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Synthesis and Properties of High Thermally Stable and Organosoluble Poly(pyridine imide)s with Pendent Diethylaminophenyl Groups

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Abstract: A novel diamine monomer 4-(4-diethylaminophenyl)-2,6-bis(4-aminophenyl)pyridine (APAP) was successfully synthesized, and then a series of poly(pyridine imide)s was prepared from APAP with five commercial dianhydrides: pyromellitic dianhydride (PMDA), 3,3′,4,4′-biphenyltetracarboxylic dianhydride (BPDA), 4,4′-oxydiphthalic anhydride (ODPA), 3,3′,4,4′-benzophenone tetracarboxylic dianhydride (BTDA), and 4,4′-(hexafluoroisopropylidene)diphthalic anhydride (6FDA) via a two-step solution polycondensation method. The structure of APAP was characterized by FT-IR, NMR, and MS. The resulting polymers have good solubility in polar solvents, such as NMP, DMF, and DMSO. Furthermore, they exhibited outstanding thermal stability. The glass transition temperature (T_g) of polymers range from 326 to 346 °C, and the temperature at 5 % and 10 % weight loss in the range of 527 °C to 570 °C and 550 °C to 601 °C, respectively. Moreover, they also presented excellent hydrophobicity with contact angels in the range of 88.4 ° to 94.0 °.

Keywords: Polyimides, Solubility, Thermal stability, Hydrophobicity, Pyridine

Introduction

Aromatic polyimides, as one of the most important of high-temperature polymers, have been widely applied in engineering fields for several decades because of their excellent combined properties, such as high mechanical strength, outstanding thermal stability, and excellent dielectric properties [1-8]. As it is well known, the conventional aromatic polyimides generally display deep color and poor solubility, which is caused by intramolecular or intermolecular charge transfer complex (CTC) formation in their conjugated molecular structures [9]. With the fast development of science and technology, the performances of the polymer materials are simultaneously put forward to high requirements. To overcome these defects, many researchers have made great efforts to focus on structural modification. The common approaches to increasing solubility include the attachment of bulky pendent [10-14], unsymmetrical structures [15-17], and flexible linkages [18,19] in the polymer chain. Liu *et al.* [13] designed and synthesized a series of novel polyimides containing bulky pendent groups and non-coplanar structures, and these polymers presented excellent solubility and transpatency. However, these strategies usually suffer from tradeoff between the thermal properties and the solubility of polyimides because the same structural features that enhance one will decrease the other [5]. Therefore, balance between the thermal properties and the solubility of polyimides become more important. The introduction of pyridine unit [20-29] in the main chain to increase thermal stability and heat resistance of polyimides is one of the frequently used methods. Obviously, pyridine would provide excellent thermal stability, good electronic, electron transporting, electron affinity characteristics, and more resistance to oxidation because of its molecular symmetry, aromaticity, as well as the polarizability resulting from the nitrogen atom in the pyridine ring [5]. Liaw *et al.* [22] have focused on synthesis of novel pyridine-containing polymers that have good thermostability and process stability.

Compared with the previous study, we hope to introduce pyridine units and bulky pendent substituents into polymer backbones, and then improve its solubility without sacrificing thermal stability. In this study, a novel diamine monomer 4-(4-diethylaminophenyl)-2,6-bis(4-aminophenyl)pyridine (APAP) was successfully synthesized by molecular structure design, and then used to prepare a series of poly(pyridine imide)s with five commercial aromatic dianhydrides via a two-step solution polymerization. The properties of the obtained polymers, including solubility, thermal stability, and hydrophobic properties, are studied.

