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Dielectric properties of carbon black and carbonyl iron filled epoxy–silicone resin coating

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Abstract The dielectric properties of epoxy–silicone resin coatings containing carbon black (CB) and carbonyl iron (CI) particles as a function of frequency (2–18 GHz) and the CB volume content (0.2–1%) have been investigated. The complex permittivity of the coatings increased with increasing CB content, which mainly attributed to the interfacial polarization at the CB/resin/CI particles interfaces. The complex permittivity also decreased rapidly with increasing frequency in the low frequency range while decreased slowly in the high frequency range. The changes of dielectric properties with frequency and the CB volume content were discussed using the power-law decay and the concept of interfacial polarization.

Introduction

Many interests have been focused on the multi-component heterogeneous composites with novel dielectric properties [1–3]. For instance, by combining different fibers or fillers with polymer matrix, the filled polymer composites possess some special microwave electromagnetic properties, which can be utilized as radar absorbers and electromagnetic wave shields [4–6].

It is well known that the carbon black (CB) filled polymers have very interesting applications. Choi et al. [7] have reported the effects of frequency, volume fraction of CB, and porosity on the complex permittivity of the CB/epoxy composites. Recently, Achour et al. [8] have reported the effective complex permittivity for series of CB/epoxy composites at ambient temperature and in the frequency range from 200 Hz to 15 MHz, and provided a detailed and quantitative characterization of the dielectric relaxation behavior. Carbonyl iron (CI) also has been used as a component of electronic devices such as plastic encapsulated inductor cores and electromagnetic wave absorbers [9]. Abshinova et al. [10] have investigated the electromagnetic properties of polymer composites based on different types of CI in the frequency range from 1 MHz to 10 GHz. The results show that the microstructure and shape of CI particles have a crucial effect on the absolute values and character of the electromagnetic properties for the CI filled polymer composites.

The coatings in this study consist of insulating resin matrix (epoxy–silicone resin), conductive CB, and magnetic CI particles. This work is mainly to investigate the dielectric properties of the filled polymer coatings in the frequency range from 2 to 18 GHz. The complex permittivity of the coatings with different CB volume content was analyzed based on the power-law decay theory and the concept of interfacial polarization.

Experimental procedures

The microwave absorbing coating contained 50 vol% CI particles and 0.2, 0.6, and 1 vol% CB was prepared as followed. First, the CB was dispersed in the ethanol solution by an ultrasonic bath at room temperature for 1 h. After mixing the CI particles into the ethanol-based solution, the suspensions were stirred for 10 min at 2000 rpm. Then, the mixtures were placed in an oven at 80 °C in order to evaporate the ethanol. After adding the resin and hardener, the mixtures were stirred at 2000 rpm for

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10 min. Finally, the hybrid mixtures were pre-cured at 90 °C for 30 min and post-cured at 120 °C for 2 h.

The morphology of the coatings was observed using scanning electron microscopy (SEM) (Model JSM-6360, JEOL, Tokyo, Japan). The complex permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) of the coatings were measured by the *T*/*R* coaxial line method in the frequency range from 2 to 18 GHz using a network analyzer (Agilent technologies E8362B: 10 MHz–20 GHz). The testing specimens have a cylindrical toroidal specimen with outer diameter of 7.0 mm and an inner diameter of 3.03 mm.

Results and discussion

Figure 1 gives the microstructure of the epoxy–silicone resin coating with CB and CI particles. It is shown that the CB and CI particles were uniformly dispersed in the epoxy–silicone resin.

Figure 2 shows the complex permittivity spectra of the coatings containing CB and CI particles. The complex permittivity spectra revealed the dielectric relaxation of the coatings. Both the real and imaginary part of permittivity increased as the CB volume content increased. For a given CB volume content, both real and imaginary part of permittivity decreased rapidly with increasing frequency in the lower frequency range while decreased slowly in the high frequency range. It is evident that the complex permittivity of the coatings possessed frequency-dependent dielectric response.

