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In Situ Preparation of Transparent Polyimide Nanocomposite with a Small Load of Graphene Oxide

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ABSTRACT: Polyimide (PI) nanocomposites with 4,4'-bisphenol A dianhydride, 4,4'-oxydiphthalic anhydride, and diaminodiphenyl methane (MDA) as comonomers and functionalized with graphene oxide (GO), were prepared by in situ polymerization. Only a small amount of GO (0.03-0.12 wt %) is added to improve the mechanical properties of PI and to avoid a substantial decrease of PI transparence. The nanocomposites are characterized by FTIR, X-ray diffraction, thermogravimetric analysis, transmission electron microscope, tensile test, and UV-vis spectroscopy. It is demonstrated that the PI/GO composite films possess transmittance of above 80% at wavelengths of 500-800 nm when the GO content is under 0.12 wt %, while the stress intensity and Young's modulus are improved by 29 and 25%, respectively. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000-000, 2012

KEYWORDS: polyimide; graphene oxide; nanocomposites; transparency

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INTRODUCTION

Polyimide (PI) has been considered as one of the most important materials because of its excellent mechanical properties, striking thermal stability, and resistance to solvents and has been widely used in many applications including optics, aerospace, and microelectronics.¹⁻³ Thus, the development in the past several years of novel PIs with improved properties has been of great interest, especially the high transparency, which has been one of the goals in many studies.^{4–7}

All General PI films present brown or yellow, which will influence their practical applications, such as backplane material of solar battery, glass substrates for liquid crystal displays and plasma discharge panels, transparent flexible plastic substrates as indium tin oxide, wave-guide materials in communication fields, protecting veil for firefighting instrument, and so on,⁸ which all demand for a transparency of at least 80% in the visible light region. The reasons that PI films present heavy color should be attributed to the formation of intermolecular and intramolecular charge transfer complex (CTC) as the interaction of the electron-donating diamine moiety and electron-accepting dianhydride moiety of the PI main molecular chain. And the more the CTC is, the darker the color is. So, the PI transparence can be improved from the viewpoint of molecular design, and many efforts so far have been made to decrease the coloration of PI, such as introducing groups with fluorine,⁹ which has strong electronegativity and can disrupt the conjugation of electron, improving the flexibility of the molecular chain by using monomers-containing flexible groups,¹⁰⁻¹² reducing the structure symmetry by copolymerization, and so on. These factors can efficiently reduce the formation of CTC, sequentially enhancing the transparency.¹³⁻¹⁵ However, all the aforementioned approaches toward improving the transparency of PI films exhibit the tendency to decrease the mechanical and thermal properties of the films due to the increase of the molecular flexibility. So, under the condition that keeping high transparency of PI, enhancing its mechanical properties has important significance. Thereinto, nanocomposite^{7,16-19} with ultrafine inorganic particles (with sizes in the nanometer range) homogeneously dispersed in polymer matrix is a candidate to solve the problem.

Graphene as a kind of two-dimensional (2D) nanofiller has drawn broad attention both in academic and industrial fields for its perfect electronic, thermal, and mechanical properties since its discovery. Many nanocomposites functioned with graphene or its derivatives have been synthesized successfully and presented more absorbing properties.²⁰⁻²⁸ For example, Koo et al.²⁹ have synthesized PI-graphene nanocomposites with amazing thermal properties, and Chen et al.¹ have prepared PI/ GO nanocomposites with better stress intensity and Young's modulus than neat PI films while the color of the films became darker with increasing GO content (0.5-2 wt %). That is to say,

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GO can improve the mechanical and thermal properties but will influence the optical transparency of PI films.

So in this work, first, 4,4'-bisphenol A dianhydride (BPADA), 4,4'-oxydiphthalic anhydride (ODPA), and diaminodiphenyl methane (MDA) were utilized as comonomers to prepare the transparent PI film by *in situ* polymerization.³⁰ Then, we mainly investigated the influence of GO on the mechanical properties, thermal stability, and optical transparency of PI films on the condition that only a small load of GO was added. As a comparison with previous work mainly focused on that 0.1-5 wt% of GO is added, here, we will discuss the properties of the nanocomposite with less than 0.12 wt % of GO. And similar work has not been reported so far.

EXPERIMENTAL

Materials

Graphene oxide (GO) prepared by Hummers' method was obtained from JCNANO (Nanjing, China). BPADA, ODPA, and MDA were purchased from Shanghai Research Institute of Synthetic Resins (Shanghai, China). Acetic anhydride, triethylamine, 1-methyl-2-pyrrolidinone (NMP), *N*,*N*-dimethylformamide (DMF), and absolute ethyl alcohol were from Tianjin Jinhuayongsheng Technology and Trade Limited Company (Tianjin, China).

