



Novel three-component nanocomposites with high dielectric permittivity and low dielectric loss co-filled by carboxyl-functionalized multi-walled nanotube and BaTiO₃

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ABSTRACT

In this work, a novel ternary polydimethylsiloxane (PDMS)-based nanocomposite co-filled by carboxyl-functionalized multi-walled nanotube (cMWCNT) and BaTiO₃(BT) nanoparticle were developed. The hybrids cMWCNT-BT was realized via the hydrogen bonding (H-bonding) interaction between –COOH groups of cMWCNT and the –OH groups of BT. Compared with the binary composite of BT/PDMS and cMWCNT/PDMS, the results showed that the kind of three-component nanocomposites had high dielectric permittivity and low dielectric loss. Different from cMWCNT, the hybrids cMWCNT-BT have good dispersion in PDMS matrix, forming many micro-capacitors and interfaces, and thus leading to remarkably high dielectric constant. Meanwhile, BT on the surfaces of cMWCNT reduce the direct contact of cMWCNT, resulting in reduced dielectric loss. More important, the BT and cMWCNT had a synergistic effect on dielectric permittivity of the cMWCNT-BT/PDMS composite. Therefore, the dielectric properties of the novel ternary composite can be improved by optimizing the synergistic effects between the charge storage behavior of the ferroelectric phase and the charge transport behavior of the conductive phase.

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1. Introduction

High-permittivity (high-*k*) polymer materials are highly desirable for their applications in the modern information and electronics industry, such as charge-storage capacitors, artificial skins and muscles, and flexible electronics because of their intrinsic advantages of flexibility, durability, easy processing and lightness [1–5]. Various strategies have been developed to fabricate high-*k* PMCs. One common approach in high-*k* polymer composites is to incorporate ceramic fillers into a polymer matrix which called as ceramic/polymer composites [6–8]. BaTiO₃(BT) as one of the most commonly used high-*k* fillers has been extensively studied for a variety of polymeric matrices including polymethylmethacrylate (PMMA) [9], polyimides [10] and epoxy resins [11]. However, there are also some challenges to be addressed [12]. First, the interfacial incompatibility between inorganic filler and polymer matrix

provides a path for the space charge migration and accumulation within the nanocomposites. The second, blending of the inorganic additive and the polymer matrix often results to filler particle aggregation [13]. A second common approach to increase the dielectric permittivity of these materials is to incorporate conductive fillers in the polymer matrix which called as electric conductor/polymer composites. Based on percolation theory, the use of conducting fillers can not only increase conductivity but also induce Maxwell-Wagner polarization. As a result, the polarization leads to a high dielectric permittivity [1]. Conversely, the addition of conductive fillers or highly polarizable conjugated polymers results in extremely high dielectric loss, which is difficult to control, because the particles can easily form a conductive path and produce local current in the composite as the filler concentration approaches the percolation threshold [3]. In this condition, transformation of dielectric loss to thermal energy occurs, followed by the reduction of dielectric strength and electromechanical transduction efficiency [14,15]. Therefore, both of ceramic/polymer and electric conductor/polymer composites have disadvantages. All these effects negatively affect the applications of high-*k* polymer materials. To overcome these shortcomings, many approaches have

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been considered including surface modification of carbon nanotubes (CNTs) [16,17] and BT [18,19], as well as advanced fabrication of core-shell structures [20,21].

In recent years, incorporate ceramic fillers and conductive fillers to prepare conductor/ceramic/polymer ternary composites has become one of effective way to obtain high- k . Yu et al. [22] added different contents of CNTs into BT/polypropylene (PP) composite with 40 wt% BT. A novel three-phase nanocomposites was prepared with PVDF as the polymer matrix, BT nano-powders were used as the high dielectric permittivity fillers. Specially, nano-Ag fillers were added into the BT/PVDF nanocomposites [23]. K. Hayashida have proposed an advanced composite system of poly(cyclohexyl methacrylate) grafted on multi-walled carbon nanotubes (PCHMA-CNT) and BT powder [24]. Functional polypropylene (PP) nanocomposites were prepared by melt compounding with multi-walled carbon nanotubes (MWNT) as the electrically conductive component and BT spherical nanoparticles as the ferroelectric component [25]. D. R Wang investigated the fabrication and dielectric properties of a novel multi-component high- k composite system consisting of poly(vinylidene fluoride)(PVDF), surface-functionalized graphene nanosheets and BT nanoparticles (rGO-BT/PVDF) [26]. However, to our knowledge, a study concerning the fabrication and performance of the multi-component high- k composite system consisting of carboxyl-functionalized multi-walled nanotubes (cMWCNT) and BT has not yet been reported in the literature. In this work, the novel ternary high- k composite based on polydimethylsiloxane (PDMS) matrix co-filled by cMWCNT and BT were developed. Herein, we first prepared cMWCNT-BT hybrid via hydrogen bonding by simply mixing the cMWCNT and BT suspension without using any modifier or surfactant. The relationship between the microstructure and properties of the composite is comprehensively analyzed. Results indicate that the cMWCNT-BT hybrid in the composites exhibit significant and synergistic effects on the dielectric properties of the composite system. The results of this research can contribute to the fabrication of composites with high dielectric permittivity and low dielectric loss.

