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New Soluble Polyimides with High Optical Transparency and Light Color Containing Pendant Trifluoromethyl and Methyl Groups

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A new aromatic diamine containing trifluoromethyl and methyl groups, namely α,α-bis(4-amino-3-methylphenyl)-4-(trifluoromethyl)phenylmethane (**1**), was synthesized from 2-methylaniline and 4-(trifluoromethyl)-benzaldehyde. A series of fluorinated polyimides (PIs) were prepared from the diamine with four commercially available aromatic tetracarboxylic dianhydrides using a one-step high-temperature polycondensation procedure. These obtained PIs showed excellent solubility, with the dissolvability at a concentration of 10 wt% in most solvents, and they could afford flexible and strong films. Thin films of these PIs exhibited high optical transparency and light color, with the cutoff wavelength at 324—357 nm and transmittance higher than 74% at 450 nm. Moreover, these PIs possessed eminent thermal stability and good mechanical properties.

Keywords fluorinated polyimides, solubility, optical transparency

Introduction

Functional polyimide films with high optical transparency and light color are a class of important advanced materials and play crucial roles for some applications in liquid crystal display devices, flexible solar cell substrates, optical waveguides and so on.[1-6] However, the wholly aromatic polyimides (PIs) are often pale yellow or deep reddish yellow and have strong absorption in the visible region observed from their UV-visible spectra because of their high conjugated aromatic structures and/or the intermolecular charge-transfer complex (CTC) formation.[7] Moreover, most aromatic PIs are difficult for both solution-casting and melt processing due to their poor solubility in organic solvents and high melting temperatures, which highly restricts their application in many fields.[8] Purely commercial PIs derived from pyromellitic dianhydride (PMDA) or 3,3',4,4'-biphenyltetracarboxylic dianhydride (BPDA), for instance, are soluble only in a few strongly protic acids. To overcome these problems, much research efforts have been focused on the synthesis of soluble and optically transparent PIs without much deterioration of their own excellent properties. Several successful approaches including insertion of flexible linkage and bulky substituents on the main chain or utilization of noncoplanar or unsymmetrical monomers have been developed.[9-19]

Recently, considerable attention has been devoted to the fluorinated PIs, especially the trifluoromethyl-containing poly(ether imide)s (PEIs).[14,18,20-26] As well-known, the bulky free volume and high electronegativity of the fluorinated groups could endow aromatic PIs with many attractive features, such as good organo-solubility, high optical transparency, light color as well as low water uptake and dielectric constants.[27] Thus, the develepment of novel trifluoromethyl substituted monomers for functional PIs has become a subject of intense research interest. In this study, a new CF_3-containing aromatic diamine was synthesized in high yield by a facile condensation reaction. Subsequently, a series of fluorinated PIs with pendant trifluoromethyl and methyl groups were prepared from the diamine and different aromatic dianhydrides. The fluorinated PIs should have diminished aromatic conjugation and intermolecular CTC formation because of incorporation of bulky free volume and noncoplanar structure into the polymer backbones. The effects of the structures on the properties of the PIs were detailedly investigated and discussed.

Experimental

Materials

2-Methylaniline, 4-(trifluoromethyl)-benzaldehyde

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and trifluoromethanesulfonic acid purchased from Shanghai Darui Finechemical Co. Ltd were used as received. Commercially available aromatic tetracarboxylic dianhydrides such as pyromellitic dianhydride (PMDA or **2a**), 3,3',4,4'-biphenyltetracarboxylic dianhydride (BPDA or **2b**), 3,3',4,4'-benzophenone-tetracarboxylic dianhydride (BTDA or **2c**), and 4,4'-oxydiphthalic dianhydride (ODPA or **2d**) were all purified by recrystallization from acetic anhydride and then dried *in vacuo* at 180 ℃ for 10 h. Commercially available *N*-methyl-2-pyrrolidinone (NMP), *m*-cresol, *N*,*N*-dimethylformamide (DMF), *N*,*N*-dimethyl-acetamide (DMAc), dimethyl sulfoxide (DMSO), tetrahydrofuran (THF) and other solvents were purified by distillation prior to use.

