Synthesis and Properties of Novel Poly(benzimidazopyrrolone amide)s Containing Pyridine Moieties

Xiaolong Wang,[1,2] Yanfeng Li,[1] Chenliang Gong,[1] Tao Ma,[1] Shujiang Zhang[1]

[1]State Key Laboratory of Applied Organic Chemistry, College of Chemistry and Chemical Engineering, Institute of Biochemical Engineering and Environmental Technology, Lanzhou University, Lanzhou 730000, China
[2]State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, China

ABSTRACT: A series of new poly(benzimidazopyrrolone amide) (PPA) copolymers were synthesized by a two-step procedure, which was the solution polycondensation of a novel pyridine-containing tetraamine with various aromatic dianhydrides at a room temperature and cyclization of the resulting prepolymers at a high temperature, respectively. The resulting prepolymers from the solution polycondensation, that is, poly(amide amino acid)s (PAAAs), had inherent viscosities of 0.82–0.91 dL/g; then, tough and flexible PPA films could be successfully prepared by the casting of the PAAA solutions onto a glass substrate followed by thermal curing with a program temperature procedure up to 350°C. The obtained PPA films exhibited not only excellent thermal properties with onset decomposition temperatures in the range 502–521°C, glass-transition temperatures in the range 299–337°C, and residual weight retentions at 700°C in air of 29.1–34.8% but also good mechanical properties with tensile strengths of 102.1–115.9 MPa and elongations at break of 6.8–7.4%. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 3369–3375, 2008

Key words: heteroatom-containing polymers; high-performance polymers; thermal properties

INTRODUCTION

Polybenzimidazopyrrolones, abbreviated as polypyrrolones, have been known as promising high-temperature heteroaromatic polymer materials because of their outstanding thermal and chemical stabilities, which are derived from their rigid and multiaromatic structures; however, their rigid semiladder structures also cause their poor processability and flexibility, which greatly limits their practical application in advanced technologies.[1–7] Therefore, some efforts to improve the processability and flexibility of polypyrrolones, including the polycondensation of biphenyl or bridged biphenyl dianhydrides with aromatic tetraamines to yield semiladder structures[4–6,8] and the incorporation of imide or imidazole segments into the polymer backbone to produce linear poly(benzimidazopyrrolone imide)s,[4,9–12] have been made; these studies have found that copolymers consisting of pyrrolone and imide segments resulted in, to some extent, an improvement in the processability without the sacrifice of thermal and mechanical properties. However, the poor processability and flexibility of polypyrrolones is still an unanswered problem. Compared with polyimides, polyamides exhibit better flexibility. Thus, it is reasonable to expect that a copolymer consisting of pyrrolone and acylamide segments should exhibit more flexibility and processability than one consisting of pyrrolone and imide segments.

Recently, polypyrrolones were found to have a preponderance when applied as functional materials, such as membranes for gas or fluid separation[13,14] and conducting materials.[15] However, the development of polypyrrolones has been greatly restricted because of the limitation of their infusibility on the basis of commercially available tetraamine monomer, which is the most important component in polypyrrolone synthesis. Therefore, the design and synthesis of new types of tetraamine monomers will play an important role in the modification of traditional polypyrrolones; for example, Koros and Walker[16] and Vora and Menczel[17] synthesized 3,3′,4,4′-tetraamine hexafluoroisophenylene biphenyl by different procedures. Polypyrrolones derived from 3,3′,4,4′-tetraamine hexafluoroisophenylene biphenyl and various dianhydrides exhibited good thermal resistance and processability and high gas permeation and permselectivity.

In other words, the synthesis of a new tetraamine monomer should one focus for the development of polypyrrolones and corresponding materials.

