion of CNT (10 wt% of composite materials) during 30 min. The CI or Fe_3O_4 was put into the CNT solution and then we carried out the sonication process for 2 hr. After sonication process, the solution was filtered using the anodisc 47 and then washed with ethanol and acetone 3 times. The com-

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posite powders were dried in vacuum oven at 80°C for 12 hr and the composite structures were analyzed by XRD and SEM. For measuring the permittivity (ε^*) and permeability (μ^*), we fabricated the toroidal sample, mixing in the epoxy matrix, adding the composite powders with mechanical stirring using the glass stick and sonication process in the glass beaker for 1 hr. The solutions were poured into the aluminum plate and dried in the air for 12 hr. The cylindrical toroidal samples with 3 mm in inner diameter, 7 mm in outer diameter and 2 mm in thickness by uniformly mixing the 50wt% composites powders being pressed into cylindrical compacts. The permittivity and permeability were measured using the network analyzer using the toroidal samples. In the measurement, the morphology and surface of composite materials was analyzed by SEM (FE-7800F, JEOL). The phase identification was measured by X-ray diffraction (SmartLab, Rigaku). The magnetic properties of pure magnetic powders and composite materials were measured by SQUID (MPMS-3 Evercool, Quantum Design Inc.) The complex permittivity and complex permeability of toroidal samples were measured by PNA Network Analyzer (N5222A, Agilent Technologies)

3. Results and discussion

In the Fig. 1, the SEM images show the morphology analysis of pure CI, Fe_3O_4, CI/CNT and Fe_3O_4/CNT. The pure CI powders have a spherical shape with smooth surface and they have a size variation, 1~5 μm (Fig. 1a). After mixing the CNT, the CI powders are well dispersed between the CNT bundled, and the morphology of CI powder is shown in Fig. 1b. The pure Fe_3O_4 nanoparticles have an octahedral structure and the size variation is distributed between 50~100 nm. (Fig. 1c) The Fe_3O_4 nanoparticles were also well mixed with CNT after sonication process and the morphology of Fe_3O_4 nanoparticles isn't changed as shown in Fig. 1d. In addition, we carried out XRD measurement for the crystal structural analysis before and after mixing CNTs under sonication process. The main peaks of pure CI and Fe_3O_4 are well matched with reference data (JCPDS #06-0696 and #75-0033, respectively) and each of peaks are indexed. Furthermore, the main peaks of XRD aren't changed after sonication process which means the sonication process doesn't affect the crystal structure of magnetic powders (Fig. 2). We measured magnetization (M) values from –2T to 2T using the SQUID equipment (Fig. 3). In a case of Fe_3O_4 nanoparticles, there was not much change depending on the existence of CNT. However, CI and CI/CNT have a different magnetization shape even their saturation values were same. Based on the M-H measurements, we could calculate the real permeability using following equations [9-10].

$$\text{Magnetization } (M) = m/v \quad (m: \text{magnetic moment}, v: \text{volume of sample}) \quad \text{(eq. 1)}$$

$$\text{Magnetic Induction } (B) = 4\pi M + H \quad \text{(eq. 2)}$$

$$\text{Permeability } (\mu) = B/H = (4\pi M + H)/H = 1 + 4\pi M/H \quad \text{(eq. 3)}$$

As mentioned above, we fabricated the toroidal specimen for measuring complex permittivity and complex permeability at 2-18 GHz range. We carried out the measurement of complex