Experimental

Materials and Chemicals

4-Diethylaminobenzaldehyde (Sinopharm Chemical Reagent Co., Ltd.), 4'-nitroacetophenone (Accela ChemBio Co., Ltd.), palladium 10 % on carbon (wetted with ca. 55 % water) (TCI), pyromellitic dianhydride (PMDA) (Sinopharm Chemical Reagent Co., Ltd.), 3,3′,4,4′-biphenyltetracarboxylic dianhydride (BPDA), 4,4′-oxydiphthalic anhydride (ODPA) (Changzhou Linchuan Chemical Co., Ltd.), 3,3′,4,4′-benzophenone tetracarboxylic dianhydride (BTDA), and

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(4,4′-hexafluoroisopropylidene)diphthalic anhydride (6FDA) (Tokyo Chemical Industry Co., Ltd.) were recrystallized from acetic anhydride and then dried in vacuum at 150 °C overnight before use. *N*,*N*-dimethyl formamide (DMF) and *N*-methyl-2-pyrrolidinone (NMP) were purified by vacuum distillation over calcium hydride before use. All other solvents were of analytical grade and were used without further purification.

Instrument and General Characterization

Fourier transform infrared (FT-IR) measurements were tested on a Thermo Nexus 470 FTIR spectrometer. ^{1}H NMR and ^{13}C NMR were measured using an Advance III 400 with deuterated dimethylsulfoxide (DMSO-d_6) as solvents. Mass spectrometry was recorded in an Elementar Vario EL III/Isoprime. Thermogravimetric analysis (TGA) was performed on a TGA Q500 analyzer in nitrogen at a heating rate of 10 °C/min. Differential scanning calorimetry (DSC) was measured on a DSC-204 phoenix thermal analyzer in a nitrogen atmosphere at a heating rate of 10 °C/min. Determination of solubility: 10 mg of polyimides dissolved in 1 m*l* of organic solvent to observe the characteristics of polymer dissolution at room temperature or heating conditions. The contact angle on the film for water was tested by a JY-PHb contact angle analysis instrument in room temperature. A crystallographic study of PI was performed at room temperature (about 25 °C) on an X'Pert PRO X-ray diffractometer (PANalytical B.V.). The X-ray diffraction (XRD) pattern was taken from 10 ° to 40 ° (2θ values) with Cu Kα radiation (λ=0.154 nm, operating at 40 kV and 40 mA). Intrinsic viscosities of polyamic acids (PAA) were tested by Ubbelohde viscometer and measured at a concentration of 0.5 g/d*l* in NMP at 25 °C.

Preparation of Monomer

Synthesis of 4-(4-diethylaminophenyl)-2,6-bis(4-nitrophenyl) pyridine

In a 250 m*l* round-bottomed, three-necked flask equipped with a condenser, a magnetic stirrer, and nitrogen inlet tube, a mixture of 4-diethylaminobenzaldehyde (10.00 g, 0.056 mol), 4'-nitroacetophenone (18.64 g, 0.11 mol), and ammonium acetate (52.19 g, 0.57 mol) in 130 m*l* acetic acid was refluxed at 125 °C and kept for 8 h. During the reaction, the progress of the reaction was followed by thin-layer chromatography. After that, the mixture was cooled to room temperature, and the precipitate was filtered off and washed with acetic, and then dried in a vacuum oven. Finally, the pure orange powder (APNP) was obtained by recrystalliztion from DMF, and then vacuum dried with a yield of 67 %.

^{1}H NMR (400 MHz, DMSO) δ: 8.60 (d, *J*=8.8 Hz, 4H), 8.37 (d, *J*=9.2 Hz, 4H), 7.96 (d, *J*=8.9 Hz, 2H), 7.93 (s, 2H), 6.79 (d, *J*=9.0 Hz, 2H), 3.47 to 3.38 (m, 4H), 1.13 (t, *J*=7.0 Hz, 6H); FTIR (KBr): 1587 cm^{-1}, 1347 cm^{-1} (-NO$_2$), 1676 cm^{-1}, 1444 cm^{-1} (pyridine); m.p. 203 °C (by DSC at a scan rate of 5 K/min).