Considering the incorporation of flexible acylamide groups, which could improve the processability and flexibility of polymers[18] and the pyridine ring, which should have contributions to thermal stability, on the basis of its symmetry and aromaticity, and solubility from its polarity,[19–23] we successfully synthesized a novel tetraamine monomer containing an acylamide group [—C(O)—NH—] and a pyridine ring: 2,6-bis[(m,p-diaminophenyl)carboxamido]pyridine (BPTA). In this study, the corresponding poly(benzimidazopyrrolone amide) (PPA) copolymers were prepared by the polycondensation of the resulting tetraamine monomer, BPTA, with commercially aromatic dianhydrides, and tough, flexible PPA films were obtained by the casting of the prepolymer solution on a glass substrate followed by a thermal curing program. The BPTA and related intermediates and the PPA copolymers and films were characterized by means of ^{1}H-NMR, ^{13}C-NMR, and Fourier transform infrared (FTIR; KBr plate) spectroscopy; elemental analysis; differential scanning calorimetry (DSC); thermogravimetric analysis (TGA); and wide-angle X-ray diffraction methods, meanwhile, the mechanical properties and dielectric constants of the resulting PPA films were measured on an Instron 1122 testing instrument and an Agilent 4291B instrument, respectively.

EXPERIMENTAL

Materials

2,6-Pyridinedicarboxyl chloride (TCI), 2-nitro-p-phenylenediamine (TCI), hydrazine monohydrate (80%, Tianjin Chemical Reagents Corp., China), and Pd/C (10%, TCI) were used as received. 4,4′-Oxydiphthalic anhydride (ODPA; Shanghai Nanxiang Chemical Co., China), 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (BTDA; Beijing Chemical Reagents Corp., China), and 4,4′-(hexafluoroisopropylidene)-diphthalic anhydride (6FDA; Aldrich) were recrystallized from acetic anhydride before use. N,N-Dimethylformamide (DMF) and N-methyl-2-pyrrolidone (NMP) were purified by distillation under reduce pressure over calcium hydride and stored over 4-Å molecular sieves. All other solvents were obtained from various commercial sources and were used without further purification.

Measurements

^{1}H-NMR (300 MHz) and ^{13}C-NMR (300 MHz) spectra were measured on a Mercury Plus 400 spectrometer with tetramethylsilane as the internal reference.

FTIR spectra of the monomer and intermediates were obtained on a Micolet NEXUS 670 spectrometer (KBr disks). Elemental analyses were determined with a PerkinElmer model 2400 CHN analyzer. DSC analyses were performed on a PerkinElmer DSC 7 differential scanning calorimeter at a heating rate of 20°C/min under flowing nitrogen. The glass-transition temperature (T_g) was taken as the midpoint of the inflection observed on the curve of heat capacity versus temperature. The melting points were measured by the DSC method. TGA was conducted with a TA Instruments TGA 2050 instrument, and experiments were carried out on approximately 10-mg samples under a controlled flux of air at a heating rate of 20°C/min. Wide-angle X-ray diffraction measurements were performed at 25°C on a Siemens Kristalloflex D5000 X-ray diffractometer, with nickel-filtered Cu Kα radiation (λ = 1.5418 Å), operating at 40 kV and 30 mA. The mechanical properties were measured on an Instron 1122 testing instrument with 100×5 mm^2 specimens in accordance with GB 1040-79 at a drawing rate of 50 mm/min. The dielectric constants were determined on an Agilent 4291B instrument with 25 μm thick specimens at frequencies of 1 and 10 MHz at room temperature. We measured water uptake by weighing the changes of a sample (30×30 mm^2) before and after immersion in water at 25°C for 24 h.
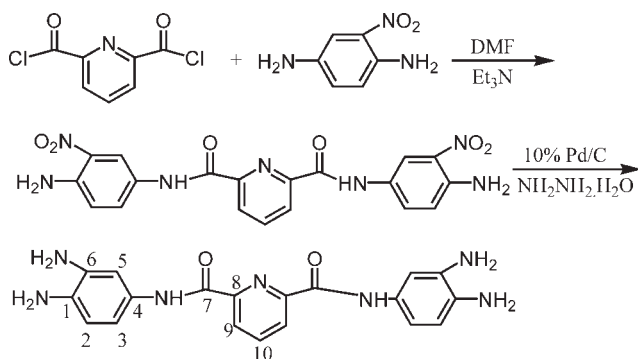
Synthesis of the tetraamine monomer