Measurements

NMR spectra were measured on a Bruker AV400 instrument with dimethyl sulfoxide-d_6 (DMSO-d_6) or $CDCl_3$ as solvent and tetramethylsilane (TMS) as an internal standard. FT-IR spectra were recorded on a Nicolet NEXUS 670 spectrometer. Elemental analysis was carried out on an Elmentar Vario EL system. Gel permeation chromatography-Light scattering (GPC-LS) on soluble polymers was performed on a BI-MwA system using THF as the eluent. Differential scanning calorimetric (DSC) analysis was performed on a PE Diamond DSC instrument at a heating rate of 20 ℃/min in nitrogen and air atmosphere. Glass transition temperatures were read at the middle of the transition in the heat capacity from the second heating scan after cooling from 400 ℃ at a cooling rate of 20 ℃/min. Thermogravimetric analysis (TGA) of the polymer samples was measured on a Netzsch TG 209F1 instrument at a heating rate of 20 ℃/min in nitrogen and air atmosphere. Ultraviolet-visible spectra of the polymer films were recorded on a PerkinElmer Lambda 35 UV-Visible spectrophotometer at room temperature. The tensile properties were performed on an Instron 5565 Tensile Apparatus with a 5 kg load cell at a crosshead speed of 5 mm/min on strips approximately 40—60 um thick and 0.5 cm wide with a 2 cm gauge length. An average of at least five individual determinations was used.

Monomer synthesis

In a 100 mL, four-necked, round-bottomed flask equipped with a mechanical stirrer, a condenser and a nitrogen inlet, 2-methylaniline (25.72 g, 0.24 mol) was added and heated to reflux under nitrogen atmosphere. 4-(Trifluoromethyl)benzaldehyde (17.41 g, 0.10 mol) dissolved in 4 mL (12 mol/L) of hydrochloric acid was added dropwise over a period of 1 h. After the completion of addition, the reaction mixture was maintained at reflux (about 150 ℃) for another 10 h. The reaction mixture was then cooled to 60 ℃, and 100 mL of 15% aqueous solution of sodium hydroxide was added to the resulting suspension. The mixture was poured into methanol to precipitate a pale-white solid which was filtered, washed repeatedly with water. The crude product was further purified by recrystallization from toluene to give white crystals (31.48 g, 85%); m.p. 188—189 ℃ by DSC (10 ℃/min).

FT-IR (KBr) ν: 3441, 3354, 2924, 1638, 1598, 1163, 1104, 1058 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6) δ: 7.57 (d, $J=8.0$ Hz, 2H), 7.23 (d, $J=8.0$ Hz, 2H), 6.63 (s, 2H), 6.56 (d, $J=8.0$ Hz, 2H), 6.49 (d, $J=8.0$ Hz, 2H), 5.22 (s, 1H), 4.65 (s, 4H), 1.94 (s, 6H). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 151.2, 145.2, 131.8, 131.0, 130.0, 127.4, 126.8 ($J_{C-F}=30.8$ Hz), 125.3, 124.9 ($J_{C-F}=270.2$ Hz), 121.4, 114.3, 54.9, 18.0. Anal. calcd for $C_{22}H_{21}F_3N_2$ (370.41): C 71.34, H 5.71, N 7.56; found C 71.25, H 5.80, N 7.49.