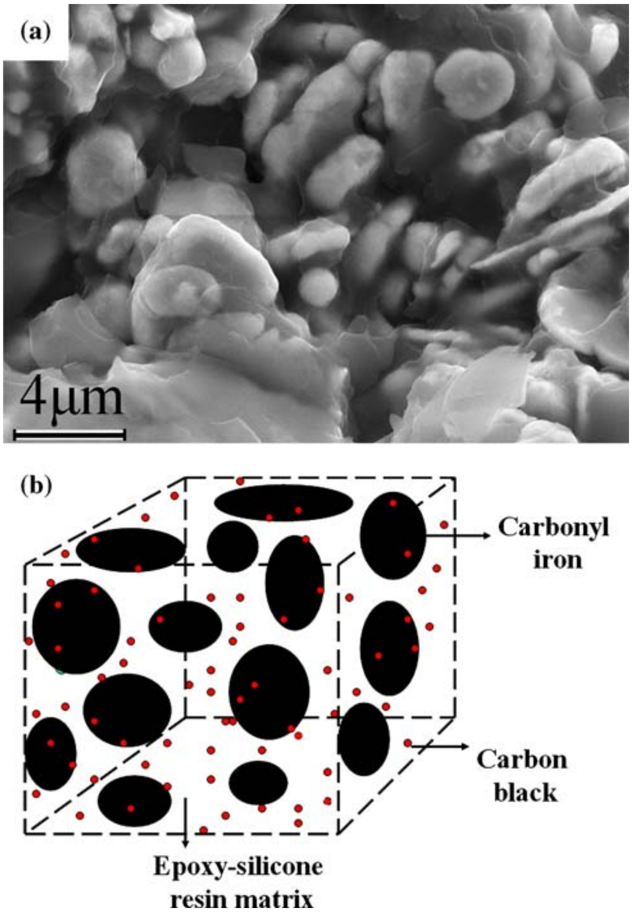

Fig. 1 **a** SEM image and **b** conceptual picture of the epoxy–silicone resin coating with CI and CB

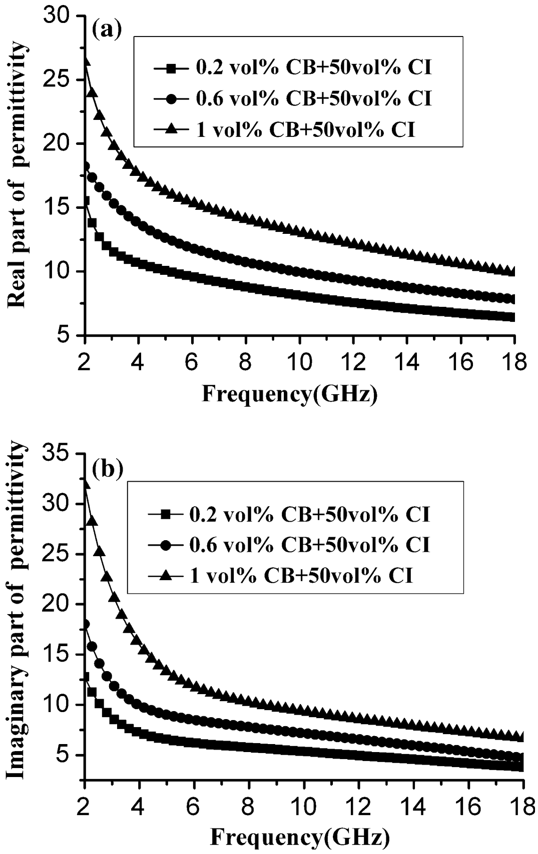

Fig. 2 Frequency dependency of the complex permittivity for epoxy–silicone resin coating (CB content increases from 0.2 to 1 vol% and the content of CI kept constant)

In general, the dielectric properties of conductive particles filled insulation resin coatings depend on the characteristics of the matrix, property and volume fraction of the filler, configuration and internal fractal structure of the composites, and frequency of the electromagnetic wave [11–13]. It is well known that the ε' of composite is proportional to the quantity of charge stored on the surface when the composite was submitted to an applied electric field [14]. As the CB volume content increased, the CB/resin/CI particles interfaces increased, as shown in the Fig. 1b. Thus, a great deal of charge forms on the CB/resin/CI particles interfaces due to the interfacial polarization. From this viewpoint, the higher real part of permittivity can be obtained when the coatings are with higher CB volume content. When the conducting CB is separated by highly resistive epoxy–silicone resin and CI particles, the main term affecting the ε'' is the interfacial polarization and its associated relaxation while the conductivity term plays a secondary role [15]. It is reasonable that the ε'' of the composites increased as the CB content increased, and showed a similar tendency with the ε' of the composites. This phenomenon can also explained by Maxwell–Wagner–Sillars theory, which is to account for the dielectric loss due to the interfacial polarization of heterogeneous materials having the volume fraction of conductive filler lower than the percolation threshold [16, 17]. Furthermore, because the interfacial polarization can occur at lower frequency more easily, the production of displacement current significantly lags behind the build-up potential as the frequency increased. Thus, both ε' and ε'' show the frequency dependence in the whole frequency range (2–18 GHz) [18].