Synthesis of 2,6-bis[(m-nitro-p-aminophenyl) carboxamido]pyridine (BPNA)

In a 250-mL flask equipped with a mechanical stirrer and a dropping funnel, 24.50 g (0.16 mol) of 2-nitro-p-phenylenediamine and 8.10 g (0.08 mol) of triethylamine were dissolved in 100 mL of dry DMF. 2,6-Pyridinedicarboxyl chloride (16.32 g, 0.08 mol), dissolved in 20 mL of dry DMF, was then added dropwise to the reaction mixture over a period of 2 h. After the reaction was allowed to continue for 6 h, the reaction mixture was poured into 400 mL of water. The precipitate was filtered, dried, and recrystallized from DMF–H$_2$O, and 12.65 g of BPNA was obtained (yield = 36.2%, mp >300°C).

IR (KBr, cm^{-1}): 3366, 3229 (N—H); 1667 (C=O); 1534 (—NO2). ^{1}H-NMR (DMSO-d_6, δ, ppm): 11.04 (s, 2H, —NH—C(O)—), 8.78 (s, 2H), 8.36–8.30 (d, J = 8.4 Hz, 2H), 8.08–8.03 (dd, J = 5.7 Hz, 1H), 7.40–7.37 (d, J = 9.3 Hz, 2H), 7.15–7.11 (d, J = 9.3 Hz, 2H), 4.47 (s, 4H). ^{13}C-NMR (DMSO-d_6, δ, ppm): 160.97, 149.66, 140.25, 138.27, 137.79, 128.67, 128.09, 127.34, 113.44, 110.28.

ANAL. Calcd for $(C_{19}H_{15}N_7O_6)_n$ $(437.37)_n$: C, 52.18%; H, 3.46%; N, 22.42%. Found: C, 52.39%; H, 3.37%; N, 22.34%.

Scheme 1 Synthesis of the tetraamine monomer BPTA.

Synthesis of BPTA

In a 250-mL flask equipped with a dropping funnel and a reflux condenser, 4.37 g (0.01 mol) of BPNA, 0.40 g of Pd/C (10%), and 80 mL of tetrahydrofuran were added. After the mixture was heated to refluxing temperature, 10 mL of hydrazine monohydrate (80%) was added dropwise over a period of 2 h, and the reaction mixture was refluxed for additional 6 h. The catalyst was filtered off, and 100 mL of distilled water was added to the filtrate, which was then held for 72 h. When the precipitate was filtered, 2.95 g of BPTA was obtained (yield = 78.2%, mp = 163°C).

IR (KBr, cm^{-1}): 3365, 3224 (N—H); 1667 (C=O). ^{1}H-NMR (DMSO-d_6, δ, ppm): 10.69 (s, 2H, —NH—C(O)—), 8.33–8.30 (d, J = 8.7 Hz, 2H, H9), 8.25–8.20 (dd, J = 6.3 Hz, 1H, H10), 7.09 (s, 2H, H5), 6.86–6.83 (d, J = 8.1 Hz, 2H, H2), 6.56–6.53 (d, J = 8.1 Hz, 2H, H3), 4.55 (s, 8H, —NH$_2$). ^{13}C-NMR (DMSO-d_6, δ, ppm): 161.56 (C7), 150.13 (C8), 140.06 (C10), 135.68 (C6), 132.85 (C1), 128.93 (C4), 125.18 (C9), 114.71 (C2), 112.00 (C3), 109.50 (C5).

ANAL. Calcd for (C$_{19}$H$_{19}$N$_7$O$_2$)$_n$ (377.40)$_n$: C, 60.47%; H, 5.07%; N, 25.98%. Found: C, 60.52%; H, 4.88%; N, 26.04%.

Synthesis of the PPA copolymers

The PPA copolymers were synthesized via a two-step, low-temperature solution polycondensation procedure, which used the precursor, poly(amide amino acid) (PAAA). The representative polymerization procedure can be illustrated by the preparation of PPA-1, as follows.