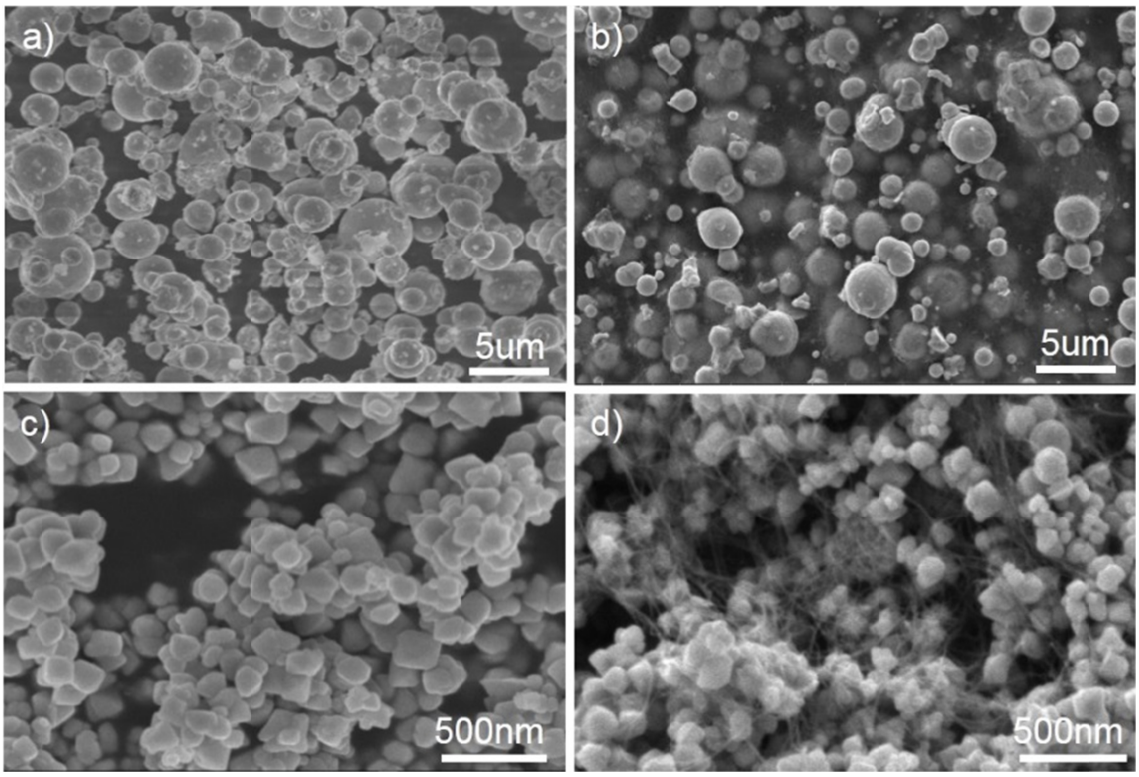

Fig. 1. SEM image of a) CI, b) CI/CNT, c) Fe_3O_4, and d) Fe_3O_4/CNT, respectively

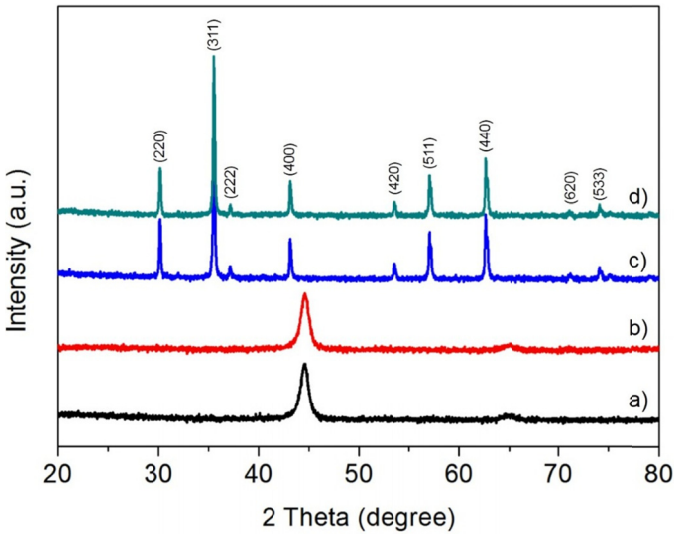

Fig. 2. XRD peaks of a) CI, b) CI/CNT, c) Fe_3O_4, and d) Fe_3O_4/CNT, respectively

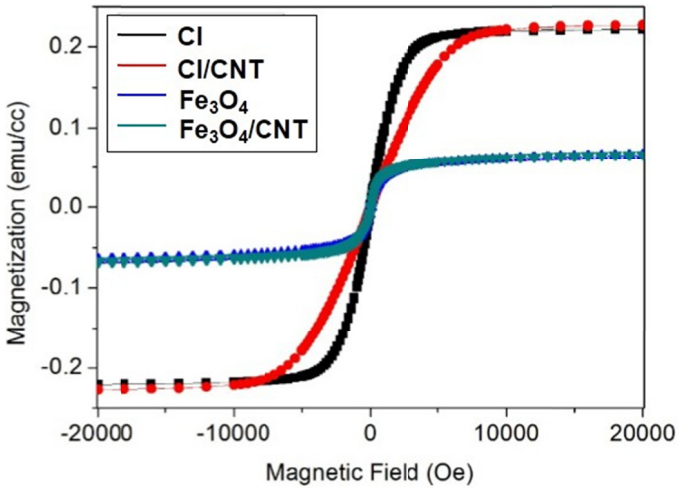

Fig. 3. SQUID measurements of CI, CI/CNT, Fe_3O_4, and Fe_3O_4/CNT

permittivity and complex permeability using vector network analyzer.

The composite materials such as CI/CNT and Fe_3O_4/CNT (about 10 and 9 at 10GHz, respectively) have larger values of real permittivity than pure CI and Fe_3O_4 (about 6 and 8 at 10GHz, respectively) because CNT has high electrical conductivity and high dielectric loss. However, real permeability of CI and CI/CNT have similar level owing to the non-magnetic properties of CNT. (Fig. 4 a and b) In a case of Fe_3O_4 and Fe_3O_4/CNT have same phenomena with CI and CI/CNT. (Fig. 4c and d) The dielectric loss tangent of CI/CNT and Fe_3O_4/CNT were about 0.3 while that of CI and Fe_3O_4 were 0.09 and 0.16 at 10 GHz, respectively as illustrated in Fig. 4c and d. We can deduce that there are magnetic-dielectric coupling effect in double compo-

