Synthesis of Aromatic Poly(urea-imide) with Heterocyclic Side-Chain Structure

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ABSTRACT: The poly(urea-imide) copolymers with inherent viscosity of 0.81–1.08 dL/g were synthesized by reacting aryl ether diamine or its polyurea prepolymer with various diisocyanate-terminated polyimide prepolymers. The aryl ether diamine was obtained by first nucleophilic substitution of phenolphthalein with *p*-chloronitrobenzene in the presence of anhydrous potassium carbonate to form a dinitro aryl ether, and then further hydrogenated to diamine. The polyimide prepolymers were prepared by using 4,4′-diphenylmethane diisocyanate to react with pyromellitic dianhydride, 3,3′,4,4′-benzophenonetetracarboxylic dianhydride, or 3,3′,4,4′-sulfonyldiphthalic anhydride by using the direct one-pot method to improve their solubility, but without sacrificing thermal property. These copolymers are amorphous and readily soluble in a wide range of organic solvents such as *N*-methyl-2-pyrrolidone, dimethylimidazole, *N,N*-dimethylacetamide, dimethyl sulfoxide, *N,N*-dimethylformamide, *m*-cresol, and sulfolane. All the poly(urea-imides) have glass transition temperatures in the range of 205–240°C and show a 10 wt % loss at 326–352°C in nitrogen and 324–350°C in air. The tensile strength, elongation at break, and initial modulus of these copolymer films range from 42 to 79 MPa, 5 to 16%, and 1.23 to 2.02 GPa, respectively. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 1719–1730, 1999

Key words: copoly(urea-imide); heterocyclic containing; thermostable polymer

INTRODUCTION

Aromatic polyimides are known as one of the best materials for use in high-temperature applications.[1–3] Its imperviousness to heat temperature is due to its aromatic and heterocyclic structure. Polyimides can resist 500°C for a few minutes or 300°C for several months. The rigid structure in the polyimide chain imparts good properties such as thermooxidative stability,[4–6] but makes the material hard to process owing to its poor solubility.[7–10] In our previous studies,[11–12] we have employed the acid-terminated poly(*m*-phenylene isophthalamide) (PmIA) prepolymer to modify polyimides. It improved the solubility, mechanical properties, and processing characteristics of the copolymers. This article addresses the use of aryl ether diamine or its polyurea prepolymer to modify the polyimide, and the effect of heterocyclic side-chain structure on the properties of copoly(urea-imides) was studied.
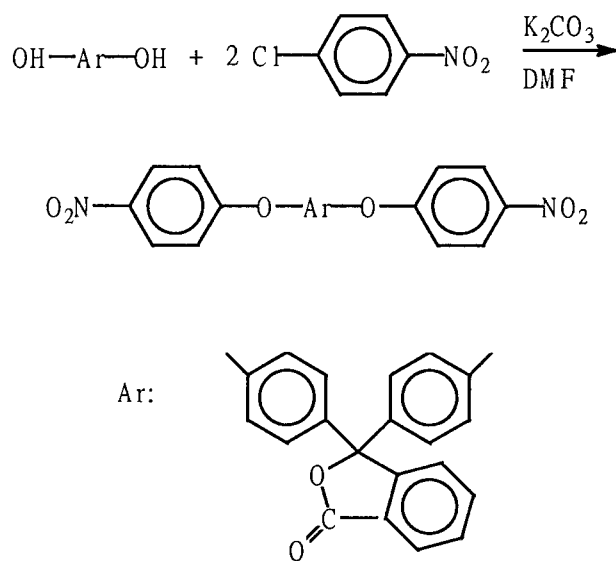
EXPERIMENTAL

Materials

Commercially available dianhydrides (Tokyo Chemical Co.), that is, pyromellitic dianhydride (PMDA), 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (BTDA), and 3,3′,4,4′-sulfonyl diphthalic anhydride (DSDA), were dried under vac-

Scheme 1

Polymer :

