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Three-dimensional graphene network/phenolic resin composites towards tunable and weakly negative permittivity

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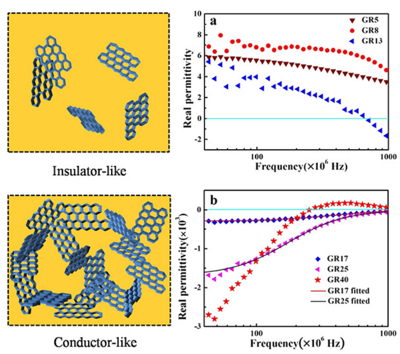
HIGHLIGHTS

- Weakly negative permittivity was first obtained from the graphene composites.
- Negative permittivity resulted from formation of graphene conductive networks.
- Negative permittivity was easily adjusted by controlling the graphene contents.
- High dielectric loss (above 25) was obtained from 13 vol% graphene.

GRAPHICAL ABSTRACT



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ABSTRACT

Three-dimensional (3D) graphene (GR) network/phenolic resin composites with tunable and weakly negative permittivity were prepared by mechanical mixed method. Dielectric properties including permittivity, dielectric loss tangent and alternating current conductivity (σ_{ac}), were investigated in detail. When the GR content was increased from 8 to 13 vol%, a ladder-shape increase of σ_{ac} was observed, indicating a percolation phenomenon. It was found that, when the GR content exceeded 13 vol% the negative permittivity appeared attributed to the formation of 3D interconnected GR network. Moreover, the negative permittivity was easily adjusted by controlling the GR content. Compared with the metal-ceramics in our previous work, the absolute values of negative permittivity were low, which was in favor of impedance matching. For the GR content of 13 vol%, high dielectric loss above 25 over the whole frequency was also observed. Finally, the equivalent circuits were used to analyze the reason of the negative permittivity.

1. Introduction

It was well known that permittivity and permeability are important parameters affecting the interaction between electromagnetic wave and materials. Recently, a new kind of material with negative permittivity and permeability simultaneously, called double negative material (DNM), has attracted much attention due to its novel electromagnetic properties, such as inverse Doppler effect, inverse Cerenkov radiation, negative refractive index [1–8]. In 2000, Smith et al. [9] first obtained the double negative property in an artificial structure with periodic arrangement of metallic wires. After that, many methods based on artificial construction have been proposed to produce DNMs [10, 11]. Recently, it has been reported that double negative property also

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can be obtained in random composites without periodic structures [12, 13]. Compared with the artificial periodic structure, the random composite shows its own advantages, such as controlled microstructures and tunable dielectric properties.

Due to these advantages, random composites not only have been used to produce the DNM but also the epsilon negative material (ENM). ENM with interesting dielectric properties shows potential applications in electromagnetic interference shielding or absorbing, capacitors and other fields [14–16]. Secondly, ENM is further expected for preparation of DNM. Currently, the ENM mainly has been produced from metal-ceramics or carbon composites. For instance, Fan et al. [17] have prepared the random Cu/YIG composites with negative permittivity by *in situ* synthesis. Values of negative permittivity from the metal-ceramic composites usually are high, which is difficult for the impedance matching. It has been reported that the carbon composites including carbon nanotubes or nanofibers have weakly negative permittivity [18–21]. Zhong et al. [20] have reported the weakly negative permittivity in the carbon nanofiber/polyetherimide composites. Our group recently has prepared carbon nanotubes/phenolic resin composites to attain the weakly negative permittivity [21]. Although the weakly negative permittivity has been observed in these works, the corresponding mechanism, regulatory, and dielectric loss have not been investigated in detail.