Polymer synthesis

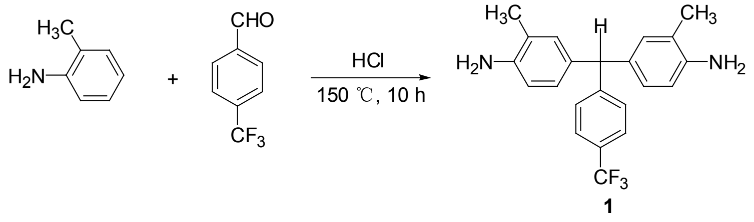
The general procedure for the preparation of the fluorinated PIs **3a**—**3d** was illustrated as follows. Diamine **1** (2.0 mmol) and dianhydride monomer **2a**—**2d** (2.0 mmol) were first dissolved in 15.0 mL of *m*-cresol in a 50 mL three-necked round-bottomed flask. After the mixture was stirred at room temperature for 30 min, isoquinoline (*ca.* 5 drops) was added, and further stirred for 3 h at 120 ℃. Then the mixture was heated at 185 ℃ for 8 h. Water formed during the imidization was continuously removed with a stream of nitrogen. The whole polymerization proceeded smoothly, and homogeneous, transparent, viscous polymer solutions were produced. White, fibrous polymer resins were obtained when the resultant polymer solution was poured into an excess of ethanol. The polymer was separated by filtration and washed with ethanol for several times and dried in a vacuum oven at 120 ℃ for 10 h.

Results and Discussion

Monomer synthesis

New CF_3-containing aromatic diamine, α,α-bis(4-amino-3-methylphenyl)-4-(trifluoromethyl)-phenylmethane (**1**), was synthesized via a condensation reaction of 4-(trifluoromethyl)benzaldehyde and 2-methylaniline catalyzed by hydrochloric acid at 150 ℃, as shown in Scheme 1. The results of FT-IR, and NMR spectra, and elemental analysis all confirmed the chemical structure of the diamine monomer. The FT-IR spectrum of diamine **1** showed the characteristic absorptions of the N—H stretching band of the amino groups at 3300—3500 cm^{-1} and the C—F stretching at about 1050—1200 cm^{-1}. The ^1H and ^{13}C NMR spectra of diamine **1** were shown in Figure 1a and 1b, respectively. In Figure 1a, the signal at δ 4.65 was peculiar to the protons of amino groups and the signal at δ 1.94 corresponded to the protons of methyl groups. In the ^{13}C NMR spectrum, all the carbon-13 atoms resonated in the region of δ 15—155, and there appeared clear quatets because the C—F coupling. The coupling effect decreased with the increase of the distance between C and F atoms. For instance, C-12 ($J_{C-F}=270.2$ Hz) showed much stronger coupling effect with F than C-1 ($J_{C-F}=30.8$ Hz). Moreover, the

Scheme 1 Synthesis of aromatic diamine **1**



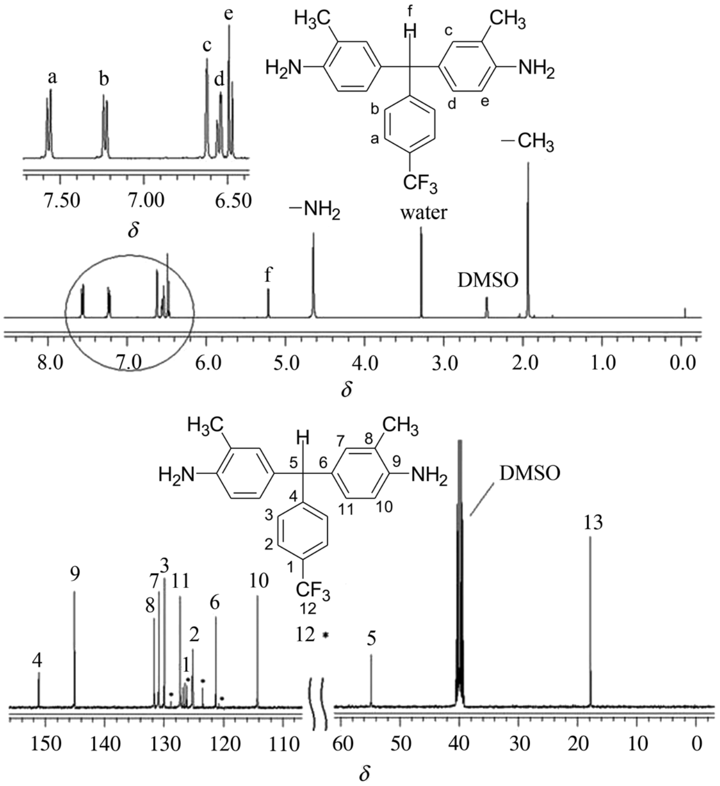


Figure 1 ^1H and ^{13}C NMR spectra of diamine **1** in DMSO-d_6.

elemental analysis values were in good agreement with the calculated values. All the above characterizations indicated the successful preparation of the fluorinated diamine monomer **1**.