Synthesis of PI and PI/GO Nanocomposites

About 0.79 g (4 mmol) of MDA and 9 mL of NMP were added to a 100 mL three-necked flask equipped with a mechanical stirrer. About 1.56 g (3 mmol) of BPADA and 0.31 g (1 mmol) of ODPA were mixed in a beaker, and 1/3 of the mixture and 2 mL NMP were added into the reaction flask every 10 min, then flushed with 5 mL NMP. The reaction was carried out in nitrogen atmosphere under room temperature for 24 h to obtain poly(amic acid) (PAA) solution. Acetic anhydride (1.13 mL) and triethylamine (1.67 mL) were added, subsequently the solution was diluted with 16 mL NMP, continued reacting for 24 h. The obtained solution with a great viscosity was diluted with NMP and dropped into 500 mL absolute ethyl alcohol slowly with durative stirring, and offwhite composite was precipitated. The solid product was filtered, washed with alcohol for several times, and then dried in the vacuum drying oven under 30°C for 24 h.

PI/GO nanocomposites were synthesized in the same way, and appropriate amount of prepared GO suspension (10 mg GO was dispersed in 10 mL NMP and treated by ultrasonication for 6 h) was added to the system together with acetic anhydride and triethylamine. The prescription was listed in Table I, and the PI/GO nanocomposites were denoted as PI/GO-1, PI/GO-2, PI/GO-3, PI/GO-4, PI/GO-5 indicating concentrations of 0.03 wt %, 0.05 wt %, 0.07 wt %, 0.10 wt %, 0.12 wt % GO to PI/GO nanocomposites, respectively.

Preparation of PI and PI/GO Nanocomposite Films

About 0.3 g of as-synthesized nanocomposite was dissolved in 5.7 g of NMP to prepare 5 wt % solution (about 6 mL). Approximately 1 mL of the solution was taken accurately with transfer pipette and coated on to four pieces of glass averagely, and the liquid films were dried in vacuum drying oven under

Sample	Monomers (g)	GO suspension of 1 mg/mL in NMP (mL)	Content of GO in composite (wt %)
PI	2.66	0	0
PI/GO-1	2.66	0.80	0.03
PI/GO-2	2.66	1.33	0.05
PI/GO-3	2.66	1.87	0.07
PI/GO-4	2.66	2.66	0.10
PI/GO-5	2.66	3.20	0.12

Table I. The Prescription of PI/GO Nanocomposites

80°C for 3 days. And the transparent PI and PI/GO films with a thickness in the range of 15–20 μm were obtained.

Characterization

The inherent viscosity of PI and PI/GO nanocomposites was determined with an Ubbelohde viscometer in DMF at 25°C. Composite (0.09 g) was dissolved in 12 mL DMF and then diluted with 3, 5, 5, and 5 mL in succession, to measure the delivery time. Then the inherent viscosity was obtained by graphing method. Infrared spectra of the nanocomposites were recorded on a FTS6000 FT-IR spectrometer manufactured by Bio-rad of USA.

An Ultima IV multipurpose X-ray diffraction (XRD) system (Rigaku Co., Sendagaya, Shibuya-Ku, Tokyo, Japan) was used for the X-ray analysis with Cu-Ka radiation ($\lambda = 1.54051$ Å) from $2\theta = 3-80^{\circ}$. Transmission electron microscope (TEM) observations were conducted using a JEM-2100 microscope (JEOL Limited, Tokyo, Japan) with 200 kV.

The thermal stability was characterized by thermogravimetric analysis on TG209 DSC204 DMA242 TMA202 thermal analysis system made by NETZSCH in Germany from 25 to 800°C in nitrogen atmosphere at a heating rate of 10° C/min.

To measure the tensile strength and modulus of the PI and PI/GO films, samples were cut into rectangular sheets with a dimension of $35 \times 6 \text{ mm}^2$ and were tested with a universal material testing machine (Testometric, M500-25KN) at a speed of 20 mm/min. The data were obtained by taking the average values from three groups of tests. The transparency of the films was examined by UV-9100 spectrophotometer (Ruili, Beijing) from 400 to 800 nm.

RESULTS AND DISCUSSION

Preparation of PI and PI/GO Nanocomposites

As is reported, the imidization process of PI is generally carried out via thermal imidization with good effect. While on some occasion, this process is unacceptable for its complexity, and the high temperature during the thermal imidization may also influence their applications.³¹ So, chemical imidization method was chosen in this work. Acetic anhydride (used as dehydrating agent) and triethylamine (used as catalyzer) together with GO are added to the reaction system following the production of PAA by polycondensation with the comonomers.^{32,33} The synthetic route is shown in Figure 1.