In a 50-mL, three-necked flask equipped with a mechanical stirrer, a dropping funnel, a dry nitrogen inlet, and a thermometer, 1.1322 g (3.0 mmol) of BPTA was stirred to dissolve in 15 mL of dry NMP, which was then cooled with an ice–salt bath and maintained at −10°C. After 0.9306 g (3.0 mmol) of ODPA, dissolved in 10 mL of dry NMP, was added dropwise to the mixture with stirring over a period of 1 h, the reaction mixture was stirred for 3 h at

−10°C and for 12 h at room temperature to yield a viscous brown PAAA solution, which was then filtered. The PPA-1 film was obtained by the casting of the filtrate on a plane glass substrate, followed by thermal curing via the following conditions: 80°C for 3 h, 120°C for 1 h, 150°C for 1 h, 200°C for 1 h, 250°C for 1 h, 320°C for 1 h, and 350°C for 1 h.

PPA-2 (BPTA/BTDA) and PPA-3 (BPTA/6FDA) were prepared according to the same procedure.

RESULTS AND DISCUSSION

Composition and structure of the tetraamine monomer BPTA

The tetraamine monomer containing acylamide bonds and the pyridine moiety, BPTA, was synthesized via a two-step synthetic route, as shown in Scheme 1. First, 2,6-pyridinedicarboxyl chloride was reacted with 2-nitro-p-phenylenediamine to offer the dinitro–diamino compound BPNA, as reported by Kumar et al.[24] Then, the tetraamine BPTA was obtained by the reduction of the dinitro–diamino compound BPNA with hydrazine monohydrate catalyzed by Pd/C. The yield of the intermediate dinitro compound BPNA, 36%, was low, which might have been the composite results of the low activity of 2,6-pyridinedicarboxyl chloride; the existence of two active amino groups, although their activities were different; and the good solubility of BPNA in DMF; however, it was higher than that reported by Kumar et al.[24] The new tetraamine monomer BPTA was stable in atmosphere at room temperature, and its purity was good enough for polymerization with commercial aromatic dianhydride monomers to prepare polypyrrolones. The detailed characterizations of the tetraamine monomer BPTA and the dinitro–diamino compound BPNA were done by elemental analyses and FTIR and NMR methods, and the related data presented in the Experimental section unambiguously supported the structure shown in Scheme 1. Figure 1 presents the ^{1}H-NMR and ^{13}C-NMR spectra of tetraamine BPTA, in which all the signals could be ascribed to the protons and carbons of BPTA, respectively. That is, the new tetraamine monomer BPTA was successfully synthesized, and its chemical structure was clearly identified.

Preparation of the PPAs and related reactions

The PPAs were prepared by the thermal cyclization of the PAAAs by polycondensation of the resulting tetraamine monomer BPTA with aromatic dianhydrides in NMP at a low temperature, as shown in Scheme 2. The polycondensation reaction of BPTA with aromatic dianhydrides was strongly affected by the stoichiometry and the addition velocity of the
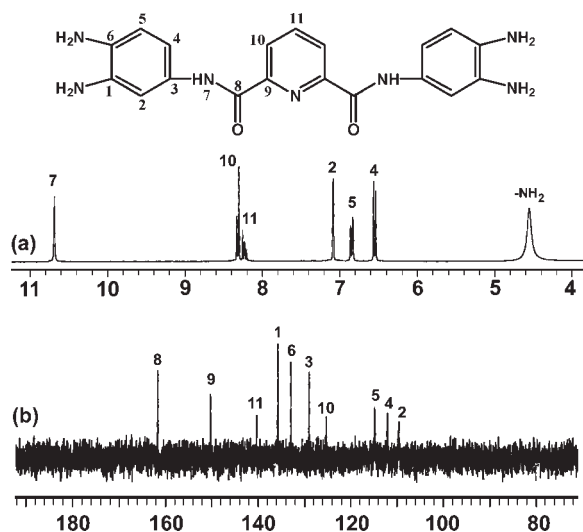
Figure 1 ^{1}H-NMR and ^{13}C-NMR spectra of the tetra-amine BPTA (300 MHz, DMSO-d_6).