Polymer synthesis

Generally, there are two main synthetic methods to PIs, namely two-step and one-step polymerizations. Commercial PIs are mainly prepared by two-step polymerizations. In this method, the poly(amic acid) (PAA) was first prepared from dianhydride and diamine in a polar aprotic solvent, then the PAA was cyclodehydrated at elevated temperatures or by adding a cyclizing agent such as acetic anhydride. Compared to two-step polymerizations, one-step polymerization is more convenient and fully cyclized PIs were obtained directly from their corresponding teteracarboxylic acid dianhydride and diamine using high boiling solvents. This method was usually used when working with soluble PIs and is considered more practical for polymerizing less reactive dianhydrides and diamines. In this study, all PIs **3a**—**3d** can be prepared conveniently from aromatic diamine **1** and various aromatic dianhydrides **2a**—**2d** (PMDA, BPDA, BTDA and ODPA) via a one-step, high-temperature solution polycondensation procedure, as shown in Scheme 2. The polymerization of diamine **1** with aromatic dianhydrides **2a**—**2d** proceeded smoothly at 185 ℃, and homogeneous, transparent, viscous polymer solutions were produced. White or pale yellow, and fibrous polymer resins were obtained when the resultant polymer solution was poured into an excess of ethanol. The PIs were obtained in almost quantitative yields and the inherent viscosities were between 0.54 and 0.87 $dL \cdot g^{-1}$ in DMAc solution (Table 1). The weight-average molecular weights (M_w) and number average molecular weights (M_n) were measured based on GPC (Table 1), the M_w and M_n values of the PIs dissolved in THF were 41400 to 105200 and 20900 to 57500, respectively. The polydispersity index (PDI) of the **3** series was in the range of 1.77—1.98. These data indicate that the obtained PIs have moderate molecular weights and relatively narrow PDI.

Scheme 2 Synthesis of fluorinated PIs **3a**—**3d**

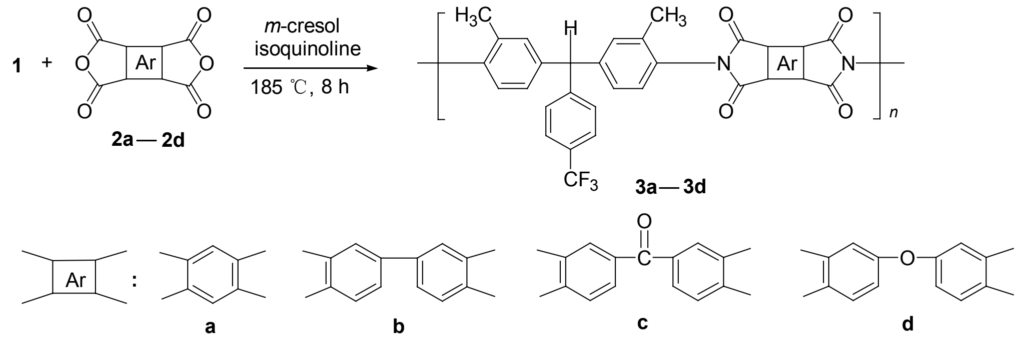


Table 1 Inherent viscosities[a] and GPC-LS molecular weights of fluorinated PIs

Polymer	η_{inh}/(dL•g^{-1})	GPC-LS data		
		$M_n\times10^{-4}$	$M_w\times10^{-4}$	PDI
3a	0.54	2.09	4.14	1.98
3b	0.87	5.75	10.52	1.83
3c	0.70	4.35	8.27	1.90
3d	0.62	3.86	6.83	1.77

[a] Measured at a concentration of 0.5 g/dL in DMAc at 30 ℃.

