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Soluble and Degradable Polyimides with Phenyl-2-pyridyl Ether Structure: Synthesis and Characterization*

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Abstract A diamine monomer *o*-phenylenedioxybis(5-amino-2-pyridine) was synthesized *via* reduction of a dinitro compound *o*-phenylenedioxybis(5-nitro-2-pyridine), producing a series of new polyimides from this diamine and various commercially available aromatic dianhydrides *via* conventional two-stage processes. The resulting polyimides are able to form tough and transparent films, with decomposition temperatures in the range of 529–551 °C, and can be dissolved in organic polar solvents. Meanwhile, these polyimides can be degraded in a hydrazine hydrate medium, a degradation mechanism proposed by analyzing the degradation products suggests that the degradable properties could be attributed to the phenyl-2-pyridyl ether structure in the polymer. In addition, the transformation of the compound structure from dinitro compound to damine monomer in the synthetic process is discussed in respect to X-ray structure.

Keywords: Monomer structure; Polymer solubility; Degradable polyimide.

INTRODUCTION

Aromatic polyimides, because of their excellent stabilities, chemical resistance and electric properties, are widely used in the aerospace and microelectronic industry in the forms of films and moldings. Other uses for these polymers including adhesives, gas separation membranes, composite matrices, coating and foams are also rapidly increasing[1−6]. However, most polyimides have high melting or softening temperatures and limited solubility in most organic solvents because of rigid backbones and strong interaction between chains, which may restrict application in some fields. Nonetheless, significant efforts and progress have been made in the development of methodologies for increasing the solubility of polyimides. Incorporation of pendent groups[7−9], molecular asymmetry[10−12], or flexible linkages[13−15] into the backbone are typical approaches to improve the solubilities.

Of these methods, introduction of aromatic ether linkages is one of the most effective in increasing solubility of polyimides, due to reduced electron-conjugation in imide rings resulting from aromatic ether

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linkages inserted in polymer main chains[16−20]. The coordinated application of inserted aromatic ether linkages and introducing pendent substituents provides significantly lower energy of internal interactions[20−22].

The *ortho*-position structure diphenol could be incorporated as flexible linkages into polymer main chains. However, reports on *o*-diphenol based monomer are much less. It is generally recognized that *o*-diphenol based structures are too twisted to make the polymer chain frizzy, thus, some active groups will be embedded into the polymer chain, which is not conducive to produce high-molecular-weight polymers. In this work, we synthesized an *o*-diphenol based diamine monomer *o*-phenylenedioxybis(5-amino-2-pyridine) (*o*-PAP). This *o*-diphenol based diamne monomer shows a balance-like structure, as shown in Fig 1. The two amine groups on *o*-PAP are almost in the same line, it was opposite to the general reorganization. To our knowledge, this is the first report of *o*-diphenol based diamine monomers with an X-ray structure. The resulting polyimides exhibited good solubility in organic solvents, possibly due to the central benzene ring that acts as a pendent substituent in the polymer chain, improving solubility by helping keep distance between chains. Additionally, this molecule was found to decompose in hydrazine hydrate medium, compared with the traditional degradation method, such as thermal oxidative decomposition[23−26], hydrolysis[27, 28] and radiation decomposition[29, 30], this method provided a green method to degrade polyimides, and the degradation of the resulted polymers was discussed in this paper.