Synthesis of 4-(4-diethylaminophenyl)-2,6-bis(4-aminophenyl) pyridine

The above mixture of APAP (10.50 g, 0.022 mol), palladium 10 % on carbon (wetted with ca. 55 % water) (4.2 g), and 200 m*l* anhydrous alcohol was put into a 250 m*l* round-bottomed, three-necked flask equipped with a condenser, a dropping funnel, a magnetic stirrer and nitrogen inlet tube, a 54 m*l* hydrazine hydrate was added drop by drop over a 1 h period through dropping funnel. The reaction mixture was refluxed at 85 °C for 8 h. The mixture was added to the excess terahydrofuran and immediately filtrated to remove Pd/C. The crude product was obtained by vacuum distillation to reduce the solvent. Finally, the pure diamine monomer, 4-(4-diethylaminophenyl)-2,6-bis(4-aminophenyl)pyridine (APAP) was obtained by crystallization from anhydrous alcohol/DMF (V_{alcohol}:V_{DMF}=10:1) and vacuum drying with yield 85 %.

^{1}H NMR (400 MHz, DMSO) δ: 7.97 (d, *J*=8.6 Hz, 4H), 7.76 (d, *J*=8.9 Hz, 2H), 7.69 (s, 2H), 6.74 (d, *J*=8.9 Hz, 2H), 6.65 (t, *J*=9.4 Hz, 4H), 5.38 (s, 4H), 3.37 (dd, *J*=12.8, 5.8 Hz, 4H), 1.10 (t, *J*=7.0 Hz, 6H). ^{13}C NMR (400 MHz, DMSO) δ: 156.96, 150.34, 149.14, 148.60, 128.54, 128.30, 127.60, 124.87, 114.30, 112.16, 111.91, 44.39, 13.15; FTIR (KBr): 3415 cm^{-1}, 3214 cm^{-1} (-NH$_2$), 1608 cm^{-1}, 1437 cm^{-1} (pyridine). MS (m/z): 409.23 ([M+H]$^+$); m.p. 108 °C (by DSC at a scan rate of 5 K/min).

Preparation of Poly(pyridine imide)s

Poly(pyridine imide)s were obtained from APAP reactioned with five commercially available dianhydrides through two-step solution polycondensation reaction. Typical reaction is as follows, in a two-necked flask with nitrogen inlet, a solution of diamine APAP (0.8171 g, 2.00 mmol) in NMP was prepared. And then, dianhydride PMDA (0.4362 g, 2.00 mmol) was added into the stirred solution at 5 °C under nitrogen. Stirring of mixture was kept for 24 h under nitrogen atmosphere at room temperature. The poly(amic acid) (PAA) was subsequently converted into poly(pyridine imide)s by a thermal imidization process at 80 °C for 8 h, 120 °C, 150 °C, 180 °C, 200 °C, 250 °C, 300 °C for 1 h each temperature. The poly(pyridine imide) films were put into water to peel off from the glass substrate, and then dried under vacuum at 150 °C. The other PAA solutions were prepared by similar procedure, and NMP solvent was used to control the PAA solutions in the range of 10 wt.% to 20 wt.%, respectively. Thermal imidization method was also adopted by the above method to obtained poly(pyridine imide) films. These poly(pyridine imide)s were named PI-1 (APAP and PMDA), PI-2 (APAP and BPDA), PI-3 (APAP and ODPA), PI-4 (APAP and BTDA), and PI-5 (APAP and 6FDA).

Results and Discussion

Monomer Synthesis

A new diamine monomer APAP was designed and synthesized by a two-step route according to Scheme 1. First, 4-diethylaminobenzaldehyde with 4'-nitroacetophenone reaction obtained an intermediate product, and then pure dinitro compound APNP by recrystallization was obtained from DMF. APNP was used to react with hydrazine monohydrate and Pd/C, and then pure diamine monomer APAP was obtained by recrystallization from the mixture of anhydrous alcohol and DMF. The chemical structure of APAP was identified by NMR, FT-IR, and MS spectrometry. As shown in Figure 1(a), the spectra FT-IR of APNP showed nitro group characteristic bands at 1587 cm^{-1} (asymmetrical stretching) and 1347 cm^{-1} (symmetrical), and a strong absorption band at 1676 cm^{-1} and 1444 cm^{-1} belonged to the pyridine ring. In Figure 1(b), the spectra FT-IR of APAP showed the characteristic absorptions of the nitro group disappeared and the typical N-H stretching absorption band of amino group appeared at 3415 cm^{-1} and 3214 cm^{-1}. The structure of APNP and APAP was also confirmed by NMR spectra. As shown in Figure 2, the signals in the range of 7.97 to 6.65 ppm were ascribed to the protons of the aromatic rings, and the -NH$_2$ proton was at 5.38 ppm in ^{1}H NMR of APAP. There are 17 resonance signals in the region of 156.96 to 13.15 ppm in the ^{13}C NMR spectrum, and three signals (156.96, 149.14, and 112.16 ppm) were also confirmed the formation of the pyridine ring. The data values of MS were consistent with the theoretical structure. The melting point of APAP was 108 °C by DSC measurement.

