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Magnetic properties and microwave absorption properties of chlorosulfonated polyethylene matrices containing graphite and carbonyl-iron powder

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ABSTRACT

In this work, the microwave absorbing properties (reflection loss, RL) of composite microwave absorbers with different thickness made of chlorosulfonated polyethylene matrices and different kinds of active fillers were investigated in the frequency range of 2–18 GHz. The materials used as microwave absorbents were graphite (GRP) and carbonyl-iron powder (CIP), and the complex permittivity ε_r and the complex permeability μ_r of the CIP/GRP composites were measured in 2–18 GHz frequency range. It was found that, with the thickness increases from 0.5 mm to 1.5 mm, the absorption peak shifts to lower frequency and the absorption band becomes wider. The frequency width with RL under −10 dB reached 8.96 GHz and the minimum RL was −33.19 dB at 13.24 GHz for the composite microwave absorber of 1.5 mm in thickness.

1. Introduction

Electromagnetic (EM) waves with frequency between 2 and 18 GHz are increasingly widely used in both civil and military applications, especially the radar detection technique for military purpose. Thereby the developments of the stealth technologies for confronting radar detection have become more important[1]. Radar absorbing materials (RAMs) have been broadly used to eliminate or minimize EM waves reflections from large objects such as aircraft, ships, tanks and electronic equipment and to cover the walls and the floor of anechoic chambers [2–4]. The key problem for designing and manufacturing RAMs is related to the choices of the materials [5–8].

The microwave absorbers have dielectric and magnetic losses according to the type of filler used. Carbonyl-iron powder (CIP) is key active ingredient for many EM waves absorbing materials because of the specific magnetic loss originates from the magnetic hysteresis, magnetic domain resonance, natural resonance, eddy current loss and remanence loss [9–12]. The dielectric lossy materials include carbon black (CB), graphite (GRP), carbon nanotubes (CNTs and MWCNTs), conductive polymer, etc. [13–16]. These two loss materials having different advantages and disadvantages of each can be used as a mixture (Fe–Ni alloy/C nanotubes [17], Fe/C nanotubes [18] and nickel-coated carbon fibers[19]).

As for chlorosulfonated polyethylene matrices, commonly referred to as CSM or CSPE, possesses high resistance to hydrocarbon oil, ozone, heat and weathering, which attributed to the addition of chlorine and sulfur atoms, on the polyethylene backbone [20–22].The aim of this paper is to prepare CIP-GRP/CSM composite microwave absorbers and to discuss the absorbing performance of the layered composites.

2. Experiment

The conductive fillers used to absorbing electromagnetic (EM) waves, were carbonyl-iron powder (CIP) and graphite (GRP) purchased from commercial suppliers. The elastomer used was chlorosulfonated polyethylene matrices (CSM) (density = 1.19 g/cm^3; Cl% = 25–45%, S% = 0.8–1.7%) obtained from Du Pont. All others reagents were of commercial grade and used without purification.

CIPs and GRP were blended with CSM in a given proportion on a planetary ball mill at ambient temperature with a rotor speed of 20 and 25 rpm (clockwise and counterclockwise). After filtering, the compositions were compression molded at 100 °C in a hydraulic press at 6.5 MPa. Vulcanized test specimens were obtained in different sizes:800 × 800 × 0.5 mm, 800 × 800 × 1.0 mm, 800 × 800 × 1.5 mm and 800 × 800 × 2.0 mm for reflectivity measurements.

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The particle size and the surface area were determined by laser light scattering. A laser particle analyzer BT-9300H of BETTER Instruments was employed for determination of parameters such as D (ν, 0.5), D (ν, 0.9) and the surface area. The samples were analyzed in ultra-sonic bath.

Scanning electron microscopy (SEM) was carried out in a FEI Quanta 200 microscope for the CIP and the elastomeric composites. The complex permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$), the complex permeability ($\mu_r = \mu' - j\mu''$) and the RL of the composite microwave absorber were measured using a vector network analyzer (Agilent Technology, E8363B) in the 2–18 GHz.

3. Results and discussion

3.1. BET determination

The BET surface area determined for CIP was 0.38213 m^2/g and a smaller value of 0.22151 m^2/g was obtained for graphite. These results indicate a larger particle size for graphite than CIP.

3.2. Determination of particle size

Table 1 illustrates the sizes of CIP and graphite particles determined by the LLS technique. The graphite particles showed greater diameters than CIP.

3.3. Microstructure and chemical composition of CIP

The SEM analysis of carbonyl-iron powders is presented in Fig. 1, it shows that they have spherical surface morphology like an onion bulb and the main size of particles is 4–10 μm.

Table 1
Medium diameters (μm) of CIP and graphite.