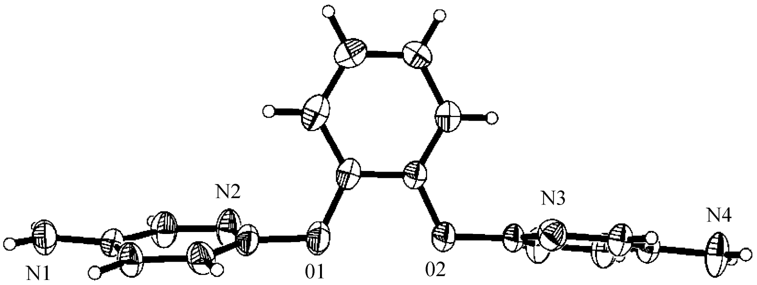


Fig. 1 X-ray structure of diamine monomer *o*-PAP

EXPERIMENTAL

Materials

Commercially available 2-chloro-5-nitropyridine (Shanghai Chemical Reagents Corp., China), *o*-dihydroxybenzene (Fuchen Chemical Reagents Corp., Tianjin, China) and $SnCl_2$ (Fuchen Chemical reagents Corp., Tianjin, China) were used without further purification. 4,4′-Oxydiphthalic anhydride (ODPA, Aldrich), 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (BTDA, Aldrich) and 2,2′-bis(3,4-dicarboxyphenyl) hexafluoropropane dianhydride (6FDA, Aldrich) were recrystallized from acetic anhydride before use. Pyromellitic dianhydride (PMDA, Beijing Chemical Reagents Corp., China) was purified by sublimation under vacuum at 200–220 °C. *N*,*N*-Dimethylacetamide (DMAc) and *N*-methyl-2-pyrrolidone (NMP) and *N*,*N*-dimethylformamide (DMF) were purified by distillation under reduced pressure over calcium hydride and stored over 4 Å molecular sieves. All the other solvents were obtained from various commercial sources and used without further purification.

Measurements

The inherent viscosities of the resulting polyimides were measured with an Ubbelohde viscometer at 30 °C. FTIR spectra (KBr) were recorded on a Nicolet Nexus 670 FTIR spectrometer. ^{1}H-NMR and ^{13}C-NMR spectra were measured on a JEOLEX-400 spectrometer with tetramethylsilane as the internal reference. Elemental analyses were determined with a Perkin Elmer model 2400 CHN analyzer. Differential scanning calorimetry (DSC) testing was performed on a Perkin Elmer DSC 7, differential scanning calorimeter at a scanning rate of 10 K/min in the flowing nitrogen (30 cm^3/min), and the glass transition temperature (T_g) values were read at the DSC curves at the same time. Thermogravimetric analysis (TGA) was conducted with a TA Instruments TGA 2050, and the experiments were carried out with approximately 10 mg samples in the flowing nitrogen (flow rate

= 100 cm^3/min) at a heating rate of 20 K/min. The mechanical properties were measured on an Instron 1122 tensile apparatus with 100 mm × 5 mm specimens in accordance with GB 1040-79 at a drawing rate of 100 mm/min.

Monomer Synthesis

o-Phenylenedioxybis(5-nitro-2-pyridine) (o-PNP)

2-Chloro-5-nitropyridine (7.9 g, 0.05 mol) and *o*-dihydroxybenzene (2.75 g, 0.025 mol) were first dissolved in 40 mL of DMF in a 100 mL flask with stirring. After the mixture was completely dissolved, potassium carbonate (8.00 g, 0.06 mol) was added into it in one portion. After 30 min of stirring at room temperature, the mixture was heated at 80 °C for 6 h. The obtained mixture was poured into 300 mL of distilled water. The precipitated solid was collected by filtration and dried in vacuo at 60 °C for 12 h. The crude product was purified by column chromatography over silica gel (7:1 petroleum ether/ethyl acetate), 9.17 g was obtained (83%). *o*-PNP: ^{1}H-NMR ($CDCl_3$, δ): 8.98 (d,2H), 8.43 (dd, 2H), 7.4 (m, 2H), 7.34 (m, 2H), 6.9 (d, 2H). ^{13}C-NMR: 165.97, 144.81, 144.23, 140.56, 134.86, 127.21, 123.69, 110.82; MS (*m*/*e*) 354.06, Calcd 354 for $C_{16}H_{10}N_4O_6$.

o-Phenylenedioxybis(5-amino-2-pyridine) (o-PAP)