Figure 1. Synthetic route of PI.

First, to obtain PI films with high transparency, we selected BPADA, ODPA, and MDA as the comonomers for the reason that the existence of ether bond in BPADA and ODPA can improve the flexibility of PI molecular chain. In addition, allowing for the convenience of film preparation, it is needed that PI has good solubility in some common solvents. Therefore, a series of different prescriptions including copolymerization and homopolymerization were conducted to investigate the transparency and solubility. As shown in Table II, the PI composites synthesized without BPADA have worse solubility in the solvents of THF, NMP, DMF, and acetone. Figure 2 presents the transparency of PI films with different components (MDA, BPADA, and ODPA with a mole ratio of 1:1:0, 2:1:1, 3:2:1, 4:3:1 and 5:4:1). Allowing for these two factors of solubility and transparency, we selected 4:3:1 of MDA, BPADA, and ODPA to synthesize PI and PI/GO composite in this work.

 Table II. Solubility of PI with Different Component in Some Common

 Solvents

	NMP	DMF	Acetone	THF
BPADA+MDA	+	+	_	0
ODPA+MDA	_	_	_	_
BPADA+ODPA+MDA	+	+	_	0

+: Soluble; -: Insoluble; O: Swelling.



The inherent viscosity is one of the important basic properties

of PI, which is related to the molecular weight and film forming

condition. Table III exhibits the inherent viscosity values of PI and its composites. The results do not present special orderli-

ness as other literatures,³⁴⁻³⁶ probably attributed to the disperse

state and the very small load of GO in PI matrix. In general,

Figure 2. Transparency of PI films with different component. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



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Samples	Inherent viscosity (mL/g)	Temperature at the Maximal rate of weight loss (°C)
PI	49.7	541.2
PI/GO-1	51.6	537.7
PI/GO-2	50.7	544.5
PI/GO-3	51.3	543.4
PI/GO-4	45.6	542.9
PI/GO-5	46.9	542.2

 Table III. Inherent Viscosity and Temperature at the Maximal Rate of

 Weight Loss of PI and its Nanocomposites

the inherent viscosity of PI is not influenced dramatically as the addition of a small amount of GO. Thus, it is also not necessary to change the film forming condition with the addition of a small amount of GO.

Figure 3 shows the FTIR spectra of PI and PI/GO (0.1 wt %) nanocomposites. The characteristic imide groups are observed as below: 1778 cm⁻¹ (C=O asymmetry, stretching), 1724 cm⁻¹ (C=O symmetry, str.), and 1369 cm⁻¹ (C–N, str.), 744 cm⁻¹ (imide ring, deformation). The peak at 3035 cm⁻¹ is the stretching vibration of =CH on benzene ring. Synchronously, the characteristic absorption peaks, at 1660 cm⁻¹ (C–O of amid), 1550 cm⁻¹ (C–NH), and 1710 cm⁻¹ (C=O of – COOH) do not occur, indicating the typical completion of the imidization reaction with the presence of the four characteristic absorption bands from the imide rings.^{10,37} Comparing the two curves, there is no obviously different absorption peak, probably because the amount of GO is too small.

Morphology

To evaluate the dispersion state of GO in PI substrate, which determines the performance of the nanocomposites, TEM was used to investigate the morphology of the samples. Figure 4 shows the TEM images of (a) GO (b) PI/GO composites (0.1 wt %). From Figure 4(a), we can see the GO sheets have a size of about several micrometers with some corrugations clearly,



Figure 3. FT-IR spectra of PI and PI/GO nanocomposite (0.1 wt %). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



Figure 4. TEM images of (a) GO, (b) PI/GO nanocomposites.

and Figure 4(b) shows that the few-layers GO looks like little black dot is dispersed in PI matrix with the form of stacks, and the exhibition of diffraction fringes suggests the existence of single layer. The scale of GO stack is less than 100 nm, indicating the good compatibility between GO and PI matrix. It is expected that the functional groups on the surfaces of GO probably formed hydrogen bonds with the carbonyl groups on the polymer chains of PAA or PI.^{1,38}

XRD Patterns

As is shown in Figure 5, the crystallinity of GO, PI, and PI/GO nanocomposites were characterized by XRD. GO has a sharp peak at $2\theta = 9.4^{\circ}$, corresponding to interlayer spacing of about 0.92 nm, indicating high oxidation degree of GO. And it should be noted that the sharp peak of GO disappeared in the composites, probably because that the amount of GO was not enough to get up to the sensitivity.^{18,39–41} Moreover, the expanded XRD peaks of PI and PI/GO nanocomposites denote their low crystallinity. The neat PI film has a broad peak at $2\theta = 18.1^{\circ}$ which is corresponding to the literature data.^{42–45} In addition, no peak of GO was detected, thus suggesting the GO sheets were homogeneously dispersed in PI nanocomposite.