Sample	Diameter (μm)	
	$D(\nu,0.5)$	$D(\nu,0.9)$
CIP	6.57	12.35
Graphite	18.98	47.37

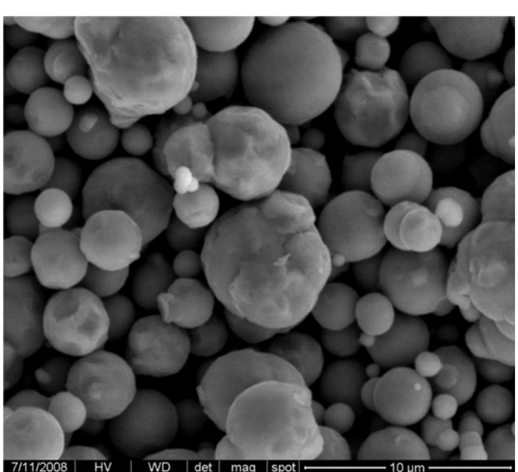

Fig. 1. SEM micrographs of carbonyl-iron powder.

Table 2
Chemical composition of carbonyl-iron powder.

Element	Weight (%)	Atomic (%)
Fe	97.74	90.29
C	2.26	9.71

The chemical composition in Table 2 reveals that the content of Fe in the CIP is above 97%.

3.4. SEM analysis of CIP-GRP/CSM

Fig. 2 shows SEM photographs of fractured cross-section and the surface of CIP-GRP/CSM composite, CIPs are uniformly dispersed in the polymer matrix and the graphite particles dispersed around the surface of the composite.

3.5. Reflectivity measurement

The results of reflection loss versus frequency for the CIP-GRP/CSM composites absorbers are illustrated in Fig. 3(a)–(d), for the composites with different thickness of 0.5 mm, 1.0 mm, 1.5 mm and 2.0 mm, respectively. For the composite microwave absorber of 0.5 mm in thickness, the RL decreases from −0.71 dB to −17 dB in the 2–18 GHz range. As showed in Fig. 3(b), the minimum RL is −22.64 dB at 14.8 GHz and the absorption range under −10 dB is from 12.04 to 16.84 GHz with a thickness of 1.0 mm. However, Fig. 3(c) shows that the minimum RL is −33.19 dB at 13.24 GHz and its bandwidth for RL under −10 dB is from 9.04 to 18 GHz with a thickness of 1.5 mm. From Fig. 3(d), the minimum RL and the bandwidth for RL under −10 dB are −32.37 dB at 10.84 GHz and 5.16 GHz. The minimum RL shifts to lower frequency with the thickness increases from 0.5 mm to 2.0 mm.

Fig. 4(a) shows the frequency dependence of the real μ' and the imaginary μ'' parts of the complex permeability of the CIP-GRP/CSM composite sample. The value of μ' decreases from 2.98 to 1.83 in the 2–18 GHz range with a small peak at 13.07–14.43 GHz and a platform at 14.43–18 GHz. μ'' increases from 0.529 to 1.21 in the 2–12.82 GHz frequency range and decreases to 0.64 at 16.64 GHz with a platform at 14.35–14.64 GHz range, and the value of μ'' increases to 0.76 at 18 GHz. It is note worthy that the maximum value of μ'' appearing at 12.82 GHz, which implies that the natural resonance occurred in the composites.

As shown in Fig. 4(b), the maximum value of the real part ε' and the imaginary part ε'' of the complex permittivity is 11.16 at 12.98 GHz and 3.82 at 13.24 GHz, respectively. It can be found that ε' begins to increase before ε'', and the maximum value of ε'' is corresponding to the degressive region of ε'. The dielectric properties of particles arise mainly due to space charge polarization and eddy current effect. According to the free electron theory[23], $\varepsilon'' \approx 1/2\pi\varepsilon_0\rho$, where ρ is the resistivity. It can be speculated that the lower ε'' values indicate a higher rersistivity with respect to other EM microwave absorbing materials, so the rersistivity of the CIP-GRP composite characterizes a sudden change.

The dissipation factors represented by the dielectric loss tangent ($\tan\delta_\varepsilon = \varepsilon''/\varepsilon'$) and magnetic loss tangent ($\tan\delta_\mu = \mu''/\mu'$) shown in Fig. 5. As can be seen from Fig. 5, the magnetic loss factor ($\tan\delta_\mu$) increases from 0.178 to 0.634 (the maximum value of $\tan\delta_\mu$) in the 2–12.82 GHz frequency range and quickly decreases to 0.363 at 13.75 GHz. Both the μ'' and the $\tan\delta_\mu$ come to the maximum value at 12.82 GHz where the composite absorber has the maximum magnetic loss for EM wave. The value of $\tan\delta_\mu$ shows a platform at 13.75–16.40 GHz