The structures of the PIs **3a**—**3d** were characterized by FT-IR and ^{1}H NMR spectra. All the PIs showed characteristic imide absorption bands around 1782 and 1730 cm^{-1} (imide carbonyl asymmetrical and symmetrical stretchings), 1374 cm^{-1} (C—N stretching), and 1040 and 735 cm^{-1} (imide ring deformation), together with strong C—F stretching absorption peak at about 1100 cm^{-1}, while the amino groups at the region of 3300—3500 cm^{-1} disappeared. A representative of FT-IR and ^{1}H NMR spectra of PI **3b** are illustrated in Figure 2 and 3 respectively. The assignments of each proton designated in the ^{1}H NMR spectrum are in complete agreement with the proposed polymer structures. The sharp peak at δ 4.65 corresponding to the amine protons in ^{1}H NMR spectrum of diamine **1** (Figure 1a) disappears completely here, and new peaks appear at δ 8.26 and δ 8.08—8.13 which correspond to the protons in the dianhydride units. All the results confirm the complete imidization between diamine and dianhydrides.

Polymer solubility

The solubility of these fluorinated PIs was tested qualitatively in various organic solvents, and the results are reported in Table 2. The obtained PIs **3a**—**3d** were soluble and had high dissolvability at a concentration of

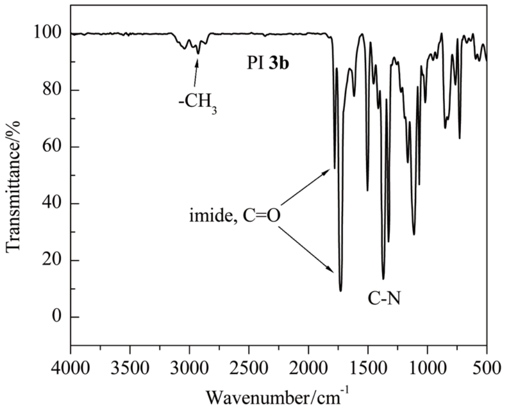


Figure 2 FT-IR spectrum of PI **3b**.

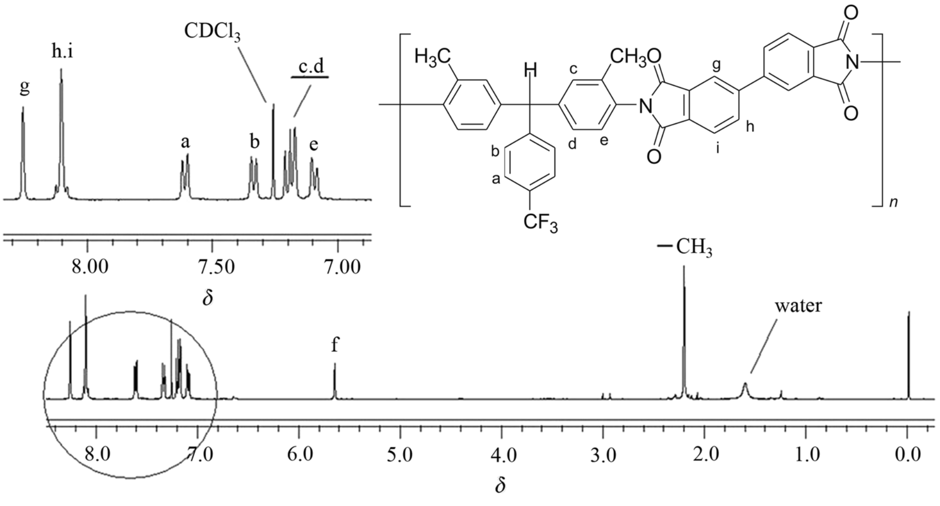


Figure 3 ^{1}H NMR spectrum of PI **3b** in CDCl$_3$.

