



Ductile polymer-based films with ultrahigh permittivity and low dielectric loss



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ABSTRACT

High-permittivity (high- k) materials play a key role in advanced electronics and electrical power systems. How to prepare the flexible material with high permittivity and low dielectric loss turns out to be a hard work due to the essential low permittivity of polymers. In this work, a kind of ductile polymer-based composites was fabricated by incorporating ionic liquids into polymer matrices. The optimized permittivity of composite with 50 wt% ionic liquid reached up to 3.3×10^4 , while the dielectric loss was as low as 0.65. The temperature dependence of dielectric properties was also investigated, and a high permittivity as well as low dielectric loss at 100 Hz of about 2.0×10^4 and 0.23 were achieved at 80 °C, respectively. Furthermore, the study reveals that the dielectric loss peaks of the composite films can be affected by the content of ionic liquid, temperature even the size of ions. In short, the finding indicates that the addition of ionic liquids into polymers provides a big opportunity to improve dielectric properties of polymer materials.

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1. Introduction

Functional high-permittivity (high- k) materials are of important significance for the electronic and electric power industries [1–5]. Higher requirements have been promoted to fabricate miniaturized, light weight and ductile electronic equipment. Some new applications, such as actuators and bionics, also ask for high- k materials with excellent flexibility [6,7]. Ceramic-based dielectric materials with high permittivity (about 10^4 – 10^5) cannot be applied into these fields, due to their obvious brittleness, high density and rigorous preparing process [8]. Thus, scientists designed polymer-based high- k materials for the outstanding ductility of polymers. However, compared with the ultrahigh permittivity of ceramics, the inherent low permittivity (about 2–10) of most polymers hinders the development of high- k polymer-based dielectrics [1]. To solve this problem, high- k fillers/polymer composite strategy has been carried out to fabricate ductile dielectric materials [2,9]. The

problem is that the incorporation of high- k ceramic particles only can promote the permittivities of composites from tens to hundreds, which are still much less than those of ceramic-based dielectric materials [9]. The conductive fillers, such as Ag [10,11], carbon nanotubes [12,13] and graphene [14,15] etc. also have been used to improve the dielectric performances of polymers on the basis of percolation theory. Although the permittivities of conductive fillers/polymer composites dramatically increase to hundreds or even thousands when the content of conductive fillers is over the percolation threshold, the dielectric losses also greatly rise up to tens or even more than one thousand due to the booming leakage current from the formation of conducting paths [16]. What's more, it often takes a long time and complicated work to modify the inorganic fillers for better compatibility with polymer matrix.

Actually, ionic liquids (ILs) show high permittivities at room temperature such as 1-Methyl-3-octylimidazolium hexafluorophosphate [17] and 1-butyl-3-methylimidazolium tetrafluoroborate [18] (about 10^5 – 10^6 at 100 Hz). Recently, ILs are becoming superstars in electrochemistry, physical chemistry, nanotechnology, chemical reactions and purifications, and they are usually used as electrolytes or powerful solvents due to the high

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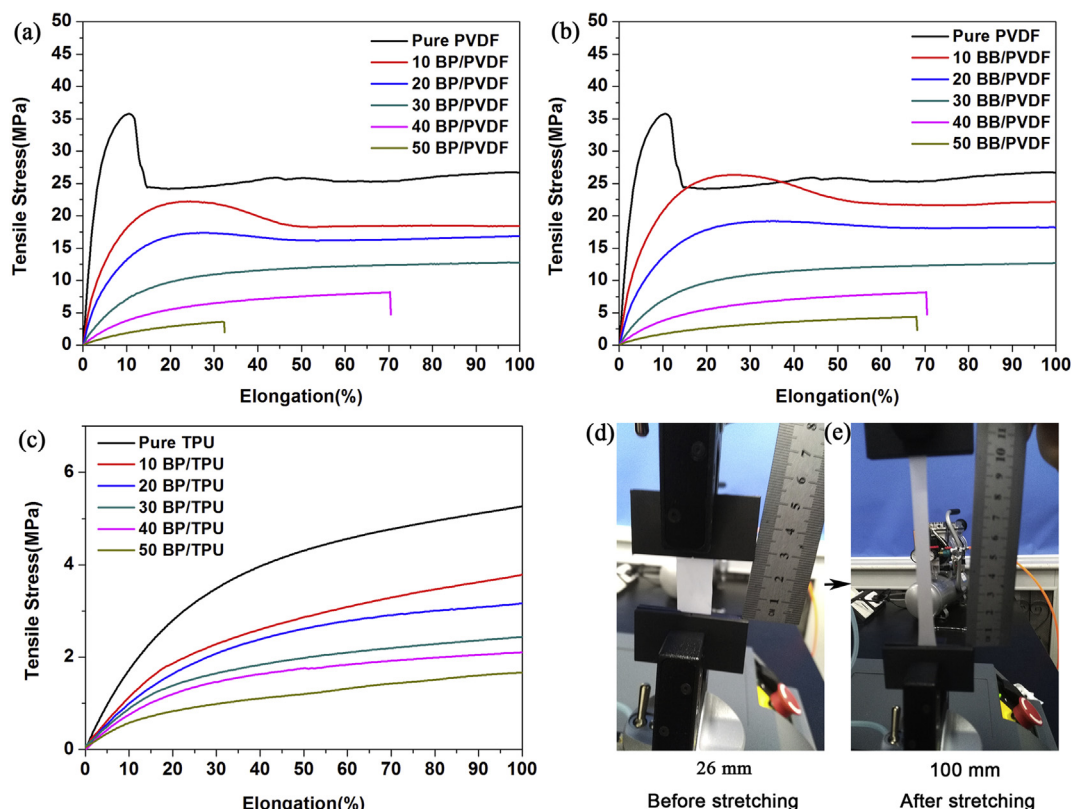