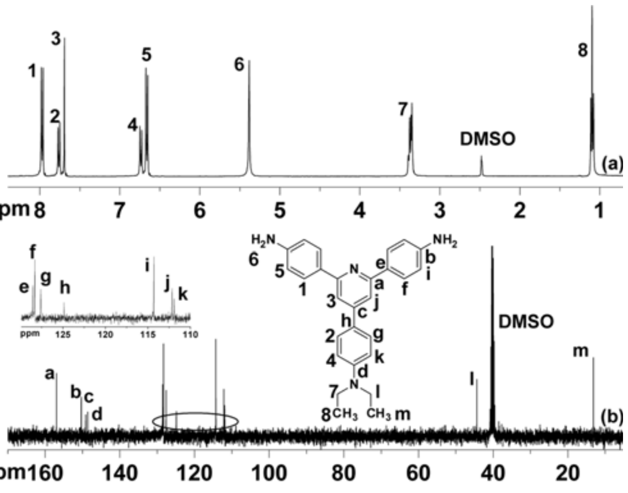
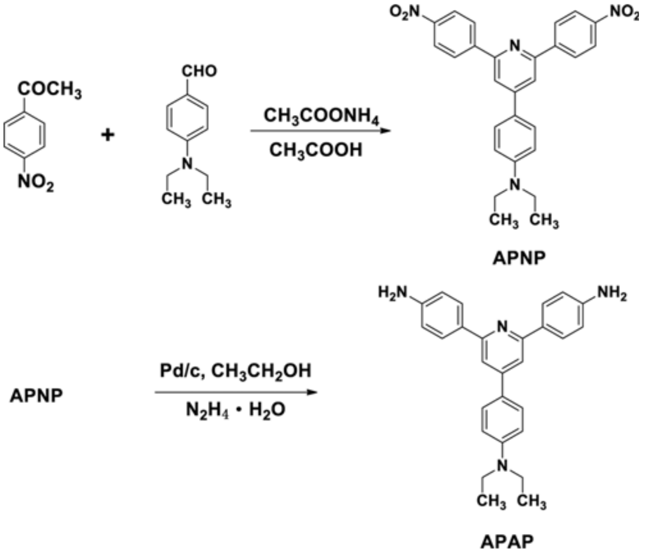


Figure 2. ^{1}H NMR (a) and ^{13}C NMR (b) spectra of APAP.



Scheme 1. Synthesis of diamine monomer APAP.

