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Microwave absorbing property of silicone rubber composites with added carbonyl iron particles and graphite platelet

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ABSTRACT

Silicone rubber composites filled with carbonyl iron particles (CIPs) and graphite platelet (GP) were prepared using non-coating or coating processes. The complex permittivity and permeability of the composites were measured using a vector network analyzer in the frequency range of 1–18 GHz and dc electric conductivity was measured by the standard four-point contact method. The results showed that CIPs/GP composites fabricated in the coating process had the highest permittivity and permeability due to the particle orientation and interactions between the two absorbents. The coating process resulted in a decreased effective eccentricity of the absorbents, and the dc conductivity increased according to Neelakanta's equations. The reflection loss (RL) value showed that the composites had an excellent absorbing property in the L-band, minimum −11.85 dB at 1.5 mm and −15.02 dB at 2 mm. Thus, GP could be an effective additive in preparing thin absorbing composites in the L-band.

1. Introduction

Radar absorbing materials (RAM) have been widely used in military applications and civil aspects as a coating or plate structure. There are mainly three types of absorbing materials according to the absorbing mechanisms, dielectric loss, magnetic loss and conductive loss materials [1–3]. Recently, absorbing materials with low thickness, wide absorbing band, light weight and high absorption ratio, are the promising candidates and attract many researchers. Carbonyl iron particles (CIPs) are considered as fine magnetizable particles that are used not only in magnetorheological fluids due to their large values of saturation magnetization value [4], but also in gigahertz (GHz) frequency due to the high Snoek's limit. For the spherical CIPs, Snoek's limit could be described in the following equation [5,6]: $(\mu_s-1)f_r=\gamma M_s/(3\pi)$, where the right part of the expression is defined as Snoek's constant, γ denotes the gyromagnetic ratio, and M_s is the saturation magnetization, μ_s is the static permeability and f_r is the resonance frequency; both μ_s and f_r could not be increased at the same time. Previous researches showed that the spherical CIPs had a limitation in fabricating excellent absorbing materials with low thickness and low frequency such as L-band (1–2 GHz) and S-band (2–4 GHz). For example, the reflection loss (RL) of composites to which 55 vol% CIPs was added just reached −3 dB at 2 GHz with 1 mm thickness [7], and −5 dB at 4 GHz with 1 mm thickness as 93 wt% CIPs was added [8]. However, as the particles were flaky shaped with the same density, the saturation magnetization occurred at a much smaller field, which means that the static permeability was higher than that of spherical CIPs [9,10]. And the flaky CIPs composite shows a dramatic enhancement of complex permeability and a higher resonance frequency compared with that of spherical CIPs composite, for the parameters μ_s and f_r of flaky particles can be constrained in the following equation [11,12]: $(\mu_s-1)f_r=\gamma M_s\sqrt{H_{ha}/H_{ea}}/(3\pi)$, where H_{ha} and H_{ea} denote the effective anisotropy field when the magnetization deviates from the easy axis in the hard plane and in the easy plane respectively. The permeability of flaky CIPs would be much higher than that of spherical CIPs because H_{ha} was much larger than H_{ea}; as a result, the flaky CIPs composite with the same volume content and thickness could achieve a higher absorption ratio at lower matching frequency.

Meanwhile, as one of the carbonous materials, the graphite platelet (GP) has been studied in the development of light-weight and electrically conductive multi-functional materials [13]. With 5 vol% GP added, the GP/epoxy resin composites had a first matching thickness 8.2 mm at frequency 15 GHz with RL −20 dB [14]. In order to enhance the absorbing property of the composites filled with GP, hybrid absorbents were prepared (such as FeNi/GP [15], Fe/GP [16], etc.) due to the dielectric loss of GP and magnetic loss of absorbing metals. However, the complicated preparation processes and rare production restrict the extensive use of these materials. Thus fabricating composites mixing CIPs and GP directly may be an effective way to achieve absorbers of high absorption and low cost.

The present work is to investigate the microwave absorbing property of composites filled with CIPs/GP. First the complex

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permittivity and permeability of silicone rubber composites with added CIPs and GP absorbents were analyzed. Then EM parameters and the dc conductivity of composites using two fabricating processes were compared, and the enhancement mechanism was established. Finally the effects of GP on the absorbing property of the CIPs/rubber composites were analyzed by simulating the reflection loss at frequency 1–18 GHz.

2. Material and methods

2.1. Materials preparation

Methyl vinyl silicone rubber was used as the matrix and 2,5-dimethyl hexane was used as the vulcanized assistant; both were supplied by LaiZhou Jintai Silicon Industry Co. Ltd., China. Raw commercial flaky CIPs were supplied by Shenyang Hangda Technology Co. Ltd., China, and the GP was purchased from Qingdao Shenshu Graphite Manufacturing Co. Ltd., China. The average diameter of flaky CIPs was 5 μm and the thickness was about 0.5 μm; the average diameter of the GP was 2 μm, and the thickness was about 0.5 μm. The morphology of the two particles is shown in Fig. 1.

