



Graphene encapsulated rubber latex composites with high dielectric constant, low dielectric loss and low percolation threshold



Ming Tian^{a,b}, Jing Zhang^b, Liqun Zhang^{a,b}, Suting Liu^b, Xiaoqing Zan^b, Toshio Nishi^c, Nanying Ning^{a,b,*}

^a State Key Lab of Organic–Inorganic Composites, Beijing University of Chemical Technology, Beijing 100029, China

^b Key Laboratory of Carbon Fiber and Functional Polymers, Ministry of Education, Beijing University of Chemical Technology, Beijing 100029, China

^c Department of Applied Physics, The University of Tokyo, Hongo, Bunkyo-ku, Tokyo, Japan

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ABSTRACT

A dielectric composite with high dielectric constant, low dielectric loss and low percolation threshold was prepared by using the combined strategy of encapsulating of graphene oxide nanosheets (GONS) on carboxylated nitrile rubber (XNBR) latex particles and the in situ thermal reduction in GONS at a moderate temperature. The encapsulation of GONS on XNBR latex particles was mainly realized via the hydrogen bonding interactions between GONS and XNBR during latex mixing. A segregated graphene network was obtained at a low content of thermally reduced graphene (TRG), resulting in a low percolation threshold (0.25 vol.%). The dielectric constant at 100 Hz obviously increased from 23 for pure XNBR to 2211 and 5542 for the composite with 0.5 vol.% and 0.75 vol.% of TRG, respectively. The dielectric loss of the composites retained at a low value (less than 1.5). Meanwhile, the elastic modulus only slightly increased with the presence of 0.1–0.5 vol.% of TRG, keeping the good flexibility of the dielectric composites. This study provides a simple, low-cost and effective method to prepare high performance dielectric composites, facilitating the wide application of dielectric materials.

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1. Introduction

Dielectric elastomers (DEs), as a kind of electro-active polymers (EAPs), can convert electric energy to mechanical energy without gearing and work efficiently over a broad frequency range [1–3]. Owing to its low modulus, large strain, fast response, lightweight, reliability, high energy density, and high electromechanical coupling efficiency, DEs find many applications in industry such as artificial muscles, sensors, micro-air vehicles, flat-panel speakers, micro-robotics and responsive prosthetics [4–6].

A disadvantage for common DEs is that the dielectric constant (k) is very low (less than 10). Thus, a key issue is to increase the k of DEs while retaining other excellent properties such as low dielectric loss, and good flexibility. One common method to improve the k of DEs is to introduce high-dielectric-constant ceramic powders such as BaTiO₃ [7], and TiO₂ [8] into the elastomer matrix. In this case, a high content (up to 50 vol.%) of ceramics is required to effectively improve the dielectric constant, resulting in high elastic modulus, low flexibility, poor processability, all of

which limit the wide application of DEs. Another method is to prepare percolative composites by adding conductive fillers such as carbon nanotubes (CNTs) or graphene sheets into the matrix [9–11]. Comparing with CNTs, graphene sheet is usually synthesized from natural graphite, which is cheap and easily available. More importantly, graphene sheet has unique layered structure with a larger aspect ratio, and thus has been considered to be an ideal candidate as high k filler.

Some studies have been carried out to increase the dielectric properties of polymer by introducing graphene oxide nanosheets (GONS) into the matrix [9,11,12]. For instance, Seveyrat et al. [12] reported that the k of polyurethane (PU) composites filled with GONS was increased from 8 for pure TPU to 60 at 1000 Hz near the percolation thresholds (7.2 vol.%). In addition, Romasanta et al. [11] reported that the k of poly(dimethyl) siloxane (PDMS) increased from 2.7 to 23 at low frequency (10 Hz) by adding 2.0 wt.% of thermally expanded graphene sheets using melt mixing method. Obviously, the increase in k of these polymers by adding graphene sheets is far less than expected. The major reasons are the poor dispersion of graphene in matrix, the restacking of graphene sheets during preparation, and/or weak interfacial adhesion between graphene sheets and polymer matrixes [11,12]. Recently, an interesting work was reported by Jiang et al. [13]. They first prepared a hyperbranched aromatic polyamide functionalized

* Corresponding author at: State Key Lab of Organic–Inorganic Composites, Beijing University of Chemical Technology, Beijing 100029, China. Fax: +86 10 6443 3964.

E-mail address: ningny@mail.buct.edu.cn (N. Ning).

graphene sheets (GS–HBA) and then introduced the as prepared GS–HBA into the thermoplastic polyurethane (TPU) matrix to increase the dielectric properties of TPU. As a result, a uniform dispersion of GS–HBA in TPU matrix and a strong adhesion between GS–HBA and TPU were obtained. The k of the composite with 5 wt.% GS–HBA at 1000 Hz was largely increased to 850, and the dielectric loss retained at a low value (less than 1.5) owing to the suppression of the leakage current by hyperbranched chains.

The dielectric constant of percolative composites can be dramatically enhanced when the volume fraction of the dielectric filler increases to the vicinity of the percolation threshold [14]. Therefore, the formation of dielectric filler network at low percolation threshold is a key to obtain dielectric composites with high dielectric constant. As has been reported in some studies, polymer composites with three-dimensional (3D) segregated filler network can be prepared by distributing CNTs or carbon black at the boundary between the polymer particles [15,16]. These composites with segregated network structure have much lower percolation threshold than the conventional composites with randomly distributed conductive fillers. This motivated us to construct a segregated graphene network in a rubber matrix to prepare dielectric composites with high dielectric constant at lower filler content. Fortunately, an interesting work has been conducted by Xia et al. to obtain natural rubber composites with segregated graphene network by using latex mixing [17]. It was found that the electrical conductivity, water vapor permeability and mechanical properties of the as-prepared composites were largely improved. To the best of our knowledge, high performance dielectric composites with a segregated network structure have not been reported yet.