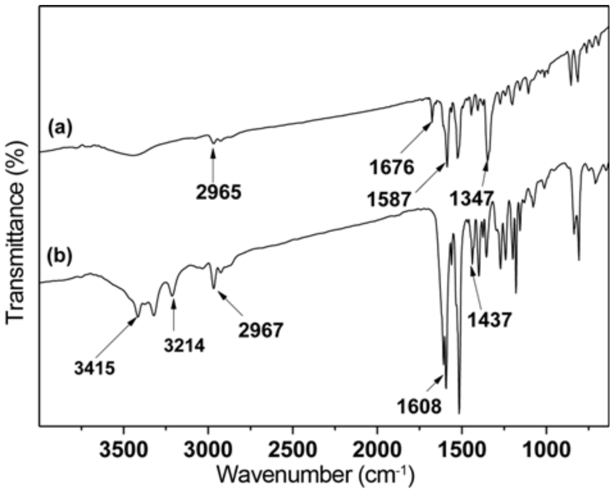
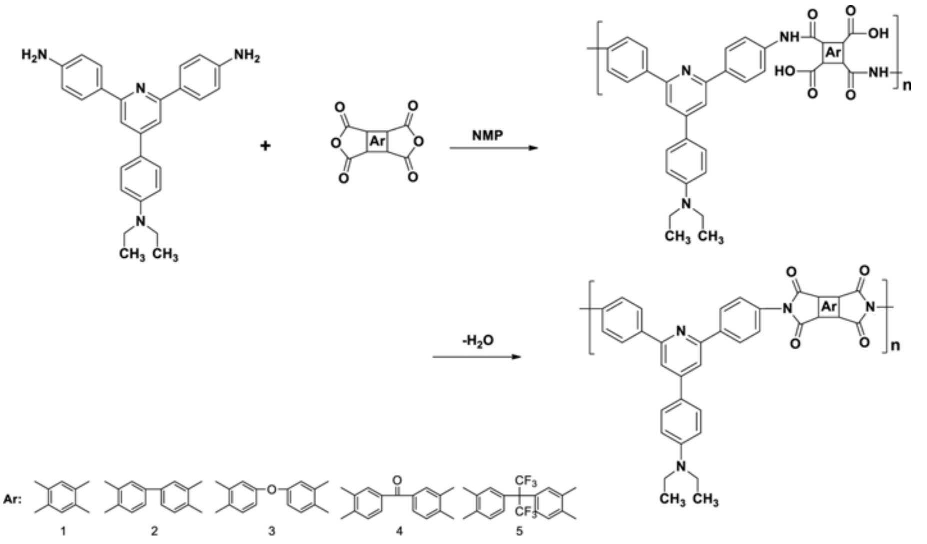


Figure 1. FT-IR spectra of APNP (a) and APAP (b).

Synthesis of Poly(pyridine imide)s

The poly(pyridine imide)s were synthesized by the two-step method in Scheme 2. First, polyamic acids (PAA) were obtained from the equimolar amount of aromatic dianhydrides and aromatic diamine APAP in NMP at room temperature under nitrogen atmosphere, and then the poly(pyridine imide)s were prepared by high-temperature imidization technique. The various characteristics of polymers were summarized in Table 1. Intrinsic viscosities of polyamic acids (PAA) were measured at a concentration of 0.5 g/d*l* in NMP at 25 °C. The values of intrinsic viscosity were in the range of 0.42 to 1.06 g/d*l*, which indicated polymers presenting high molecular weight. The representative FT-IR spectrum of the poly(pyridine imide)s was given in Figure 3. The FT-IR spectrum of polymers showed absorption at 1783 cm^{-1} (imide C=O symmetric stretching), 1721 cm^{-1} (imide C=O asymmetric stretching), 1367 cm^{-1} (imide C-N stretching), and 720 cm^{-1} (imide ring) associated with the structure of imide ring. The disappearance of strong



Scheme 2. Synthesis of poly(pyridine imide)s.

Table 1. Thermal properties, viscosity, and contact angles of poly(pyridine imide)s

Polymer	T_g (°C)[a]	T_d (°C)[b]	$T_{5\%}$ (°C)[c]	$T_{10\%}$ (°C)[c]	Char yield (%)[d]	η PAA (d*l*/g)	θ_w (°)
PI-1	346	549	555	591	69	0.66	89.8
PI-2	337	548	551	590	70	0.42	89.0
PI-3	326	551	570	601	74	0.52	93.0
PI-4	332	532	536	580	71	0.75	88.4
PI-5	336	511	527	550	66	1.06	94.0

[a]Glass transition temperature, obtained from DSC at a heating rate of 10 °C/min in N_2, [b]onset decomposition temperature, obtained from TGA at a heating rate of 10 °C/min in N_2, [c]temperature at 5 % and 10 % weight loss recorded by TGA at a heating rate of 10 °C/min in N_2, [d]char yield (wt%) at 800 °C in N_2.