Graphene (GR) exhibits good two-dimensional sheet structure, high electron mobility and large surface area, so it is often used as an additive to improve mechanical and thermal properties of the composite

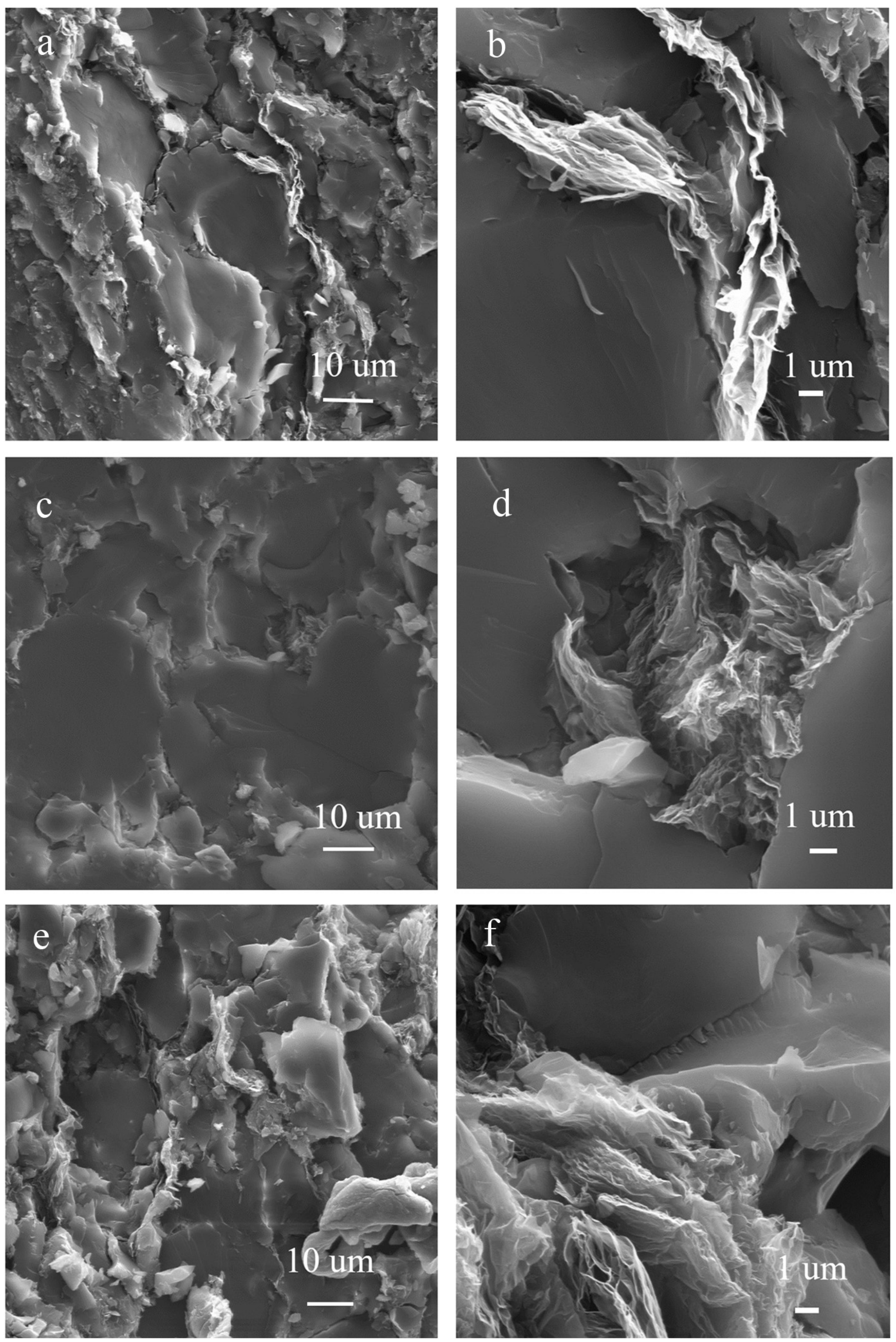

Fig. 1. FESEM images of the GR/PR composites with different GR contents. (a) and (b) GR8, (c) and (d) GR13, (e) and (f) GR17. (b), (d) and (f) were the corresponding magnification images.