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Dielectric and mechanical properties of polyimide composite films reinforced with graphene nanoribbon

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ABSTRACT

Graphene nanoribbon (GNR) was introduced into the polyimide (PI) composite films. The effects on dielectric and mechanical properties of PI/GNR composites were investigated. Results show that the dielectric and mechanical properties of the composite films are significantly enhanced compared to pure PI. This can attribute the excellent dispersion of and the strong interfacial interaction between GNR and the PI matrix. Tensile strength of PI/GNR composite films shows a first increasing and then decreasing trend with increased GNR content. With 0.1 wt% loading, the tensile strength is increased from 120.1 MPa to 166.7 MPa, and the dielectric constant of PI/GNR composite film is decreased from 3.6 to 3.1 compared to pure PI, respectively. The success of this preparation is believed to afford new avenue for the development of high strength polyimide based composites.

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1. Introduction

The research on polyimide (PI) based composites, thanks to their remarkable insulation properties, superior mechanical properties, excellent thermal stability, and good resistance to solvents, has been catching great attention [1–6]. With rapid development in some special applications, the properties of pure PI film need be enhanced to meet the extreme requirements. Recently, it was found that the mechanical, dielectric and electrical properties of PI films are significantly improved by the incorporation of some inorganic contents [7–9]. Graphene, an intriguing single-atom thick layered carbon material possessing outstanding properties, becomes very promising new material for PI based composites in various applications [10–16]. In that sense, graphene has been widely used as an effective content mixture to enhance mechanical and dielectric properties of PI [17,18]. At present, two methods, the solution-mixing and the in-situ polymerization, are used to prepare PI/graphene composites by incorporating the functionalized graphene. Liu et al. obtained the maximum mechanical property in PI/graphene oxide composite with 2 wt% graphene oxide, and the tensile strength and the Young's modulus increased 34% and 31%, respectively [19]. However, Tseng and co-workers found the mechanical property of PI/

FG composite with 10 wt% FG enhanced (what perities?) slightly [20]. Shi et al. prepared (3-aminopropyl) trimethoxysilane (APTMS)-functionalized reduced graphene oxide (APTMS-rGO)/polyimide (PI) composite (APTMS-rGO/PI). The results exhibited that the uniform dispersion of APTMS-rGO increases the glass transition temperature and the thermal decomposition temperature and, the tensile strength of the composites with 0.3 wt% APTMS-rGO is 31% higher than that of pure PI, and Young's modulus is 35% higher than that of pure PI [17]. S. Ramakrishnan et al., reported that adding up to 2 wt% Graphene oxide (GO) to PI leads to an improvement in the storage modulus from 1.4×10^8 to 3.8×10^8 Pa and an improvement in the glass transition temperature from 317 to 323 °C over pure PI [21]. T. Huang et al., reported that the PI based composites with 2 wt% modified graphene exhibited a 20-fold increase in wear resistance and a 12% reduction in friction coefficient, constituting a potential breakthrough for future tribological application [22]. These results show that the incorporation of graphene with PI matrix plays a crucial role for improving the mechanical property of resulting composites. Lots of studies have focused on the mechanical behavior of PI composites at a high graphene loading, however, few studies report the interfacial behavior and the mechanical property of PI/graphene composite at ultralow contents. Graphene nanoribbon (GNR), thin elongated strips of graphene, can be fabricated by unzipping carbon nanotubes [23]. The outstanding electronic and spin transport properties of GNR make them attractive materials in a wide range of device applications [24–27]. GNRs have been produced by several techniques including lithographic [28], chemical [29],

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sonochemical [30], and chemical vapor deposition (CVD) [31]. To the best of our knowledge, the preparation of composites with PI and graphene nanoribbon (GNR) has not been reported so far.

In this paper, we evaluated the effects of the incorporation of GNR on mechanical, dielectric and electrical properties of PI composite. The results indicate that the GNR not only can dissolve in *N,N*-dimethylacetamide (DMAC) well, but also provide a starting platform for the in-situ fabrication of composite films. The PI/GNR composites exhibit a dramatic enhancement in mechanical properties at very low GNR doping. We found that the elongation at break and tensile strengths of synthesized PI/GNR composites is greatly enhanced in comparison with pure PI film. For example, with an incorporation of only 0.1 wt% GNR, the elongation at break is increased from 7.2% to 11.7%, which is >50% improvement compared to that of pure PI film. In addition, the volume resistivity and hydrophobic performance are also enhanced. The potential applications of PI/GNR composites with these enhanced properties are greatly increased in future.

2. Experimental setup

2.1. Synthesis of graphene nanoribbon

The Formation of graphene nanoribbon from multiwalled carbon nanotubes (MWCNTs) is synthesized in two steps as previously described [29]. First, MWCNTs (150 mg) were suspended in 30 mL of concentrated H_2SO_4 for a minimum of 1 h and up to 12 h. $KMnO_4$ (750 mg) was then added, and the mixture was constantly stirred for 1 h at room temperature. The reaction mixture was poured onto 100 mL of ice containing H_2O_2 (30%, 5 mL). The resulting light-brown colored precipitate was collected on a 200 nm pore size PTFE membrane, washed several times with HCl (20 vol%) and re-suspended in H_2O by stirring for 2 h. Then HCl (30 vol%, 60 mL) was added to coagulate the product, which was then collected on the same PTFE membrane, washed several times with HCl (20%, 6 mL each), and dispersed in ethanol (40 mL) for 2 h with stirring. Then the product was again coagulated, filtered over the same PTFE membrane, washed several times with ether, and freeze dry to obtain graphene nanoribbon oxide. Second, graphene nanoribbon was prepared by high temperature annealing graphene nanoribbon oxide powder in Ar. 500 mg of graphene nanoribbon oxide powder was put in a tube furnace under a flow of Ar (50 sccm) and annealed for 1 h after reaching the desired annealing temperature (1000 °C).