Samples with only single 50 vol% CIPs and 50 vol% CIPs/5 vol% GP were fabricated separately. The silicone rubber and absorbents were mixed in a two-roll-mixer for 15–30 min. GP was added into the silicone rubber, and then the vulcanized assistants and flaky CIPs were added to the compound. The effects of the absorbent aggregation could be eliminated for the roll-mixer provided the shearing force in the mixing process could overcome the intermolecular van der Waals force between the particles [17,18]. The testing samples for electromagnetic (EM) parameters measurement were modeled to a toroidal shape with an outer diameter 7.0 mm, inner diameter 3.04 mm and thickness 2 mm. The toroidal samples were prepared in two ways, namely the non-coating process and the coating process. As shown in Fig. 2, the non-coating process involved adding the mixed composites to the toroidal mold directly, while the coating process involved stacking several sheets each with a thickness of 0.1–0.2 mm and then shaping to the toroidal shape. As a result, the flaky particles were dispersed randomly in the non-coating process and were aligned in the coating process [19]. Then all the samples were vulcanized into pieces at 180 °C under a pressure 10 MPa for 5 min.

2.2. Sample measurement

The morphology of the composites was observed by scanning electron microscopy (SEM CamScan CS3400) to evaluate the dispersion state and microstructure of GP and CIPs in silicone rubber composites. The dc electric conductivity was measured on pressed rectangular composites prepared at room temperature by the standard four-point contact method. The effective complex permittivity and permeability of the samples were measured using an AV3627 vector network analyzer in the frequency range of 1–18 GHz and then the RL could be calculated. For a single-layer absorbing material, the RL of normal incident EM wave at the absorber surface is given by [20]

$$R = 20\lg\left|(Z_{in} - Z_0)/(Z_{in} + Z_0)\right| \qquad (1)$$

$$Z_{in} = \sqrt{\mu_r\mu_0/(\varepsilon_r\varepsilon_0)}\tanh(j2\pi d\sqrt{\mu_r\varepsilon_r}/\lambda) \qquad (2)$$

where Z_{in} is the normalized input impedance of the absorber, $Z_0(=\sqrt{\mu_0/\varepsilon_0} = 120\,\pi\Omega)$ is the intrinsic impedance of free space, ε_r, μ_r and ε_0, μ_0 are complex permittivity and complex permeability of the absorber and free space respectively, λ is the wavelength, and d is the thickness of the absorber.

3. Results and discussion

3.1. Effects of GP on permittivity and permeability of composites

The complex permittivity (ε) and complex permeability (μ) of each composite depending on the frequency are shown in Fig. 3. Among all the composites, it was clearly observed that the real part permittivity (ε') of composites decreased first, and then remained nearly unchanged in 8–18 GHz, while the imaginary part (ε'') fluctuated. CIPs/GP composites prepared in the coating process had the highest ε and μ; ε' was nearly 2 times larger than that of composites fabricated in the non-coating process, while ε'' was about 6 times larger. The real part permeability (μ') of each

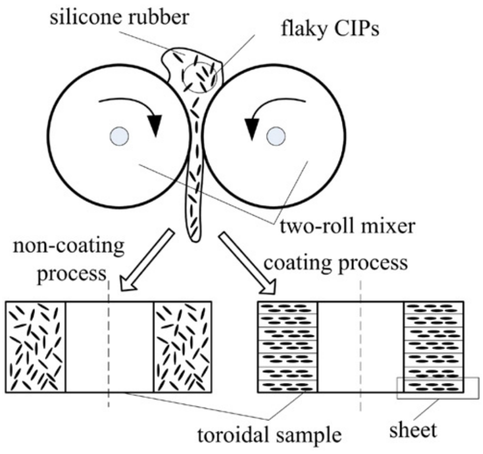

Fig. 2. Schematic diagram of the non-coating process and the coating process.

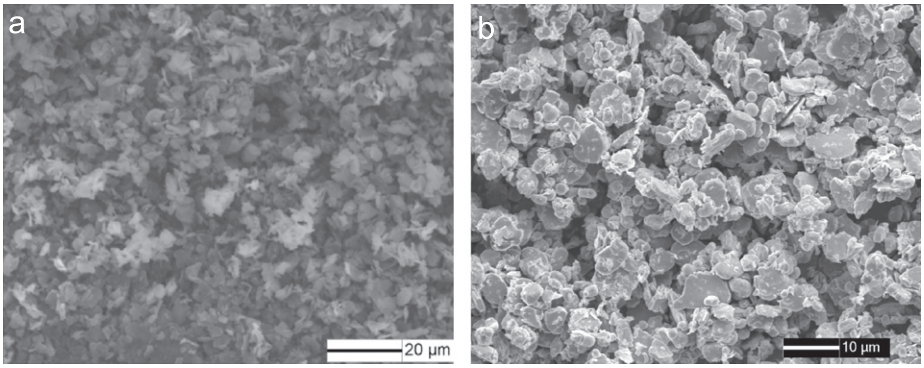

Fig. 1. SEM images of (a) graphite platelet and (b) flaky CIPs.