2. Experimental

2.1. Materials

The PDMS (Sylgard 184) prepolymer and curing agent were supplied by Dow Corning Corporation. Carboxyl-functionalized multi-walled nanotubes(cMWCNT) were purchased from Chengdu Organic Chemicals Co. Ltd., Chinese Academy of Sciences. BaTiO₃(BT) ceramic powder (average diameter 100 nm), were purchased from Aladdin. Ethyl acetate, chloroform and other chemicals used in the experiments were used as received.

2.2. Preparation of the composites

2.2.1. Preparation of the BT/PDMS composites

The BT/PDMS composite with 1.53 vol% BT was prepared via the solution method in order to get a good dispersion of BTs in rubber matrix. First, BT was dispersed in ethyl acetate and stirred for 2 h in order to form a stable suspension, then the BT suspension was ultrasonically vibrated for 30 min at room temperature. After that, the BT suspension was added into the Sylgard 184 pre-polymer followed by careful stirring. Subsequently, Sylgard 184 curing agent was added into the mixture and the ratio of curing agent to Sylgard 184 pre-polymer is 1:10. Then the final solution was carefully mixed for 10 min, cast onto a Teflon mold, and cured at 80 °C for 24 h to form a thin film. The thickness of the BT/PDMS film was controlled by changing the amount of the solution poured onto the mold.

2.2.2. Preparation of the cMWCNT/PDMS composite

Similarly, 0.45%vol cMWCNT was added to PDMS to fabricate the cMWCNT/PDMS composite. First, cMWCNT was dispersed in chloroform and stirred for 2 h in order to form a stable suspension, then the cMWCNT suspension was ultrasonically vibrated for 30 min at room temperature. After that, the cMWCNT suspension was added into the Sylgard 184 pre-polymer followed by careful stirring. Subsequently, Sylgard 184 curing agent was added into the mixture and the ratio of curing agent to Sylgard 184 pre-polymer is 1:10. Then the final solution was carefully mixed for 10 min, The mixture solution was concentrated with heating stirring in order to get rid of the solvent quickly and thus can effectively prevent the aggregation of MWCNT. and cured at 80 °C for 24 h to form a thin film.

2.2.3. Preparation of the cMWCNT-BT/PDMS composites

0 vol%, 0.04 vol%, 0.13 vol%, 0.18 vol%, 0.27 vol%, 0.33 vol%, 0.45 vol%, 0.89 vol%, 1.34% cMWCNT with 1.53 vol% BT were added to PDMS separately to fabricate the cMWCNT-BT/PDMS composites. Firstly, BT was dispersed in ethyl acetate and stirred for 2 h to form a stable suspension. The resulting BT suspension, denoted as A, was ultrasonically vibrated for 30 min at room temperature. cMWCNT were dispersed in chloroform and stirred for 2 h to form a stable suspension, which was then ultrasonically vibrated for 30 min at room temperature. The cMWCNT suspension was denoted as B. The A and B suspensions were both added into the PDMS prepolymer. A curing agent was then added to the mixture to a concentration of 10 wt.% with respect to the total weight of the PDMS prepolymer. The resulting cMWCNT-BT/PDMS solution was carefully stirred for 10 min, cast onto a Teflon mold, and cured at 80 °C for 24 h. The thickness of the film was controlled by changing the amount of the solution poured onto the mold. Fig. 1 shows the preparation process of the cMWCNT-BT/PDMS composites. Therefore, the binary composites BT/PDMS and cMWCNT/PDMS and ternary cMWCNT-BT/PDMS composite were all prepared via solution coating. For the as-prepared composites, before curing, the mixture solution was first concentrated with heating stirring in order to get rid of the solvent quickly which can effectively prevent the aggregation of cMWCNT during cure. When most of the solvent was removed from the polymer matrix, the cMWCNTs were immobilized in the PDMS matrix, Therefore, uniform dispersion and distribution of cMWCNTs in PDMS matrix can be obtained.

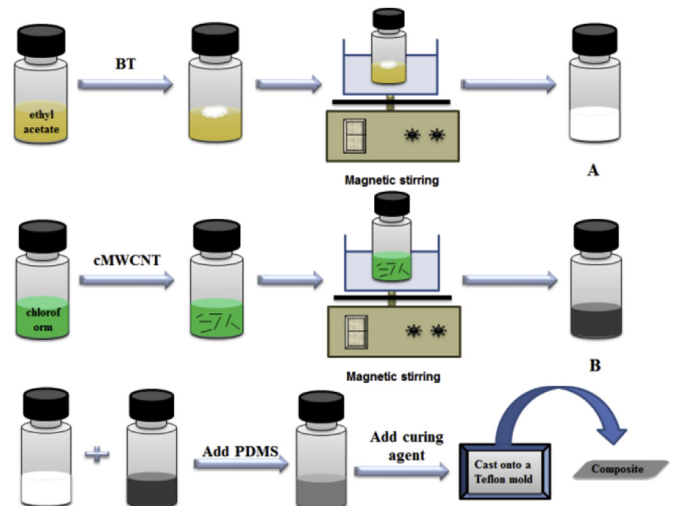


Fig. 1. The preparation process of the cMWCNT-BT/PDMS composites.

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