A mixture consisting of 0.02 mol (7.08 g) of *o*-PNP, 27.08 g (0.12mol) of anhydrous $SnCl_2$ and 500 mL of 95% C_2H_5OH was put into a reaction flask, while 20 mL of concentrated HCl was added slowly under stirring. After addition of hydrochloric acid was finished, the mixture was refluxed for 12 h. Excess ethanol was evaporated, and the remaining solution was poured into 400 mL of distiller water, the mixing solution was basified with 15% K_2CO_3 solution to form a precipitate, and the precipitate was filtrated off, washed with water and methanol, recrystallized from toluene to get a yellow product. The product was dried in a vacuum at 40 °C for 2 days, giving rise to 4.74 g of PAP, yield 76%. *o*-PAP: ^{1}H-NMR ($CDCl_3$, δ): 7.61 (s, 2H), 7.16 (s, 2H), 6.98 (d, 2H), 6.6 (d, 2H), 4.9 (s, NH_2, 4H); ^{13}C-NMR (DMSO, δ): 153.34, 146.10, 140.91, 131.28, 124.82, 123.76, 121.56, 110.79; MS (*m*/*e*) 294.11, Calcd 294 for $C_{16}H_{14}N_4O_2$.

Polymer Synthesis

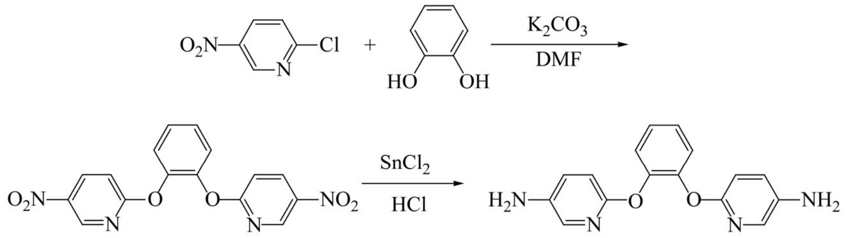
The polyimides were synthesized from various dianhydrides and diamine PAP *via* a two-step method. The synthesis of polyimide PI-1 (ODPA-*o*-PAP) is used as an example to illustrate the general synthetic route that used to produce the polyimides. To a solution of 0.7356 g (2.50 mmol) of diamine *o*-PAP in 11.6 mL of CaH_2-dried NMP, 0.7756 g (2.50 mmol) of ODPA was added in one portion. The solution was stirred at room temperature under N_2 for 24 h to yield a viscous poly(amic acid) (PAA) solution. PAA was converted into a polyimide with thermal or chemical imidization methods. For the thermal imidization method, the PAA solution was cast onto a clean glass plate and heated (80 °C/3 h, 120 °C/30 min, 150 °C/30 min, 180 °C/30 min, 210 °C/30 min, 250 °C/30 min, 300 °C/1 h) to produce a fully imidized polyimide film. Chemical imidization was carried out by the addition of an equimolar mixture of acetic anhydride and pyridine to the aforementioned PAA solution *via* heating at 80 °C for 4 h. The polyimide solution was poured into methanol solution (methanol: water = 1:1). The precipitate was collected by filtration, washed thoroughly with methanol, and dried at 80 °C in vacuo to give the followings.

PI-2 (BTDA-*o*-PAP), PI-3 (6FDA-*o*-PAP), and PI-4 (PMDA-*o*-PAP) were synthesized by the similar method.

RESULTS AND DISCUSSION

Monomer Synthesis

The diamine monomer, *o*-PAP, was synthesized *via* a two-step procedure, as shown in Scheme 1. In the first step, *o*-PNP was synthesized by a nucleophilic aromatic substitution reaction in which 2-chloro-5-nitropyridine is reacted with *o*-dihydroxybenzene by using potassium carbonate as a base in DMF. The resulting 2-chloro-5-nitropyridine has two electron withdrawing groups (nitro-group and N atom), facilitating the separation of the chlorine atom. Thus, in this step, the reaction could finish at low temperature with high yield. In the second step, dinitro compound *o*-PNP was reduced to *o*-PAP by $SnCl_2$ and hydrochloric acid in ethanol. In this step, the key point is how to neutralize hydrochloric acid. We found strong-bases such as NaOH and KOH to be unsuitable for this reaction, while weak-bases such as K_2CO_3, and ammonia could give higher yields and easily controlled reactions.