PI-P:PMDA-MDI=1-1
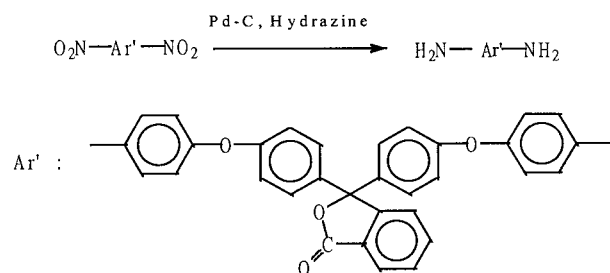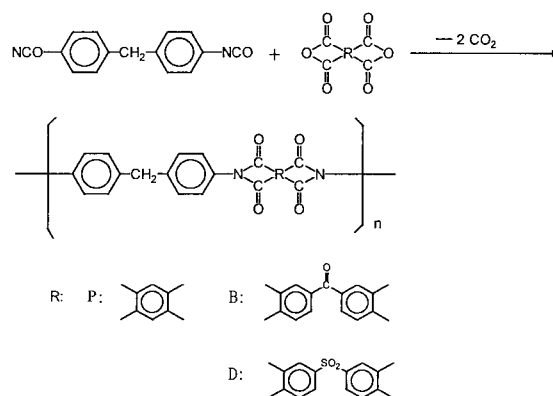PI-B:BTDA-MDI=1-1
PI-D:DSDA-MDI=1-1

Scheme 3

uum at 40°C for 4 h. 4,4-Diphenylmethane diisocyanate (MDI) was used without purification. Phenolphthalein and *p*-chloronitrobenzene were of high purity and used without purification. Solvents such as *N*-methyl-2-pyrrolidone (NMP), *N,N*-dimethylacetamide (DMAc), *N,N*-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), 1,3-dimethyl-2-imidazolidone (DMI), *m*-cresol, and sulfolane (cyclo-butylsulfone) (purchased from Merck Co. and from Aldrich Chemical Co.) were purified by vacuum distillation on calcium hydride. Triethylamine was used as a catalyst, without further purification.

Synthesis of Dinitro Monomer[13–14]

A mixture of 0.15 mol of phenolphthalein, 0.35 mol of *p*-chloronitrobenzene, and 0.40 mol of anhydrous potassium carbonate in 160 mL of dry DMF was refluxed at 160°C for 8 h and cooled. The mixture was then poured into 1 : 1 ethanol : water. The precipitate was collected by filtration
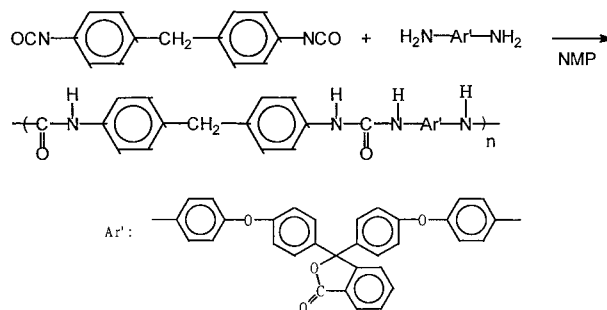
and recrystallized from DMF (yield: 87%). The reaction is shown in Scheme 1.