Fig. 1. The stress-strain curves of (a) BP/PVDF, (b) BB/PVDF and (c) BP/TPU composite films with different IL loading; the pictures of 50 BP/TPU composite film (d) before stretching and (e) after stretching.

electric conductivity, wide electrochemical window, high chemical and thermal stability [19–21]. The excellent electric conductivity of ILs composing of free ions and ion pairs are organic salts stems from the combination of ionic character and fluidity [20,22]. Although ionic conductivities of IL/polymer composites have been carefully investigated [23], the unique dielectric behaviors with high permittivity and low dielectric loss are ignored. Actually, the effect of IL in dielectric materials has already been reported, for example, Kim et al. found that the addition of IL not only improved the dispersion state of CNTs in polydimethylsiloxane (PDMS) but also lowered the Young's modulus of CNTs/PDMS composites, and even a low loading of 8 wt% IL could significantly promoted the dielectric properties of composites [24]. In addition, IL was also applied to decorate reduced graphene oxide (rGO) through $\pi_{\text{cation}}-\pi$ interaction by Gu and Liang to fabricate the IL-rGO/cyanate ester composites, and the results displayed a huge promotion of dielectric properties for composites comparing with the as-prepared rGO/cyanate ester composites [25].

In this manuscript, two polymers were applied as matrices including polyvinylidene fluoride (PVDF) and thermoplastic polyurethane (TPU). PVDF is famous for its excellent piezoelectric and pyroelectric properties while it is poled under an electric field, and it is also a popular candidate as a dielectric polymer matrix due to its relative high permittivity about 10–12 [26]. TPU as a common dielectric elastomer is also introduced in this work, which usually is applied in the actuator and electric generation fields [27].

Two ILs were used as high- k fillers to promote the dielectric properties of polymers containing 1-Butyl-3-methylimidazolium hexafluorophosphate (BP) and 1-butyl-3-methylimidazolium tetrafluoroborate (BB). And the results demonstrated that the permittivities of IL/polymer composites were greatly improved, while

the dielectric loss showed an amusing tendency at the range from 10^2 Hz to 10^7 Hz.

2. Experimental section

2.1. Fabrication of IL/polymer composite films

Both of the ILs (BP and BB) were purchased from Shanghai Chengjie Chemical co., LTD; PVDF (KF-850, Kureha Chemicals, Japan) and TPU (9380A, Bayer, German, shore A = 90) were provided by Guanhua plastic raw materials co., LTD. Firstly, using the fabrication of BP/PVDF composite films as an example, a series of different weight fractions of BP (0 wt%, 10 wt %, 20 wt %, 30 wt%, 40 wt% and 50 wt%) blending with PVDF in *N,N*-Dimethylformamide (DMF) were stirred in beakers at 50 °C for 2 h till both BP and PVDF were absolutely dissolved in DMF. After that, the solution was poured onto a piece of glass and a scraper was applied to carefully control the thickness of films, Finally the films were dried in the oven for 10 h at 80 °C to remove the solvent. For convenience to discuss, we named the composites films with different weight fractions of BP (0 wt%, 10 wt %, 20 wt %, 30 wt%, 40 wt% and 50 wt%) as pure PVDF, 10 BP/PVDF, 20 BP/PVDF, 30 BP/PVDF, 40 BP/PVDF and 50 BP/PVDF, respectively.

To prove the enhanced dielectric properties of IL/polymer composites compared with pure polymers, BB/PVDF and BP/TPU composite films were also prepared and named in the same way as the BP/PVDF system.

2.2. Characterization

The tensile properties of composites were tested through a

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