Scheme 1 Synthesis of *o*-PAP

The structure of both *o*-PNP and *o*-PAP were confirmed by NMR spectroscopy. As shown in Fig. 2, the absorption signals of aromatic protons for *o*-PNP and *o*-PAP appeared in the range of δ = 6.5–9.1. In the ^{13}C-NMR spectrum (Fig. 3), the carbon 13 atoms in *o*-PNP and *o*-PAP showed 8 signals which resonated in the regions of δ = 110–170. All the spectroscopic data obtained agreed with the expected structures. Meanwhile, the protons of the amine group of *o*-PAP appeared at δ = 4.9, which indicated that this diamine monomer has higher activity.

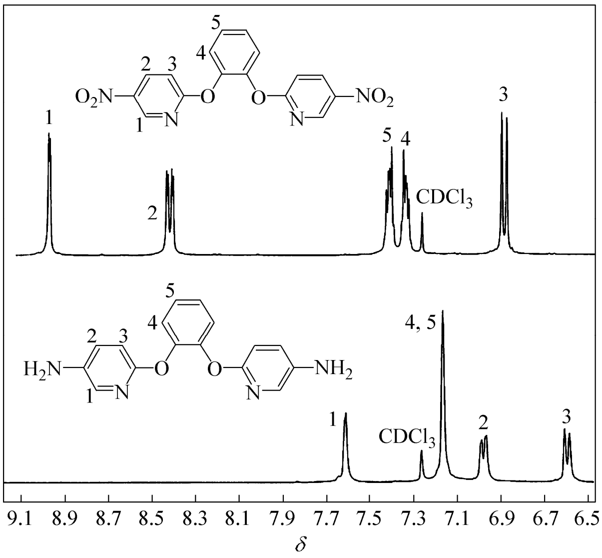


Fig. 2 ^{1}H-NMR spectra of *o*-PNP (top) and *o*-PAP (bottom)

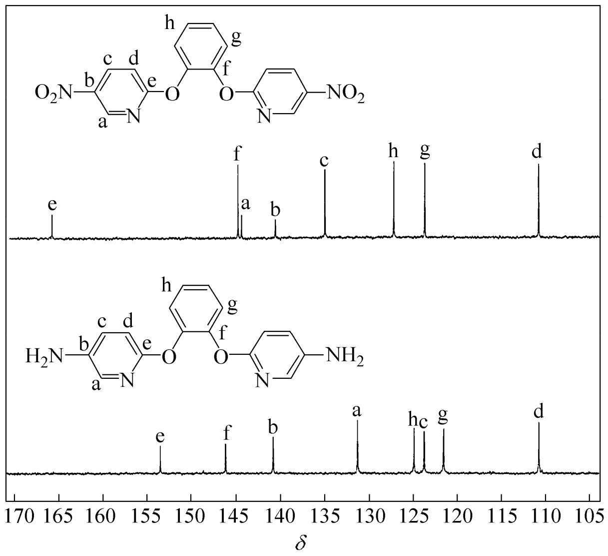
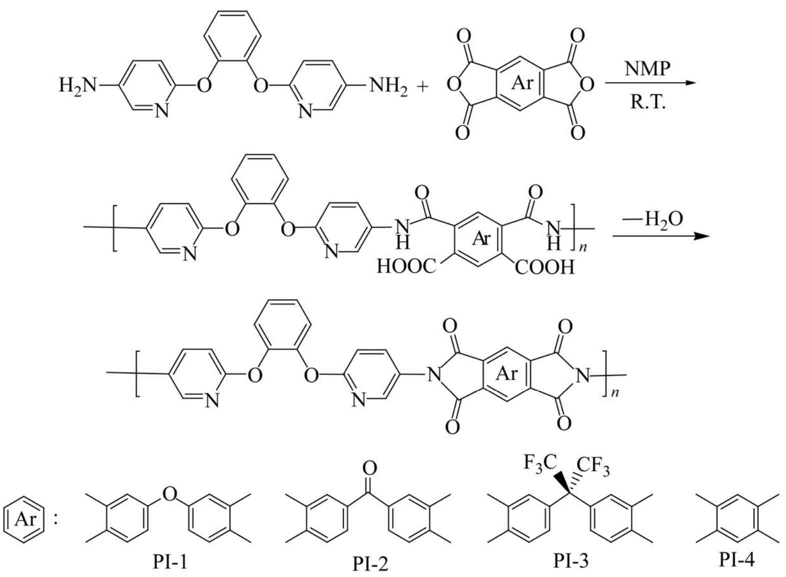