There exists large number of oxygen-containing groups (C–O–C, C–OH, and C=O) on GONS [18]. These oxygen-containing groups enable GONS to uniformly disperse in rubber latex by ultrasonic, facilitating the preparation of GONS dielectric composites

with segregated network structure. However, the oxidation of graphite severely disrupts the graphite structure, resulting in a decrease in dielectric constant [19,20]. Therefore, reduction in GONS is required to obtain a DE composite with both high k and low dielectric loss. In most cases, the reduction in GONS was realized via chemical reduction by using hydrazine hydrate [21], Vitamin C [22], etc., resulting in a high reduction degree of GONS, and thus a good electric conductivity of the composites. A disadvantage of chemical reduction method is the high dielectric loss of the composites caused by the large direct current (DC) conductance [13]. In recent years, several studies have found that in situ thermal reduction in GONS at a moderate temperature (180–200 °C) within a polymer is a simple and effective technique for the moderate reduction in graphitic structure [23,24]. This motivates us to ask the following question: Can we simultaneously increase the interfacial polarization ability of GONS and keep a low DC conductance of the composites by the moderate reduction in graphitic structure using the in situ thermal reduction method to obtain dielectric composites with both high k and low dielectric loss?

Therefore, in this study, we aim to prepare dielectric composites with high dielectric constant, low dielectric loss and low elastic modulus at low filler content by the encapsulating of GONS on the surface of rubber latex particles and in situ thermal reduction in GONS at 180 °C for 2 h. Carboxylated nitrile rubber (XNBR) containing a large amount of strong polar groups (carboxyl groups and cyano groups), has a high dielectric constant (>10), good mechanical strength, good oil and solvent resistance, and thus XNBR latex was used as the elastomer matrix. A homogeneous aqueous suspension of GONS and carboxylated nitrile rubber (XNBR) latex was first obtained through ultrasonically-assisted latex mixing. The encapsulation of GONS on the surface of XNBR rubber particles is expected during mixing process because of the hydrogen bonding between –COOH groups on XNBR and oxygen groups on GONS.

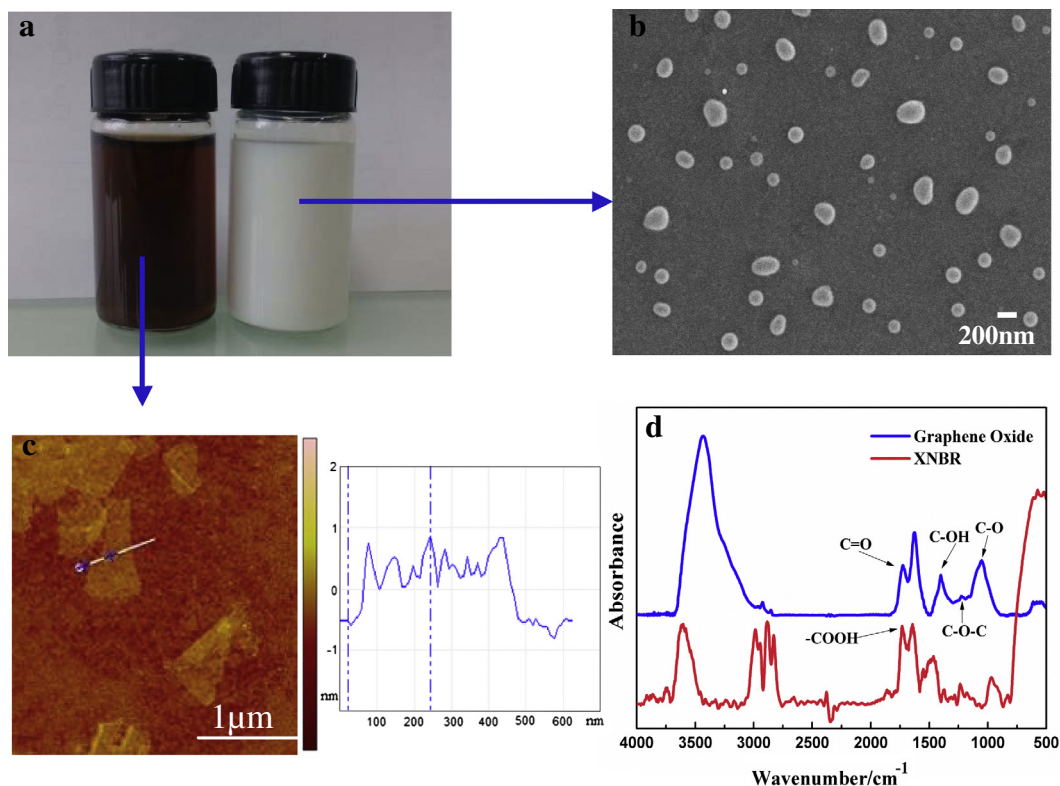


Fig. 1. (a) Digital photograph of GONS aqueous solution (left) and XNBR particles suspension (right); (b) SEM image of XNBR particles; (c) AFM image (left) and cross-section (right) of GONS nanosheets (position of cross-section in the image indicated by white, straight line); (d) FT-IR spectra of XNBR particles and GONS.

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