monomers. In our cases, aromatic dianhydride monomers were selected to add to the NMP solution containing BPTA, and the addition time was controlled within 1 h. If dianhydride monomers were superfluous or the addition velocity was too fast, the reaction mixture was easily gelled. Therefore, an exact molar ratio of dianhydride and tetraamine very close to 1.0 and a low addition velocity were required to obtain a PAAA solution with a suitable viscosity; then, tough, flexible PPA films could be prepared by the casting of the PAAA solution onto a

glass substrate followed by thermal curing with the temperature program as follows: 80°C for 3 h, 120°C for 1 h, 150°C for 1 h, 200°C for 1 h, 250°C for 1 h, 320°C for 1 h, and 350°C for 1 h.

The inherent viscosities of the resulting PAAAs, which were measured by an Ubbelohde viscometer with 0.5 g/dL of an N,N-dimethylacetamide (DMAc) solution at 30°C, ranged from 0.82 to 0.91 dL/g (Table I). Strong, tough, and flexible PPA films were obtained by the casting of the PAAA solutions onto the glass substrate followed by thermal curing with a final temperature of 350°C. FTIR spectroscopy technology was used to monitor the curing progress, and Figure 2 compares the FTIR spectra of PPA-1's thermally cured at different temperatures. According to the variations of the FTIR spectra presented in Figure 2, the characteristic absorptions of the imide ring (1780 and 1725 cm^{-1}) were observed when the curing temperature was lower than 150°C, which indicated that poly(amino imide) was the major product. When the curing temperature reached 200°C, the absorption band at 1780 cm^{-1} disappeared, and new absorption bands at 1756 and 1600 cm^{-1} appeared, which were assigned to the characteristic absorptions of the stretching vibration of C=O in benzimidazole groups.[25] A gradual change for the absorption band at 1470 cm^{-1}, which should have been the characteristic absorption of C=N in polypyrrolone groups, occurred from 150 to 350°C. The absorption bands at 1756, 1600, and 1470 cm^{-1} did not disappear when the curing temperature reached 350°C, which indicated the formation of the
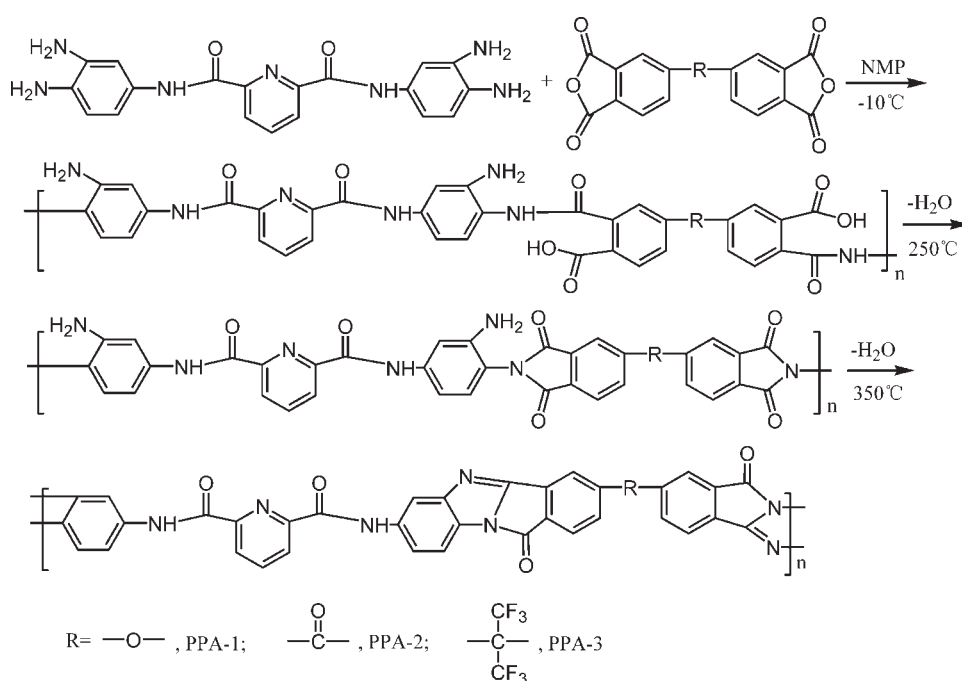
Scheme 2 Synthesis of the PPA copolymers.

