



Interfacial polarization and dielectric properties of aligned carbon nanotubes/polymer composites: The role of molecular polarity

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ABSTRACT

Dielectric elastomer (DE) composites have attracted much attention owing to their potential applications such as flexible electronics, energy storage, artificial muscles and sensors. The underlying mechanism to improve the electromechanical performance of DE is believed to be the interfacial polarization between fillers and matrix. Thus, understanding the interfacial polarization mechanism is a key to design and produce high performance DE composites. Despite of extensive works on studying the effect of fillers on interfacial polarization, the role of molecular polarity of matrix is still lacking. Thus, in this study, we prepared three kinds of carbon nanotubes (CNTs)/hydrogenated nitrile butadiene rubber (HNBR) DE composites with different molecular polarity of HNBR as the matrices and a special kind of aligned CNTs as the high dielectric constant (ϵ') fillers to study the role of molecular polarity of HNBR on interfacial polarization and dielectric properties of the composites. These composites exhibit the similar dispersion and filler network of CNTs in matrices, which is a precondition of this study. Interestingly, at the same content of CNTs (higher than the percolation threshold), the ϵ' of these composites largely increases with the enhancement of the molecular polarity of HNBR, whereas the ϵ' of these pure HNBR matrices is very close. An equivalent circuit was then established to quantitatively analyze the interfacial polarization mechanism, and it fits well with the experiment data. This study demonstrates for the first time that the higher molecular polarity of matrix leads to the stronger interfacial polarization and thus results in higher ϵ' of the composites.

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1. Introduction

Dielectric elastomer (DE) is able to generate large deformation under external electric field thus converting electrical energy into mechanical energy. By the virtue of high energy density, high electromechanical coupling efficiency, reliability, durability and fast response, DE materials enable the development of light-weighted actuators, micro-robotics, artificial muscles and sensors, and thus

have attracted much attention in recent years [1–4]. The underlying mechanism of the transduction of DE is ascribed to the electrical energy stored in capacitors originating from the materials' capacitance. Despite of these intrinsic advantages, the application potential of DE is still limited by the low dielectric constant (ϵ'). One way to increase the ϵ' is incorporating ceramic fillers [4] such as TiO₂ [5] or BaTiO₃ [6] into the polymer matrix. However, the ϵ' of composites cannot be significantly increased unless the content of the incorporated fillers reaching a high value (e.g. 50 vol%) [7]. In addition, high content of stiff ceramic fillers will result in severe deterioration of mechanical properties and flexibility [8]. Another way to enhance the ϵ' is to incorporate conductive fillers, which is based on percolation theory. The incorporation of conductive fillers can give a drastic increase in ϵ' when the filler content is approaching the percolation threshold [9].

There are mainly two underlying mechanisms on the

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enhancement in ϵ' in conductive fillers/polymer composite [10]. One is the microcapacitor mechanism, according to which two parallel arranged fillers with the insulating polymer matrix in between could form a microcapacitor. Each microcapacitor can make a contribution to the increase in capacitance of the composites. The ϵ' of the composites depends on the network structure of microcapacitors. Within a certain range, increasing the filler content results in an increase in the number of the microcapacitors, which helps to achieve high- ϵ' composites. However, when the filler content reaches a certain value, the conductive fillers interconnect with one another, resulting in the increase in leakage current and thus the drop in ϵ' [10–13]. The other one is the interfacial polarization effect (also known as the Maxwell-Wagner-Sillars effect), according to which the enhancement in ϵ' is owing to the accumulation of polarized charges at the interfaces of heterogeneous systems [4]. The accumulation of polarized charges arises from the difference in conductivities and permittivities of the constituents [14]. Especially in the low-frequency ranges, interfacial polarization effect makes a remarkable contribution to the improvement in the ϵ' . In many cases, the fillers are generally not arranged in parallel, and thus the mechanism in improving the ϵ' is the interfacial polarization mechanism.

Interfacial polarization is influenced by many factors. These factors can be divided into two aspects: intrinsic physical and chemical structure/properties of fillers and intrinsic properties (mainly polarizability) of polymers. Extensive studies have proven that the fillers with strong interfacial interaction with the matrix can effectively enhance the ϵ' of the composites [15,16]. For example, the ϵ' of the composite filled with reduced graphene oxide is much higher than that of composite filled with graphene oxide at the same filler content. This is ascribed that the reduction of graphene oxide restores the graphite structure and thus enhances the interfacial polarization. Thus, to improve the electromechanical properties of these DE composites, the widely used approach is the optimization of filler [17]. However, to the best of our knowledge, hardly any attention was paid on the influence of molecular polarity of polymer matrix on the interfacial polarization effect and the ϵ' of the composites.