Fig. 3 ^{13}C-NMR spectra of *o*-PNP (top) and *o*-PAP (bottom)

Preparation of Polyimides

The related polyimides were prepared from *o*-PAP and commercially available aromatic dianhydrides, such as ODPA, BTDA, 6FDA, and PMDA, *via* a conventional two-step procedure as shown in Scheme 2. The polymerization was carried out by reacting stoichiometric amounts of diamine monomer *o*-PAP with aromatic dianhydrides at a concentration of 15% solids in NMP. First, the polymerization was conducted at room temperature for 24 h to form poly(amic acid)s (PAA), followed by sequential heating to 300 °C or mixing with Ac_2O/Py to obtain the corresponding polymers. Both thermal and chemical methods could transform PAA to polyimide, but the former method is easier to handle and used to make polyimide thin films, while the latter is better suited to prepare soluble polyimides. The chemical structures of the polyimides were characterized by FTIR and element analysis.



Scheme 2 Synthesis of the polyimides

All of the polyimides were able to form tough and transparent films, and showed characteristic imide absorption bands around 1780 cm^{-1} attributed to the asymmetrical carbonyl stretching vibrations, and at 1720 cm^{-1} attributed to the symmetrical carbonyl stretching vibrations. The absorption around 1380 cm^{-1} was assigned to C—N stretching, and C—O multiple stretching absorptions were also detected in the range of 1300–1100 cm^{-1}. There was no existence of the characteristic absorption bands of the amide group near 3200–3360 (N—H stretching) and 1650–1670 (C=O stretching) cm^{-1}. Table 1 shows the element analysis data for the novel polyimides, which were in agreement with the calculated values for the proposed chemical structures.

Table 1. Physical properties and elemental analysis of the polyimides

Polyimide	Inherent viscosity of PAA(dL/g)	Composition of repeating unit	Elemental analysis (%)			
				C	H	N
PI-1	0.74	$C_{32}H_{17}N_4O_7$	Calcd	68.22	3.70	9.36
			Found	68.49	3.83	9.16
PI-2	0.67	$C_{33}H_{17}N_4O_7$	Calcd	68.85	3.63	9.18
			Found	68.66	3.89	9.35
PI-3	0.55	$C_{35}H_{17}F_6N_4O_6$	Calcd	60.66	3.03	7.65
			Found	60.97	3.37	7.82
PI-4	0.83	$C_{26}H_{13}N_4O_6$	Calcd	66.40	3.58	11.06
			Found	66.54	3.47	11.22

Monomer Structure and Polymer Solubility

Fortunately, the monocrystalline structures of both *o*-PNP and *o*-PAP[†], as shown in Fig 4, were obtained. Unexpectedly, *o*-PNP showed a very different structure compared with *o*-PAP, with the structure of *o*-PNP similar to a running person, while the *o*-PAP resembles a balance. In the reductive process, through the rotation of the C—O bonds, the structure of "running person" could transfer to that of the "balance". This considerable

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[†] The crystal structure of *o*-PNP and o-PAP (ethyl acetate) is a colorless block form. Crystallographic data for the structure reported in this paper have been deposited with the Cambridge Crystallographic Data Centre (CCDC) as supplementary publication no. CCDC: 1012824 (*o*-PNP) and 1012825(*o*-PAP). Copies of the data can be obtained free of charge from the CCDC (12 Union Road, Cambridge CB2 1EZ, UK; Tel.: +44 1223 336408; Fax: +44 1223 336003
E-mail: deposit@ccdc.cam.ac.uk; Web site: http://www.ccdc.cam.ac.uk)

change in structure could be helpful to polymerization, because the two amine groups were almost in the same line. In this case, the polymer chain should grow in this direction, and the active groups are not embedded easily into the polymer chain. Meanwhile, from the monocrystalline structure of *o*-PAP it was found that the middle benzene ring could act as a pendent substitute in the polymer chain. The pendent substituent would keep distance between the chains, which is helpful for the solubility of polymers. As expected, the resulting polyimides show good solubility in organic solvents, as shown in Table 2. It can be seen that almost all the polyimides, except PMDA-*o*-PAP, could be dissolved in organic polar solvents, such as *m*-cresol, NMP, or DMAc, even at room temperature in most cases.