TABLE I
Inherent Viscosity (η_{inh}) of the PAAAs and Quality of the PPA films

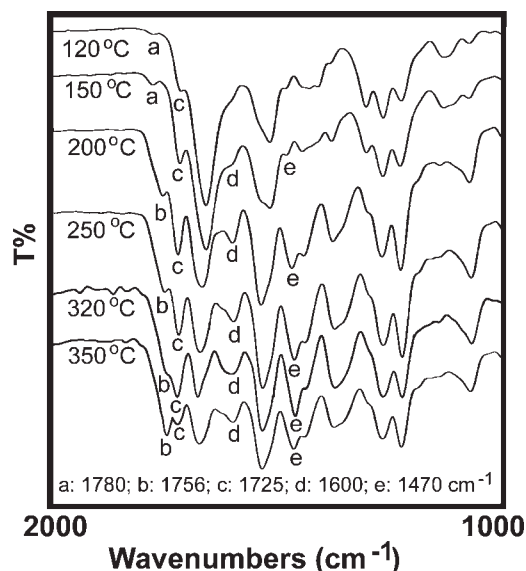| PAAA	η_{inh} (dL/g)[a]	PPAs	Film quality[b]
PAAA-1	0.91	PPA-1	T and F
PAAA-2	0.87	PPA-2	T and F
PAAA-3	0.82	PPA-3	T and F

[a] Measured with a 0.5-g/dL DMAc solution at 30°C
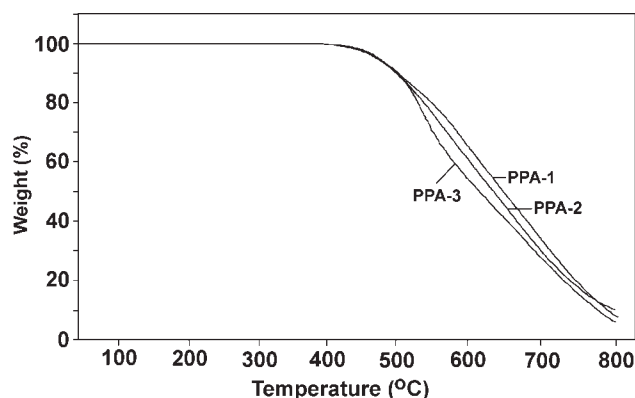[b] F = flexible; T = tough.

polypyrrolones. Obviously, when the curing temperature was low, thermal cyclization based on the carboxyl group with the —NH in the acylamide first occurred, and then, a second thermal cyclization based on C=O in the imide group with the α —NH₂ in the aromatic rings occurred with further increases in the curing temperature. In other words, there were two thermal cyclizations to form the polypyrrolone unit in the PPA chain.

Thermal properties of the PPA films

The thermal properties of the resulting PPA films were investigated by DSC and TGA methods. Figure 3 shows the TGA curves of these copolymers, and the data that express their thermal properties, including T_g, onset decomposition temperature (T_d), temperature at 5% weight loss (T_5), and residual weight retention (R_w) at 700°C in air, are listed in Table II. According to Table II, the T_d values ranged from 502 to 521°C, whereas the T_5 values were all higher than 480°C for the resulting copolymer films; meanwhile, the T_d values of the resulting copolymer films based on different aromatic dianhydrides decreased

Figure 3 TGA curves of the PPA films.

in the following order: ODPA > BTDA > 6FDA. The PPA-1 film based on ODPA exhibited the highest T_d value of 521°C, and PPA-3 based on 6FDA exhibited the lowest T_d value of 502°C. The T_d values of the resulting copolymer films over 500°C indicated a high thermal stability, which was mainly attributed to the rigidity resulting from multiaromatic structures and the rigid symmetrical pyridine rings. Also, R_w at 700°C in air was in the range 29.1–34.8%, and the cause of low R_w was the presence of brittle acylamide bonds in the copolymer backbone, which were easily broken at high temperatures, which led to fast weight loss.