10 wt% in most tested solvents. It is surprising that PI **3a** and **3b**, which were prepared from the very rigid dianhydrides PMDA and BPDA, respectively, were quickly soluble in NMP, DMAc, $CHCl_3$, CH_2Cl_2 and THF at room temperature. It should be noted that good solubility in low-boiling-point solvents is critical for preparing PI films or coatings at a relatively low processing temperature, which is desirable for advanced manufacturing applications. The good solubility of these PIs was governed by the structural modification through incorporation of the bulky 4-(trifluoromethyl)phenyl groups in the polymer backbone, which could decrease the packing density and intermolecular interactions of macromolecular chain, thereby enhancing solubility. Moreover, the methyl group introduced at the *ortho*-position of nitrogen in the polymer chains appeared to hinder the rotation of C—N imide bond and force the two aromatic rings to adopt a noncoplanar structure, which was also effective to improve solubility.

Table 2 Solubility behavior of fluorinated PIs[a]

Polymer	NMP	DMAc	DMF	DMSO	$CHCl_3$	CH_2Cl_2	THF	Acetone
3a	○	++	+	+	○	++	○	−
3b	○	○	++	+	○	○	○	−
3c	○	○	○	++	○	○	○	−
3d	○	○	○	○	○	○	○	S

[a] ○, 100 mg sample dissolved in 1 mL solvent (10 wt%); ++, soluble at 5 wt%; +, soluble at 1 wt%; S, swelling; −, insoluble.

Thermal properties

DSC and TGA were used to evaluate the thermal properties of the PIs **3a**—**3d**. The results are summarized in Table 3. It was inspiring that these PIs showed extra high T_g, and the values were all beyond 300 ℃. PI **3a** based on rigid dianhydride PMDA even shows no distinct T_g at determined temperature region. Apparently, the **3** series exhibited 30—60 ℃ of T_g increment compared with those of structurally fluorinated PEIs derived from the trifluoromethyl-substituted bis(ether amine)s and corresponding dianhydrides.[20-26] High T_g values mainly originated from the inherent rigidity of these macromolecular backbones. Meanwhile, the introduction of methyl substituent into the *ortho*-position of the imide nitrogen hinders the rotation of the two aromatic rings around C—N bonds, so further increases the T_g values.

The thermal and thermooxidative stabilities of these PIs were evaluated in both nitrogen and air atmospheres. Typical TGA curves for PI **3b** are reproduced in Figure 4. These PIs showed good thermal stability and had no notable weight loss below 510 ℃ in nitrogen atmosphere. The 10% weight loss temperatures (T_{10}) in nitrogen and in air atmospheres stayed in the range of 529—541 ℃ and 447—465 ℃, respectively. They left more than 55% char yield at 800 ℃ in nitrogen. The TGA data indicated that these PIs had fairly high thermal stability even with the introduction of methyl and trifluoromethyl pendent groups.

Table 3 Thermal properties of PIs **3a**—**3d**

Polymer	T_g[a]/℃	T_d[b]/℃	T_{10}[c]/℃		Char yield[d]/%
			In N_2	In air	
3a	no	518	538	450	58
3b	348	517	541	465	56
3c	324	516	534	453	58
3d	309	519	529	447	55

[a] From the second trace of DSC measurements conducted at a heating rate of 20 ℃/min. [b] Onset decomposition temperature at 20 ℃/min heating rate. [c] 10% weight loss temperature in TGA at 20 ℃/min heating rate. [d] Residual weight retention at 800 ℃.

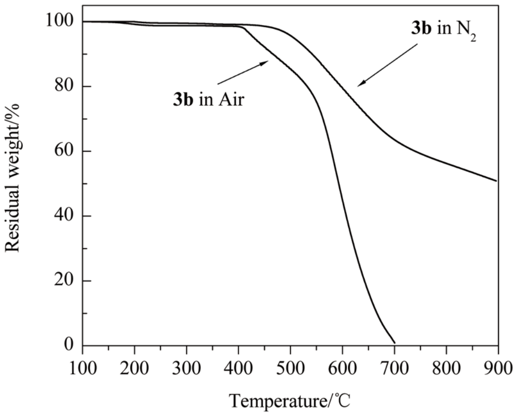


Figure 4 TGA curves of PI **3b** at a heating rate of 20 ℃/min.