Figure 5. XRD patterns of PI and PI/GO nanocomposites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Thermal Stability

Thermal stability is one important property of PI-based composites. As shown in Figure 6(a), TGA curves and (b) DTG curves, at about 200°C, there is a weight loss of about 10%, which may be due to two reasons: (1) the evaporation of residual solvent NMP, suggesting that there exist still a little of residual solvent under this film forming condition; (2) the removal of water during the total imidization process for PI, as known that the chemical imidization usually proceeds not totally. And at about 500°C, PI and its composites begin to decompose, substantiating the excellent thermal stability of the films, which may rest with the existence of rigid benzene rings in the monomer. The temperature at the maximal rate of weight loss is obtained from the DTG curves, as listed in Table III, we can see that there is no obvious improvement, demonstrating that very small amount of GO will not influence the thermal stability of PI film remarkably.

Mechanical Properties

Typical stress-strain curves of neat PI and PI/GO nanocomposite films are shown in Figure 7, and the data of mechanical properties are presented in Figure 8(a,b). As expected, the tensile stress and Young's modulus of the films increase with the addition of GO. When only 0.03 wt % of GO is added, the tensile stress and Young's modulus are improved by 9.7 and 4.1%, respectively. When the GO content is further increased to 0.07 wt %, the modulus decreases slightly but still much higher than neat PI. And when the amount of GO is lifted to 0.12 wt %, the tensile stress of the PI/GO nanocomposite was increased from 58 to 75 MPa (about 29%) and the Young's modulus was increased from 1.4 to 1.8 GPa (about 25%) compared with those of neat PI films. Although the elongation at break was not decreased markedly, which indicating that only a small amount of GO did not lead to brittleness of PI films. The enhancement in both tensile strength and Young's modulus of PI/GO nanocomposites could be attributed to the good dispersion of GO in PI matrix and effective load transfer from the PI matrix to graphene due to the



Figure 6. (a) TGA and (b) DTG curves of PI and PI/GO nanocomposites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

strong interaction between the OH groups in GO and the main backbone of PI or the strong π - π interaction between the functional group of GO and PI.^{46,47}



Figure 7. Stress-strain curves of neat PI and PI/GO composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



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Figure 8. (a) Tensile strength and (b) Young's modulus of PI and PI/GO composites.

Transparency

Figure 9 displays the color of PI and PI/GO nanocomposite films, demonstrating that the color of the films becomes deeper slightly with the increasing of GO content. The transparency can be characterized by UV-vis absorption spectroscopy,³⁰ as shown in Figure 10, the neat PI film with a thickness in the range of 15-20 µm has a high transparency of nearly 95% at long wavelength region, which indicates that this method of copolymerization with BPADA, ODPA, and MDA to enhance the transparency of PI is effective comparing with other literatures.⁵⁻⁸ The high transparency should mainly rest with the flexibility of molecular chain due to the existence of ether bond and methylene and that two different dianhydrides (BPADA and ODPA) are used in the system to reduce the structure symmetry of PI, thus contributing to preventing the formation of CTC. When only 0.03 wt % GO is added, the transparency decreases a little, but still keeps above 90%. Whereas, the transparency drops below 90% when increasing GO content because of its stacks in PI substrate. And when the content of GO is less than 0.12 wt %, the transparence maintains above 80% at 500-800



Figure 9. Optical images of PI and PI/GO composite films (from left to right: neat PI, PI/GO-1, PI/GO-2, PI/GO-3, PI/GO-4, PI/GO-5). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

nm, suggesting that small quantities of GO will not influence the transparence remarkably.

CONCLUSIONS

Transparent PI nanocomposites functionalized with GO were prepared with BPADA, ODPA, and MDA as comonomers and GO as nanoadditives. And the addition of small amount of GO did not influence the transparence significantly with the fact that PI/ GO films with 0.12 wt % GO content possess a transparency of above 80% at 500–800 nm. Tensile test revealed the considerable mechanical properties of the PI/GO nanocomposite films with the stress intensity and Young's modulus enhanced by 29 and 25%, respectively as the addition of only 0.12 wt % GO.



Figure 10. UV-vis spectra of PI and PI/GO composite films (thickness of 15–20 μ m). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

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