Figure 4 shows the DSC curves of these copolymers, and the T_g values of the resulting PPA films determined by DSC ranged from 299 to 337°C. PPA-2 from the ketone-type dianhydride BTDA exhibited the highest T_g value, and PPA-1 from the ether-type dianhydride ODPA exhibited the lowest one; this order was consistent with the results reported by Liu et al.[7] Although the T_g values of these polymers were lower than those of the polypyrrolones (T_g = 300–382°C) reported by Liu et al.,[7] they were higher than that of commercial polyimides, for example, Ultem 1000 (T_g = 217°C),[26] and pyridine-containing aromatic polyamides (T_g < 230°C).[18] The strong intramolecular interactions, rigidity, and multiaromatic structures derived from the polypyrrolone sections in the copolymer made the T_g values of the PPA copolymers higher than that of polyimides and polyamides, whereas the flexible polyamide sections

Figure 2 FTIR spectra of PPA-1 thermally cured at different temperatures.

a: 1780; b: 1756; c: 1725; d: 1600; e: 1470 cm⁻¹

TABLE II
Thermal Properties of the PPA Films

PPA film	T_g (°C)[a]	T_d (°C)	T_5 (°C)	R_w (%)[b]
PPA-1	299	521	482	34.8
PPA-2	337	510	480	30.5
PPA-3	329	502	482	29.1

[a] As measured by DSC.
[b] At 700°C in air.

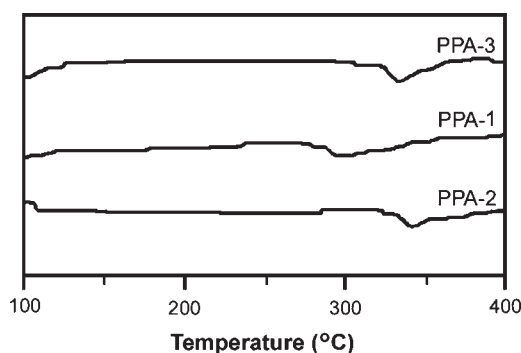**Figure 4** DSC curves of the PPA films.

Figure 5 Wide-angle X-ray diffraction patterns of the PPAs.

caused their T_g values to be lower than that of pure polypyrrolones.

Aggregate structures of the PPA films

The X-ray diffraction patterns of the PPA films are illustrated in Figure 5. According to the wide-angle X-ray diffraction patterns, all of these copolymer films had amorphism aggregate structures with slight crystalline natures; the reason might be that the flexible polyamide sections were suitable to form microcrystalline polypyrrolone sections. Consequently, the resulting PPA films exhibited good thermal properties and were translucent to a certain extent.

Mechanical properties of the PPA films

As described previously, high-quality PPA films were prepared by the casting of PAAA solutions onto glass plates followed by thermal curing according to the temperature program as follows: 80°C for 3 h, 120°C for 1 h, 150°C for 1 h, 200°C for 1 h, 250°C for 1 h, 320°C for 1 h, and 350°C for 1 h. The tensile strength and modulus values of the resulting PPA films are summarized in Table III. Table III shows that the PPA films had tensile strengths of 102.1–115.9 MPa, tensile moduli of 1.46–1.92 GPa, and elongations at break of 6.8–7.4%, which indicated that the PPA films were strong and tough poly-
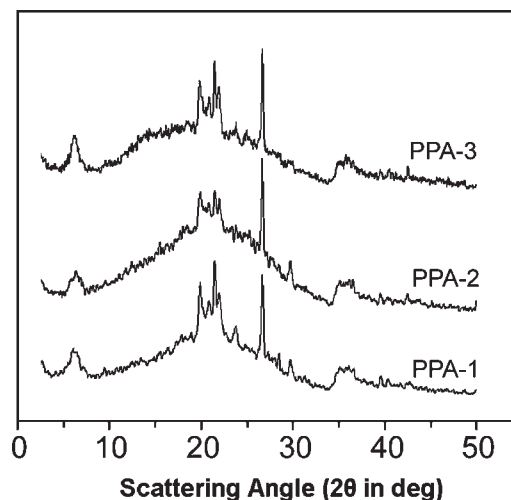
meric materials. The tensile modulus of PPA-3, derived from BPTA and fluorinated aromatic dianhydride 6FDA, was almost 0.5 GPa lower than those of PPA-1 and PPA-2. Obviously, the high fluorine concentration in the copolymer backbone was responsible for this reduction in tensile modulus.