Optical and mechanical properties

As depicted in introduction, most commercially aromatic PIs display rather deep yellow and strong absorption at UV-visible spectra because of their high conjugated aromatic structures and/or the intermolecular CTC formation. Unlike those commercial PIs, PI **3a**—**3d** films exhibit high optical transparency at UV-visible spectra and nearly colorless. Thin films were measured for optical transparency with UV-visible spectroscopy. Figure 5 shows the UV-visible transmittance spectra of these PIs, and the cutoff wavelength (λ_{cutoff}) values and the percentages transmittance at 450 nm from these spectra are listed in Table 4. All the PI films had shorter λ_{cutoff} than 360 nm, and exhibited high optical transparency of 74.4%—81.5%. For comparison, a photograph of one piece of commercial Kapton and one piece of PI 4d film is shown in Figure 6. The films were photographed against a special background to highlight the transparency. It is easy to find that PI **3d** film exhibited much lighter color than Kapton film. The good optical transparency and light color of these PIs were mainly attributed to the reducing of the conjugation and intermolecular CTC formation along the PI backbone. It may be concluded that bulky pendant groups together with the noncoplanar structure and methane moieties in the polymer chains were effective to reduce the conjugation and intermolecular CTC formation, so to improve the optical properties.

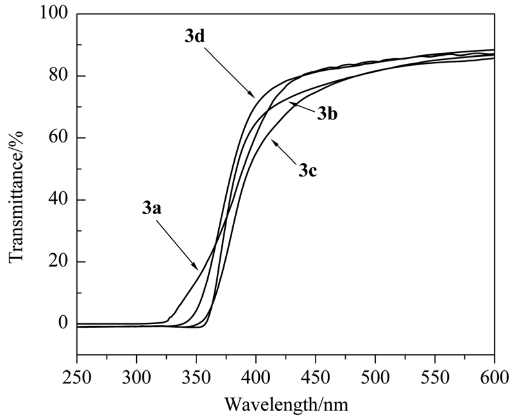


Figure 5 UV-visible spectra of the PI films (about 30—40 μm thickness).

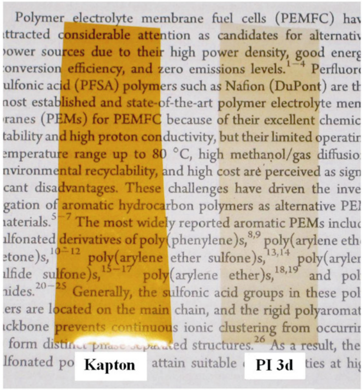


Figure 6 Photographs of a piece of the Kapton film (35 μm thickness) and a piece of the PI **3d** film (40 μm thickness).

Table 4 Optical and mechanical properties of the PI films

Polymer	λ_{cutoff}/nm	T_{450}/%	Tensile strength/MPa	Elongation to break/%	Young's modulus/GPa
3a	324	81.5	69.7	5.6	2.8
3b	357	76.3	90.6	7.4	2.8
3c	350	74.4	79.1	5.9	2.6
3d	338	81.3	80.5	11.7	2.4

The tensile properties of these fluorinated PIs are also reported in Table 4. The tensile strengths at maximum load, elongations at break, and initial moduli of them were in the range of 69.7—90.6 MPa, 5.6%—11.7% and 2.4—2.8 GPa, respectively. The results indicated these fluorinated PI films were tough enough for potential use as excellent high-T_g and optically transparent materials for advanced applications.

Conclusions

A new CF_3-containing aromatic diamine has been successfully synthesized in high yield by a facile 2-methylaniline condensation reaction. A series of fluorinated PIs with bulky free volume and noncoplanar structure, were prepared by one-step high-temperature polycondensation procedure. The obtained PIs were characterized by excellent solubility, good film-forming capability, high thermal stability and optical transparency, light color, and good mechanical properties. The eminent combination of several properties guaranteed the obtained PIs as potential high-temperature resistant materials for optical and photoelectric applications.

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