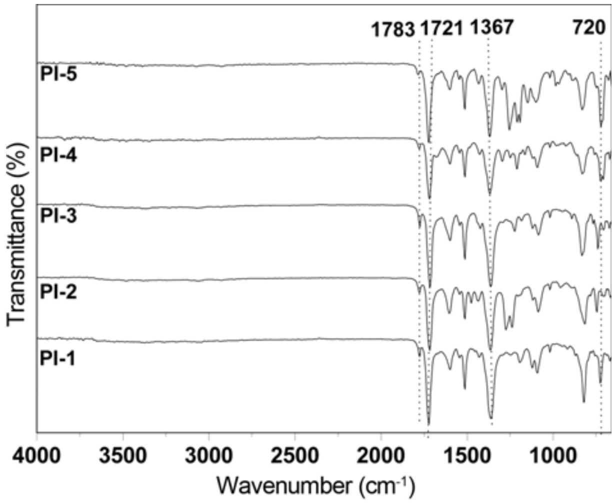


Figure 3. FT-IR spectra of poly(pyridine imide)s.

absorption corresponding to amino group at 3415 cm^{-1} also proved the complete thermal imidization of polymers.

Polymer Solubility

The solubility of polymers was tested in various organic solvents with a polymer concentration of 10 mg/m*l* at room temperature or upon heating, and the results are shown in Table 2. The obtained poly(pyridine imide)s presented good solubility. Usually, polymer solubility has a close relationship with their structure, such as bulky pendant groups, flexible linkages (-O-, -C(CF_3)$_2$-), non-coplanar structure, asymmetric structure, and so on, in the polymer chain, which are all beneficial to promote the solubility of the polymer. Obviously, the introduction of bulky pendent diethylaminophenyl groups into polyimide backbones improved the solubility of polymers, which can effectively prevent the coplanarity of aromatic rings and reduce the packing efficiency of molecular chains. Furthermore, PI-1 based on PMDA and PI-2 based on BPDA showed partial solubility on heating in DMF, DMSO, and NMP due to the introduction of rigid pyromellitic and biphenylic units in the polymer chains, thereby making it difficult for the solvent to attack. PI-3 derived from ODPA presented solubility on heating in DMF and NMP, and

Table 2. Solubility of poly(pyridine imide)s[a]

Polymer	Solvent[b]					
	DMF	DMSO	NMP	$CHCl_3$	THF	CH_2Cl_2
PI-1	+-	+-	+-	--	--	--
PI-2	+-	+-	+-	--	--	--
PI-3	+	+-	+	--	+-	--
PI-4	+-	+-	+-	--	--	--
PI-5	+	+-	++	--	+-	--

[a]Solubility was tested with a polymer concentration of 10 mg/m*l* in solvent with stirring. ++: soluble at room temperature; +: soluble on heating; +-: partially soluble on heating; --: insoluble. [b]DMF, *N,N*-dimethylformamide; DMSO, dimethyl sulfoxide; NMP, *N*-methyl-2-pyrrolidone; $CHCl_3$, chloroform; THF, tetrahydrofuran; CH_2Cl_2, dichloromethane.

partial solubility on heating in DMSO and THF because of the incorporation of the flexible group oxygen in the polymer chain, which loosened the packing density of polymer chain and decreased the intrinsic molecular interactions, and then increased the fractional free volume. PI-4 came from BTDA exhibited partially solubility on heating in DMF, DMSO, and NMP, and it may be attributed to the presence of carbonyl units in the polymer backbones. Furthermore, PI-5, which stemmed from 6FDA, displayed relatively good solubility, such as solubility at room temperature in THF, solubility on heating in DMF, and partial solubility on heating in DMSO and THF, which resulted in the introduction of hexafluoroisopropylidene groups in the polymer chains, and inhibited close packing and reduced the interchain interaction, and then enhanced polymer solubility.