Dielectric properties of the PPA films

The dielectric properties of these pyridine-containing PPA films were detected at frequencies of 1 and 10 MHz at 25°C, as shown in Table III. The dielectric constants of these PPA films at 1 and 10 MHz were in the range 3.13–3.34 and 3.03–3.22, respectively, and the dielectric dissipation factors were in the range $7.4–8.5 \times 10^{-3}$ and $7.9–8.4 \times 10^{-3}$, respectively. For each copolymer, the dielectric constant at 1 MHz was higher than that at 10 MHz. This variation was attributed to the frequency dependence of the polarization mechanisms, which comprised the dielectric constant. Among the three PPA films, PPA-3 exhibited the lowest dielectric constant, which was explained by the presence of CF_3 groups in the polymer backbone.

TABLE III
Mechanical and Dielectric Properties of the PPA Films

PPA film[a]	Mechanical property[b]			Dielectric property				Water uptake (%)
	TS (MPa)	TM (GPa)	E (%)	Dielectric constant at 1 MHz	Dielectric dissipation factor at 1 MHz	Dielectric constant at 10 MHz	Dielectric dissipation factor at 10 MHz	
PPA-1	107.4	1.88	7.4	3.29	8.5×10^{-3}	3.22	7.9×10^{-3}	0.85
PPA-2	115.9	1.92	6.8	3.34	7.4×10^{-3}	3.20	8.1×10^{-3}	0.86
PPA-3	102.1	1.46	7.0	3.13	7.9×10^{-3}	3.03	8.4×10^{-3}	0.57

[a] The thickness of the measured specimens was 25 μm.
[b] E = elongation at break; TM = tensile modulus; TS = tensile strength.

Hydrophobic properties of the PPA films

The water uptake of these PPAs was investigated (see Table III). The results indicate that these PPAs exhibited a low water uptake of 0.57–0.86%, which presented the negative influence of the moisture from the surrounding environment on the dielectric properties. PPA-3 exhibited the lowest water uptake among the PPAs, and the reason was the introduction of the hydrophobic trifluoromethyl groups in the polymer backbone. Compared with polyamides, the resulting copolymers showed low water uptake. The reason was the contribution of the hydrophobic multiaromatic structures in the repeat units of the copolymers.

CONCLUSIONS

A series of new PPA copolymers were synthesized with a novel pyridine-containing tetraamine and various aromatic dianhydrides by two-step methods. The precursors, PAAAs with inherent viscosities in the range 0.82–0.91 dL/g, were first obtained by a low-temperature solution polycondensation procedure; then, tough and flexible PPA films were prepared by the casting of the PAAA solutions onto glass substrates followed by thermal curing via a programmed temperature procedure up to 350°C. The PPA films exhibited excellent thermal properties with T_d's in the range 502–521°C, T_g's in the range 299–337°C, and R_w's at 700°C in air of 29.1–34.8% and also exhibited good mechanical properties with tensile strengths of 102.1–115.9 MPa and elongations at break of 6.8–7.4%. The dielectric constants of the copolymer films were found in the range 3.13–3.34 and 3.03–3.22 at 1 and 10 MHz, respectively.

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