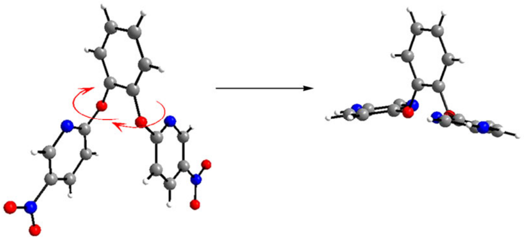


Fig. 4 Structure transformation from *o*-PNP (left) to *o*-PAP (right)

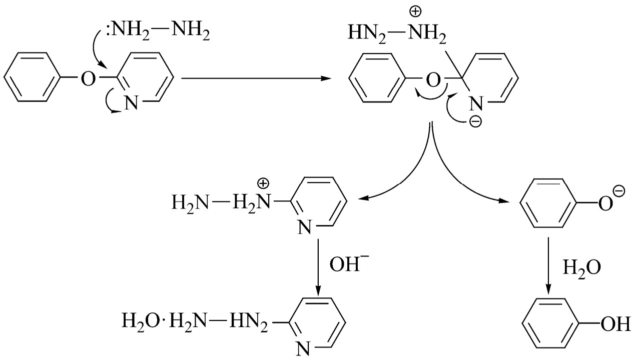
Table 2. Solubility of the polyimides

Solvents	Polyimides [a]			
	PI-1	PI-2	PI-3	PI-4
NMP	+ +	+	+ +	–
DMAc	+ +	+ –	+ +	–
DMF	+ –	–	+	–
DMSO	+ –	–	+	–
m-Cresol	+ +	+ +	+ +	–

[a] Measured by chemical cyclization polyimide derived from the corresponding poly(amic acid)s; Qualitative solubility was determined with as 3 mg of polymer in 1 mL of solvent. + +: soluble at room temperature; +: soluble on heating; + –: partial soluble on heating; –: insoluble on heating.

Polymer Degradation

Interestingly, *o*-PNP was found to decompose in a hydrazine hydrate medium at room temperature as we reported previously[31]. This could be attributed to the phenyl-2-pyridyl ether structure present in *o*-PNP, as shown in Scheme 3. The α-carbon of nitrogen on 2-hydroxypyridine has similar electrophilic reactivity with carbonyl, and this carbon can be attacked by lone pair of hydrazine hydrate, breaking the π-bond and creating a tetrahedral intermediate. Pushing back of the electrons of pyridine nitrogen results in leaving of the aminophenol to afford the protonated amino pyridine and the deprotonated aminophenol. Then, proton transfer from water and hydroxide can neutralize the protonated amino pyridine and the deprotonated aminophenol to generate the final products.



Scheme 3 Decomposation mechanism of phenyl-2-pyridyl ether structure

PI-3, which shows good solubility, was used to carry out the degradation experiments in 3-methylphenol. Hydrazine hydrate was added into the above solution and stayed at 50 °C for one week. Gel permeation chromatography (GPC) was used to test the change of polyimide before and after degradation, as shown in Fig. 5. Molecular weight of PAA-3 is about 22000 (M_n). However, the same sample treated by ammonia for one week shows the molecular weight of only about ~700. The GPC results clearly suggest that such a polyimide with phenyl-2-pyridyl ether structure could degrade fast and greenly.