Polymer Thermal Properties

The resulting of thermal properties was investigated by thermal gravimetric analyses (TGA) and differential scanning calorimetry (DSC). The DSC curves are shown in Figure 4, and data are summarized in Table 1. The values of T_g were measured in the range of 326 °C to 346 °C. Poly(pyridine imide)s presented high T_g because of the incorporation of rigid imide rings and pyridine units into polymer chains, which is better than that of pyridine-free polyimides. Obviously, PI-1 based on PMDA presented the highest T_g, which attributed to the rigid pyromellitic units. However, PI-3 showed a minimum T_g value than that of PI-2, PI-4, and PI-5 because of the flexible ether linkages moieties derived from ODPA that were introduced into the polymer backbone. As shown in Figure 5, the onset decomposition temperatures of the resulting poly(pyridine imide)s were in range of 511 °C to 551 °C, and the temperature of 5 % and 10 % weight loss were in the area of 527 °C to 570 °C and 550 °C to 601 °C under nitrogen atmosphere, respectively. Meanwhile, the residual weight at 800 °C for the resulting poly(pyridine imide)s was all above 66 %. These polymers exhibited excellent thermal properties due to the introduction of rigid pyridine ring and (diethylaminophenyl groups) into polymer backbones.

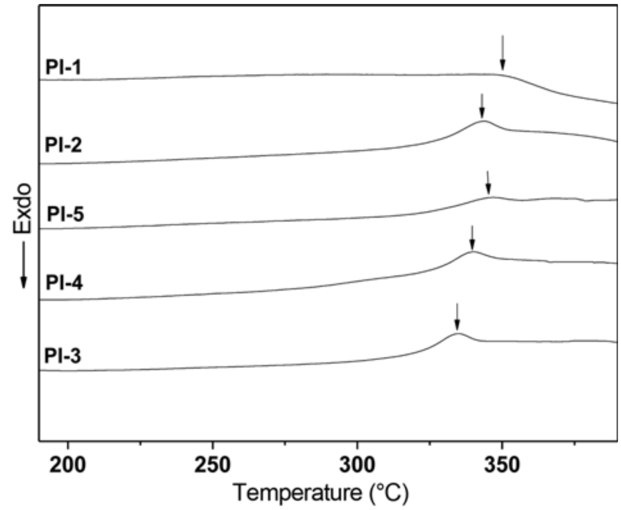


Figure 4. DSC curves of poly(pyridine imide)s.

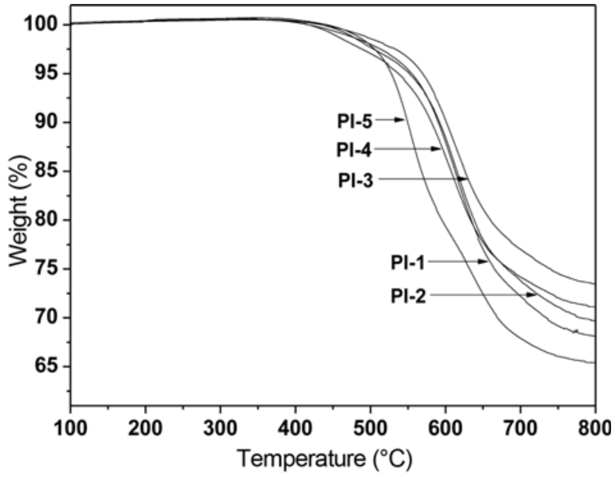


Figure 5. TGA curves of poly(pyridine imide)s.

Wide-angle X-ray Diffractograms of Polymer

The prepared poly(pyridine imide) films were examined for crystallinity by using wide-angle X-ray diffractograms