The resulting sample was obtained after washing repeatedly with DI water and drying at 60 °C for 24 h.

2.2. In-situ synthesis of PI/GNR composite film

The GNR (0.5 g) was dispersed in *N,N*-dimethylacetamide (DMAC) (500 mL) by ultrasonic mixing for 1.5 h, followed by centrifugation at 4000 rpm for 20 min to remove the precipitates. However, nearly no precipitate was found after that, indicating that most is well dispersed in DMAC. Equivalent molar ratios of 2 g 4, 4'-oxy dianiline (ODA) and 2.2 g pyromellitic dianhydride (PMDA) were dissolved in the GNR/DMAC dispersion solution to make solutions containing 0.1, 0.3, 0.5, 1 and 3 wt% of GNR (compared to monomers), respectively. After that, viscous polymer solutions of polyamic acid (PAA)/GNR were obtained. The PAA/GNR solutions were then cast onto glass substrates and dried at room temperature overnight. Next, the PAA composite films were imidized through a sequence of heat treatments at 100, 200, 260, 310, and 350 °C, each for 1 h under nitrogen environment, forming the composite films (PI/GNR) with thicknesses of 30–40 μm (see Fig. 1). All chemicals were bought from the Sinopharm Chemical Reagent Co. Ltd. (China).

2.3. Measurements

Cross section scanning electron microscope (SEM) images were obtained on a JEOL field-emission SEM machine, model JSM-6700F, at operating voltage of 15 kV. TEM was carried out on model JEOL JEM-2010. The small angle x-ray scattering (SAXS) experiments were carried out at beam line 4B9A at Beijing Synchrotron Radiation Facility. The storage ring was operated at 2.2 GeV with current about 80 mA. The incident X-ray wavelength was selected to be 0.154 nm by a double-crystal Si (111) monochromator. The tensile strength and elongation at break were measured on XLD-series liquid screen electronic tensile apparatus with specimens in accordance with GB/T13541-92 at a drawing rate of 50 mm/min. The average of five individual measurements is used with three significant digits, and the unit is MPa. The contact angles of water droplets on the films were measured using a goniometer (JY-82) equipped with a camera to catch images of the water droplets on the surface of the PI composite films. The dielectric constant of hybrid PI films was tested using an impedance analyzer (Agilent 4294A) with 16451B Dielectric Test Fixture in the frequency range of 1–107 Hz. The

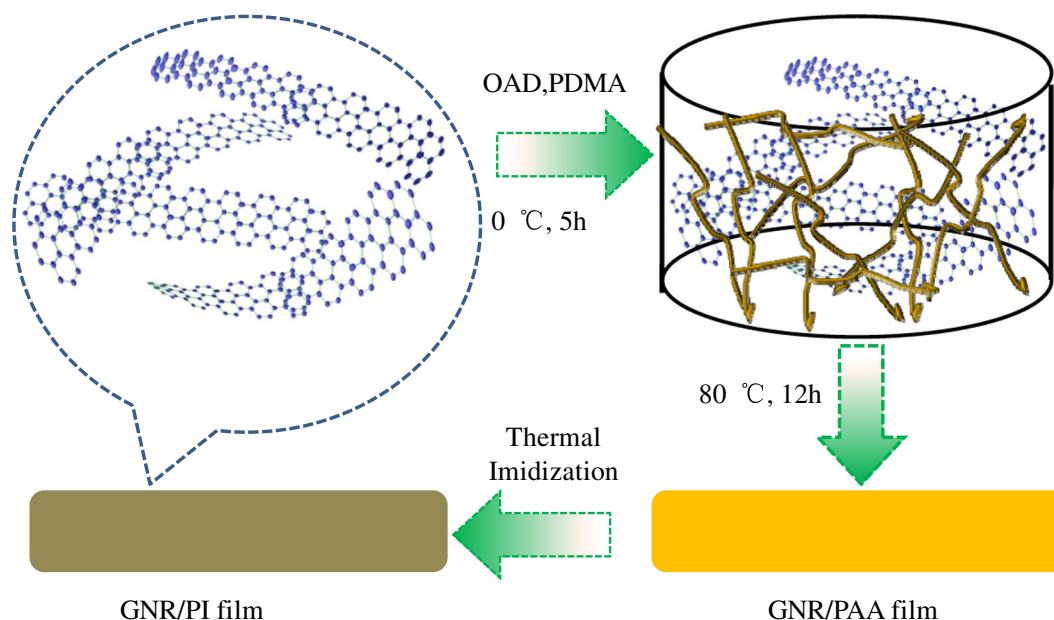


Fig. 1. Schematic flow of in-situ polymerization processes for PI/GNR composite film preparation.

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