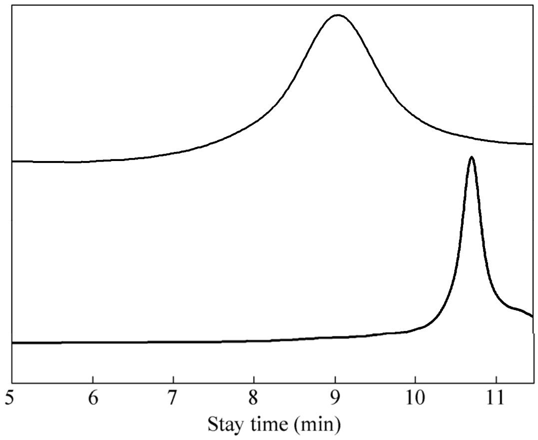


Fig. 5 The GPC data of PAA-3 (top) and PI-3 after degradation (bottom)

Thermal and Mechanical Properties

The polyimides could form tough and transparent films, as shown in Fig. 6 (insert). Additionally, for a degradable polymer, the thermal property is a matter of concern. DSC and TGA methods were applied to evaluate the thermal properties of the polyimides; the TGA curves of the polyimides are shown in Fig. 6, and thermal analysis data from the TGA and DSC curves of the polyimides are summarized in Table 3. The T_g values of PI-1 to PI-4 were in the range of 230–321 °C. As we expected, PI-1,2,3 showed a lower T_g because of the dianhydride used in these polymers decreased with increasing flexibility of the polyimide backbones. PI-4, derived from PMDA, exhibited the highest T_g because of the rigid pyromellitimide unit. The temperatures of the initial decomposition were determined to be in the range of 529–551 °C for all the polyimides tested. The temperatures of 10% gravimetric losses of the polyimides reached 541–575 °C in nitrogen. Also, the char yields at 800 °C for all the polymers were in the range of 47%–58 wt%. This suggests that this kind of polymer does not result in a loss of thermal properties. Meanwhile, all the polyimides can be processed into clear, flexible and tough films, as shown in Table 4, the polyimide films had tensile strengths of 83.7–110.8 MPa, elongations at break of 7%–14% and tensile modulus of 1.32–1.54 GPa, which indicated strong and tough materials.

Table 3. DSC and TGA data of the polyimides

Polyimide [a]	Thermal properties			
	T_g [b] (°C)	T_d [c] (°C)	T_{10} [c] (°C)	R_w [c] (%)
PI-1	235	539	541	47
PI-2	242	551	565	53
PI-3	230	529	545	52
PI-4	321	542	575	58

[a] Measured samples were obtained by thermal imidization method; [b] T_g was measured by DSC at a heating rate of 10 K/min in N_2; [c] T_d, decomposition-starting temperature; T_{10}, temperature at 10% weight loss. R_w, residual weight (%) at 800 °C in nitrogen.

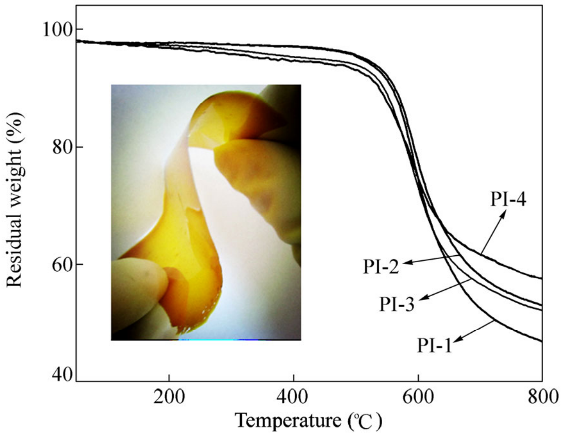


Fig. 6 TGA curves of the polyimides (insert is the film of PI-3.)

Table 4. Mechanical properties of the polyimides

Polymer	Tensile strength (MPa)	Elongation (%)	Modulus (GPa)
PI-1	93.8	10	1.39
PI-2	86.5	14	1.48
PI-3	110.8	12	1.54
PI-4	83.7	7	1.32

CONCLUSIONS

In this work, a new series of polyimides were obtained from the diamine *o*-PAP with various aromatic dianhydrides. The resulted polyimides could form tough films and showed excellent thermal property. Meanwhile, the polymer from *o*-diphenol based diamine monomer showed good solubility which could explained through examination of the X-ray structure of diamine *o*-PAP. Finally, the resulting polymer with phenyl-2-pyridyl ether structure can be degraded in a hydrazine hydrate medium.

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