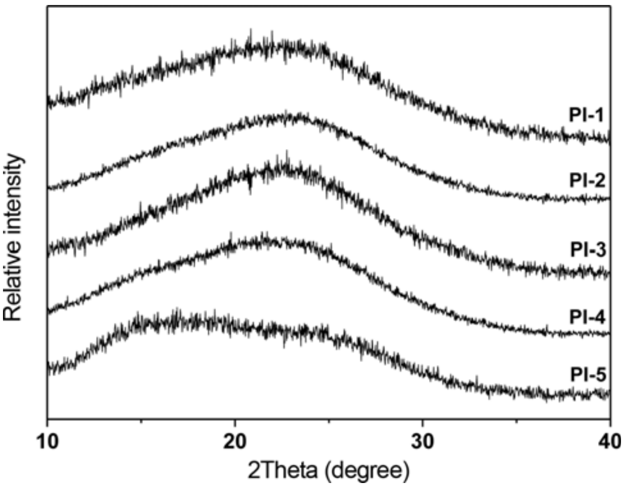


Figure 6. XRD patterns of the poly(pyridine imide)s.

analysis (WAXD) with graphite monochromatized Cu Kα radiation and with 2θ ranging from 10° to 40°. As shown in Figure 6, the representative X-ray diffractograms showed a wide dispersion peak, indicating that all the polymers are amorphous. The formation of amorphous structure is attributed to the presence of bulky pendent groups in the main backbone, which reduced the interchain cohesion and led to the formation of amorphous structure. Obviously, no melting endothermic behavior was observed except the T_g in Figure 4, which also agreed with the analysis results of WAXD.

Hydrophobic Properties of Polymer

Polyimide materials often exhibited higher water absorption than that of hydrocarbon polymers because of the existence of imide group in the main chains [30]. As shown in Figure 7, the contact angles of polyimide Kapton and poly(pyridine imide)s for water (θ_w) are listed in Table 1. Generally, the higher contact angle the polymers have, the better hydrophobicity the polymers will present. The contact angles of the obtained poly(pyridine imide)s were in the range of 88.4° to 94.0°, which is higher than that of the commercial Kapton (81.6°). Poly(pyridine imide)s presented excellent hydrophobicity, which may be because of the incorporation of hydrophobic groups, such as alkyl groups, pyridine and bulky pendent substituents, and so on. Obviously, PI-5 derived from 6FDA presented the highest hydrophobic property. It is because of the introduction of fluorine-containing group from dianhydride units into the polymer backbones, which reduced the surface energy of the polymer, and then exhibited good hydrophobicity. The contact angle of PI-4, which originated from BTDA, presented the lowest hydrophobicity, it may be because the carbonyl from BTDA easily forms hydrogen bonds with water.

Conclusion

A series of novel poly(pyridine-imide)s containing bulky pendant diethylaminophenyls and heterocyclic pyridines was successfully synthesized and characterized. These polymers showed good solubility, and they presented outstanding thermal stability with high glass transition temperature (T_g) and decomposition temperature. Moreover, they also exhibited excellent hydrophobic properties. These polymers may become potential promising high-performance polymeric materials for use in heat-resistant materials and microelectronics applications.

Acknowledgments

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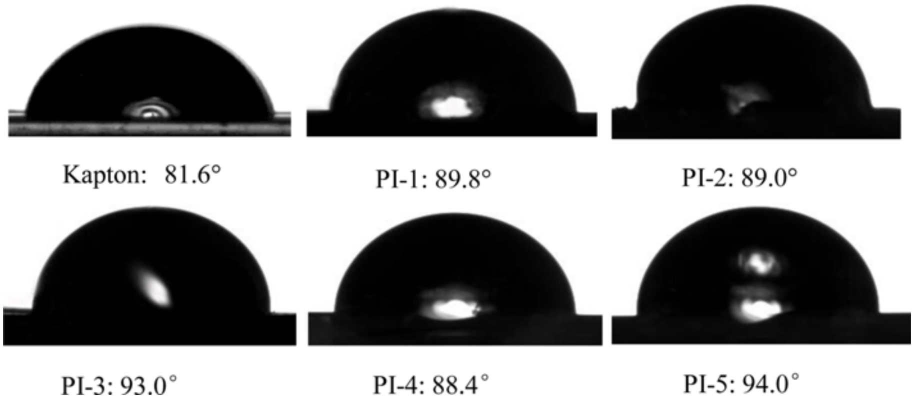

Figure 7. Contact angle of Kapton and poly(pyridine imide) films.

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