Up to now, the interfacial polarization of DE composites is usually studied by analyzing the dielectric properties of the composites. In earlier literatures, the interfacial polarization mechanism was also studied by theoretically calculating through electric modulus formalism [14,18,19], and constantly used as an interpretation of the change in dielectric properties. Dielectric properties are reflections of the impedance of materials under an alternating-current field. The impedance of materials under an alternating-current field can be interpreted by the rational ordered combination of resistance and reactance. Thus, the dielectric properties of the composites can be described by its equivalent circuits constituting elements of resistor (R), capacitor (C), and inductor (L). Therefore, the interfacial polarization mechanism can be quantitatively evaluated by establishing equivalent circuit of the composites and analyzing the composition elements of the composites [20–22].

Thus, in this study, attempts have been made to study the influence of molecular polarity of elastomer matrix on the interfacial polarization and the ϵ' of elastomer composites. Hydrogenated nitrile butadiene rubber (HNBR) with different acrylonitrile content that shows different molecular polarity was used as the elastomer matrix. A special kind of aligned carbon nanotubes (CNTs) prepared by electrostatic self-assembly were used as the conductive fillers. These CNTs can be well dissociated into single CNTs and dispersed uniformly in these HNBR matrices during mechanical shearing and blending [4]. We then studied the electrical properties and dielectric properties of the three kinds of CNTs/HNBR composites.

An equivalent circuit was established to separate the influence of the interfacial polarization from the intrinsic influence of filler and matrix. The interfacial polarization and its influence on dielectric properties of elastomer composites were fully studied through the experimentation and the equivalent circuit model. The relationship among the molecular polarity, interfacial polarization and dielectric properties were revealed.

2. Experimental

2.1. Materials

Hydrogenated nitrile butadiene rubber (HNBR, acrylonitrile (ACN) contents ranging from 24 to 44 wt%) were purchased from Zeon Corporation (Japan). The Mooney viscosity and ACN content (which are provided by the supplier) of different HNBR are shown in Table 1. Carbon nanotube bundle (CNTB, FloTube™ 7000, purity >93%) with a length of 15–25 μm and a width of 2 μm was supplied by Beijing CNano Technology (China) Co., Ltd. Dicumyl peroxide (DCP, 99%) was purchased from Beijing Chemical Reagents (China) Co., Ltd. All chemicals were used as received.

2.2. Preparation of CNTs/HNBR nanocomposites

In this study, the volume fractions of CNTs in the composites were 0.5 vol%, 1 vol%, 1.5 vol%, 2 vol%, and 2.5 vol%. The CNTs/HNBR composites were prepared as follows. A two-roll mill (Zhanjiang Machinery Factory, China) was used to mix HNBR and the CNTB, with the gap between the two rolls about ~ 0.5 mm for high-shear blending. Then DCP (as vulcanizer) was added to the mixture with a mass fraction of 1 wt%. The vulcanization of the composites was then carried out on a lab platen press, with the pressure of 15 MPa, the vulcanization temperature of 170 $^{\circ}\text{C}$ and the optimum curing time that was determined by using an oscillating disc rheometer (P3555B2, Beijing Huanfeng Chemical Machinery Experimental Factory, China). In this study, HNBR with similar Mooney viscosity were used in order to obtain the same dispersion of CNTs in matrix.

2.3. Measurements

Scanning electron microscope (S-4800, Hitachi, Japan) was applied to observe the microstructure and morphology of the CNTB and CNT/HNBR composites under an acceleration voltage of 5 kV. The samples of CNT/HNBR composites were first cryo-fractured and coated with gold.

Transmission electron microscope (H-800-1, Hitachi Co., Japan) was used to observe the dispersion and filler network of CNTs in composites. The TEM specimens were prepared by using a Reichert-Jung Ultra-cut microtome (Leica Camera AG, Germany) at the temperature of 100 $^{\circ}\text{C}$ and then mounted onto 200-mesh copper grids.

CNTs network of CNTs/HNBR composite was investigated by using the rubber process analyzer (RPA 2000, Alpha Technologies, USA) at a fixed frequency of 1 Hz and a temperature of 60 $^{\circ}\text{C}$. The strain was varied from 0.28% to 450%.

Table 1
Mooney viscosity and ACN content of different hydrogenated nitrile butadiene rubber.

HNBR	ACN content /%	Mooney viscosity
Zetpol 3310 (HNBR-24)	24	65.7
Zetpol 2010L (HNBR-36)	36	77.6
Zetpol 1010 (HNBR-44)	44	74.6

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