Polymer 54 (2013) 2335-2340

Contents lists available at SciVerse ScienceDirect

Polymer



Intermolecular interactions of polyimides containing benzimidazole and benzoxazole moieties

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ARTICLE INFO

Article history: Received 29 November 2012 Received in revised form 28 February 2013 Accepted 28 February 2013 Available online 6 March 2013

Keywords: Intermolecular interaction Hydrogen bonding Polyimides

ABSTRACT

Thermal analysis and infrared spectroscopic studies were performed to investigate the strength of intermolecular interactions of the polyimides derived from 5,4'-diamino-2-phenyl benzimidazole (DAPBI) and 5,4'-diamino-2-phenyl benzoxazole (DAPBO). Polyimide films were prepared based on biphenyltetracarboxylic dianhydride (BPDA) isomers, difference of glass transition temperatures (ΔT_g) between DAPBI and DAPBO type polyimides decreased consistently as the steric impedance increased, implying that polyimides containing benzimidazole group had higher degree of intermolecular interactions. Infrared spectra of polyimide films displayed that the imide carbonyl (C=O) stretching band for DAPBI derived polyimides shifted to lower frequency compared to the DAPBO counterpart. In the IR spectra of the copolyimides based on DAPBI and DAPBO, significant red shift was observed for C=O stretching band and T_g increased as a function of DAPBI content. This suggests the presence of strong intermolecular interactions due to the hydrogen bonding for the DAPBI based polyimides.

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1. Introduction

Polyimide films having a favorable combination of properties such as high glass transition temperatures, excellent mechanical properties, and good dimensional stabilities have been extensively investigated both in academic and application studies. By modifying their chemical structures and manufacturing conditions, polyimides could be tailored to meet specific design criteria [1]. Particularly, polyimides having rigid-rod backbones have attracted much attention because PI systems possessing linear and stiff backbone structures tend to show high tensile modulus and low coefficient of thermal expansion (CTE), which are desirable since polyimides are frequently used as coatings and films adhered to metal or silicon substrates [2].

Incorporating heterocyclic units into polyimide main chains is an effective way to improve thermal and mechanical properties of polyimides since this class of groups tend to increase the rigidity of the polymer backbones and provide strong intermolecular associations [3–5], as evidenced from the extraordinarily high performance of some aromatic heterocyclic polymers [6,7]. Improved properties were obtained after quinazolinone [8] and benzothiazole [9] moieties were introduced into the polyimide backbones. In our previous work, benzimidazole and benzoxazole mojeties were incorporated into the polyimide main chains, using 5.4'-diamino-2-phenyl benzimidazole (DAPBI) [10] and 5.4'-diamino-2-phenyl benzoxazole (DAPBO). Thermal and mechanical properties of the resultant polyimide films were remarkably enhanced compared to those of the common PI films. Another characteristic we observed was an obvious T_g difference between DAPBI and DAPBO derived polyimdes: Tg of DAPBI derived polyimides was higher than that of DAPBO type polyimides for at least 40 °C when the dianhydride component was the same. It was presumed that stronger intermolecular interactions between DAPBI derived polyimide chains contributed to the higher T_{g} , more specifically, -NH- group of DAPBI acted as the proton donor and offered favorable condition for the formation of hydrogen bonding with electron-rich group, like C=O (shown in Fig. 1). In this study, homopolyimides were separately prepared from BPDA isomers and the two diamines, ΔT_{g} between DAPBI and DAPBO based polyimides were explained from the viewpoint of the different steric structures contributed by the BPDA isomers, and it was used to discuss the strength of intermolecular interactions. From the IR spectra of polyimides based on DAPBI and DAPBO, the presence of hydrogen bonding was investigated in term of the frequency shift of C=O and -NH-, and T_g dependence of polyimides on hydrogen bonding was also displayed.





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Fig. 1. Schematic diagram of intermolecular hydrogen bonding for polyimides containing benzimidazole unit.

2. Experimental

2.1. Reagents and characterizations

The chemical structures and melting points of the monomers used are listed in Table 1. DAPBI and DAPBO were supplied by Changzhou Sunlight Fine Chemicals Co. Ltd and used as received. s-BPDA, BTDA and BPADA were supplied by Sinopharm Chemical Reagent Beijing Co. Ltd. a-BPDA was a gift from UBE Industries. Inc. i-BPDA was synthesized in our lab [11]. The dianhydrides were dried at 150 °C under vacuum for 10 h prior to use. N,N'-

Table 1

Chemical structures and melting points of the monomers.

dimethylacetamide (DMAc) and xylene, products of Tianjin Chemical Reagent Co. Ltd., were of analytical grade and used without further purification. Phthalic anhydride (PA) was a product of Shanghai Shanpu Chemical Engineering Co. Ltd. and used as received.

FT-IR spectra were obtained on a Bruker Vector 22 spectrometer at a resolution of 4 cm⁻¹ in the range of 400 cm⁻¹–4000 cm⁻¹, with the sample form of thin films and powders. All spectra were measured at room temperature and represented the average of 128 scans. Dynamic mechanical analysis (DMA) was carried out with a TA instrument DMA Q800 at a heating rate of 5 °C/min and a load frequency of 1 Hz in film tension geometry, T_g was regarded as the peak temperature of the tan δ curve.

2.2. Preparation of polyimide thin films for FT-IR test

DAPBI and DAPBO were reacted with equimolar amount of s-BPDA, BTDA and BPADA, respectively, in DMAc as the polymerization solvent at room temperature, to obtain homogenous polyamic acid (PAA) solutions. Films were cast from PAA-DMAc solution onto a clean, flat glass plate followed by a preheating program ($80 \degree C/1$ h, 100 $\degree C/1$ h, 120 $\degree C/1$ h, 150 $\degree C/1$ h, 180 $\degree C/1$ h) and an imidization procedure under vacuum ($250 \degree C/1$ h, $320 \degree C/1$ h, $400 \degree C/1$ h, except the BPADA derived PI was annealed at $350 \degree C$). The thickness of the film was controlled to be $3-5 \ \mu$ m. The reaction procedure is shown in Scheme 1.

Random copolyimides based on s-BPDA, DAPBI and DAPBO were prepared in the same fashion, the molar ratios of DAPBI/DAPBO were 20/80, 40/60, 60/40 and 80/20, as shown in Scheme 2.

Monomer	Monomer	Chemical structure	Melting point/°C
DAPBI	5,4′-diamino-2-phenyl benzimidazole		236
DAPBO	5,4'-diamino-2-phenyl benzoxazole		233
s-BPDA	3,3',4,4'-biphenyltetracarboxylic dianhydride		298
a-BPDA	2,3,3',4'-biphenyltetracarboxylic dianhydride		196
i-BPDA	2,2',3,3'-biphenyltetracarboxylic dianhydride		275
BTDA	3,3',4,4'-benzophonetetracarboxylic dianhydride		227
BPADA	2,2-bis[4-(3,4-dicarboxypheoxy)phenyl] propane dianhydride		190

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