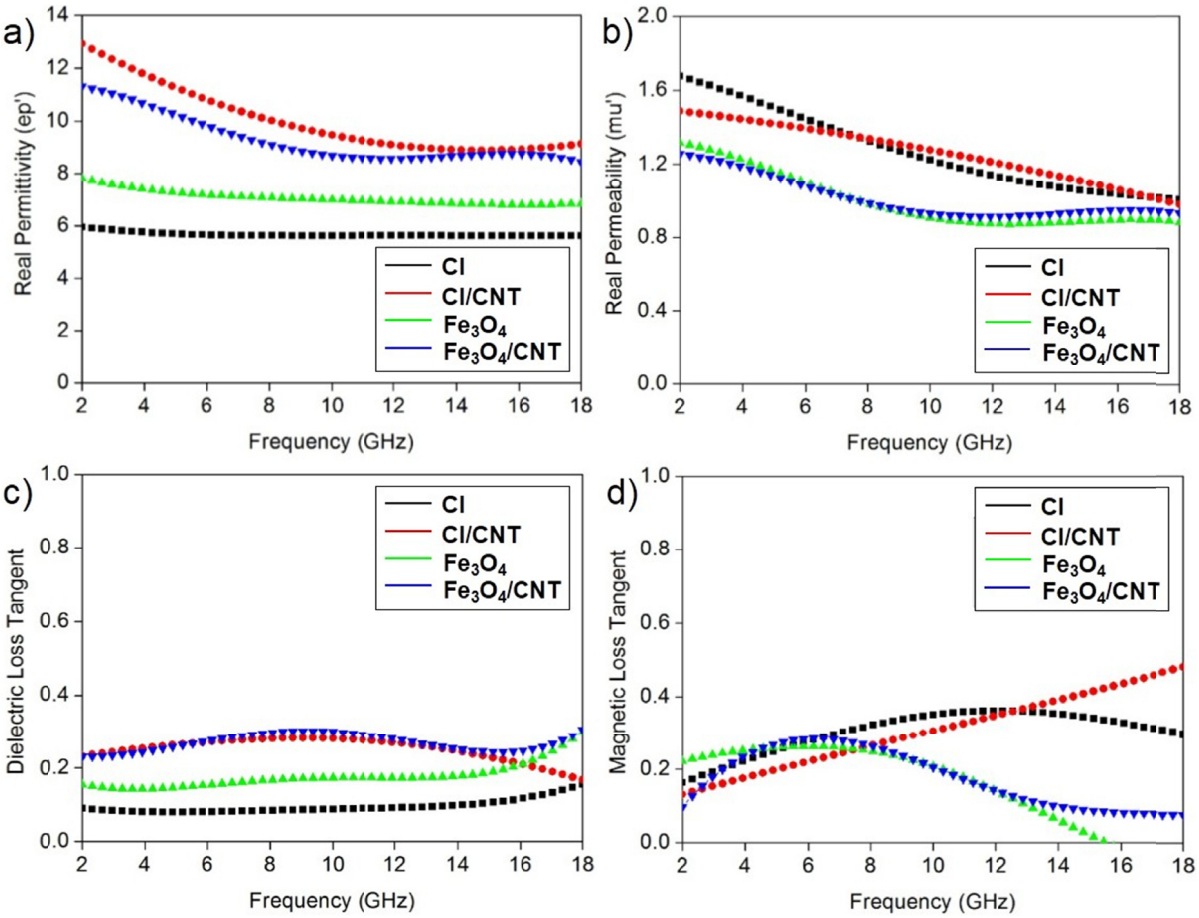

Fig. 4. a) Real permittivity of CI, CI/CNT, Fe_3O_4, and Fe_3O_4/CNT, b) Real permeability of CI, CI/CNT, Fe_3O_4, and Fe_3O_4/CNT, c) Dielectric Loss Tangent of CI, CI/CNT, Fe_3O_4, and Fe_3O_4/CNT, and d) Magnetic Loss Tangent of CI, CI/CNT, Fe_3O_4, and Fe_3O_4/CNT

nents composites. Based on the eq1-eq3, the efficient microwave absorption materials should had proper value of permittivity and permeability. The CI/CNT and Fe_3O_4/CNT had a proper value of permittivity and permeability so they could be adapted to the microwave absorption materials.

4. Conclusions

In this research, we fabricated the composite materials having each components of CI/CNT and Fe_3O_4/CNT and compared the microwave absorption properties of pure CI and pure Fe_3O_4 nanoparticles. Also, we knew that the sonication process doesn't affect the crystal structure change from structural analysis using the SEM and XRD. Also the magnetization wasn't changed but the value of real permittivity and dielectric loss tangent was improved as adding the CNT in the magnetic powders. These improved properties of real permittivity and dielectric loss tangent affect to the refection loss at 2-18 GHz region. From the research data, the CNT composites could be adapted to the microwave absorption (8-12 GHz) for radar absorbing.

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