Conventionally, concerning the dielectric relaxation in the frequency domain, the complex permittivity can be fitted empirically by power-laws

$$\varepsilon' - \varepsilon'_{\infty} \propto F^{-b'}, \quad \varepsilon' \propto F^{-b''} \tag{1}$$

where ε'_{∞} denoted high frequency permittivity and $0 < b < 1$ [19, 20]. According to the Jonscher theory [21], the plot of real part of permittivity ε' versus imaginary part of permittivity ε'' would be a straight line on a linear presentation, which is usually defined as the Cole–Cole representation. In Fig. 3a, we display the real and imaginary part of the complex permittivity as a function of frequency for the epoxy–silicone resin coating containing 0.2 vol% CB and 50 vol% CI particles. It can be seen that ε' and ε'' have nearly parallel variations in the high frequency range. Similar results (not shown) can be obtained when the CB volume content is 0.6 and 1%. As the inset of Fig. 3a clearly shows the Cole–Cole plot of ε' versus ε'' is almost linear, which is consistent with a fractional power-law response. Figure 3b shows the fitted values of the b' and b'' with different CB volume content (0.2, 0.6, and 1%), which was calculated using the results of Fig. 2. When the CB volume content is 0.2, 0.6, and 1%, the values of the b' is 0.36, 0.37, and 0.39 and the b'' is 0.45, 0.49, and 0.63, respectively. As the foregoing discussion, both of the ε' and ε'' shows much stronger frequency dependence as the CB volume content increased. Therefore, it is reasonable that both the b' and b'' increased with the increasing CB volume content.

Conclusions

The dielectric properties of CB and CI particles filled epoxy–silicone resin coatings were investigated in the frequency range from 2 to 18 GHz. As the volume content of CB increased, both the real and imaginary part of permittivity increased, and showed nearly parallel variations in the high frequency range. For a given CB and CI particles content, both real and imaginary part of permittivity decreased rapidly with increasing frequency in the lower frequency range and decreased slowly in the high frequency range. The changes of dielectric properties with frequency and the CB volume content could be explained using the power-law decay and the concept of interfacial polarization.

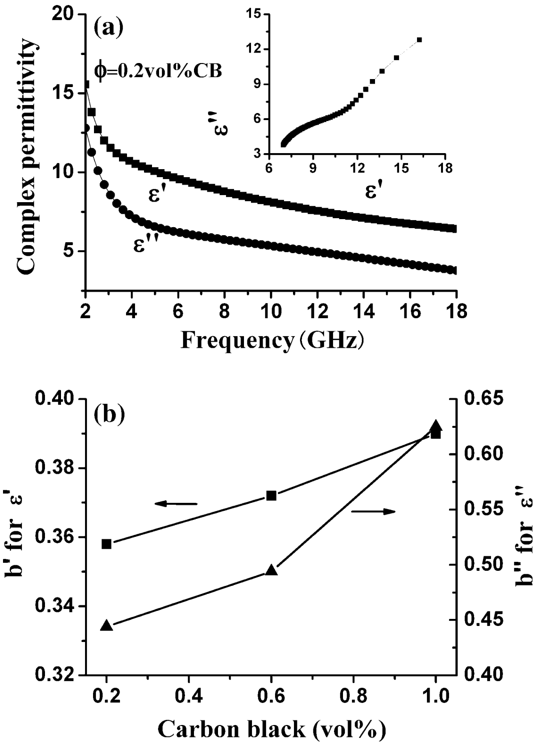

Fig. 3 **a** The real and imaginary part of the complex permittivity versus frequency, for the CB content is 0.2 vol%. The inset shows the Cole–Cole plot of the complex permittivity. **b** Fitted values of the b' and b'' versus the CB content

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