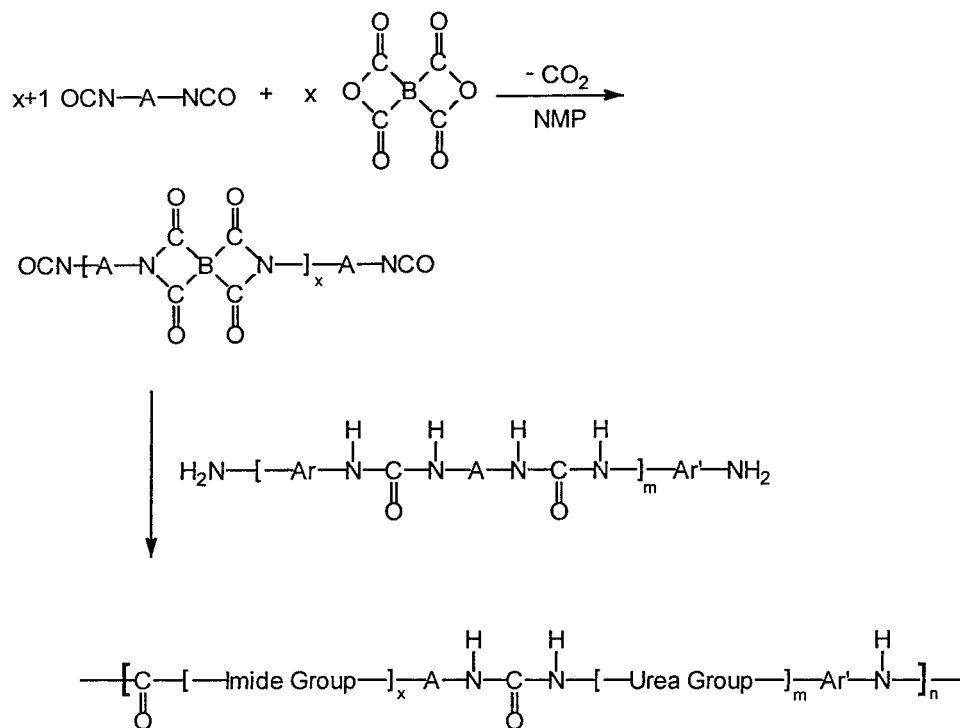### Synthesis of Diamino Monomer[14–16]

The 0.09 mol of dinitro compound and 0.31 g of Pd-C was dissolved in 180 mL of ethanol and poured into a four-necked flask. The 90 mL of hydrazine monohydrate was added dropwise over a period of 1 h at 85°C. Then, the reaction was continued to reflux for another 6 h. After cooling, the mixture was filtered to remove Pd-C, and the solution was concentrated under reduced pressure and poured into water. The precipitate was collected by filtration and dried to yield a white solid (yield: 81%). The reaction is shown in Scheme 2.
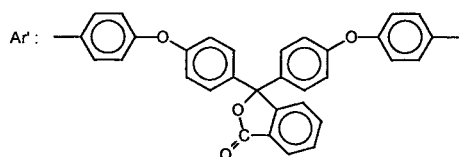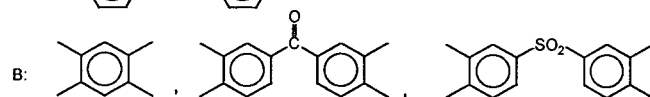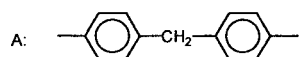
Synthesis of Aromatic Polyimide

Equimolar MDI and dianhydride were dissolved in NMP (solid content of 15%) into the reaction

Scheme 2

Scheme 4

x+1 OCN—A—NCO + x [structure] $\xrightarrow[\text{NMP}]{-\text{CO}_2}$

OCN—[A—N—B′—N—]$_x$—A—NCO

H$_2$N—[—Ar—N—C—N—A—N—C—N—]$_m$—Ar′—NH$_2$

—[C—[—Imide Group—]$_x$—A—N—C—N—[—Urea Group—]$_m$—Ar′—N—]$_n$—

Where x=1,2,3,4 m=0,1,2

A: [structure] —CH$_2$—

B: [structures] , [structure with C=O] , [structure with SO$_2$]

Ar′ : [structure]

PUI- 1 :	MDI-PMDA-Amine-MDI=2-1-1-0	PUI- 8 :	MDI-BTDA -Amine-MDI=5-4-1-0
PUI- 2 :	MDI-PMDA-Amine-MDI=3-2-1-0	PUI-15:	MDI-BTDA -Amine-MDI=5-4-2-1
PUI- 3 :	MDI-PMDA-Amine-MDI=4-3-1-0	PUI-16:	MDI-BTDA -Amine-MDI=5-4-3-2
PUI- 4 :	MDI-PMDA-Amine-MDI=5-4-1-0	PUI- 9 :	MDI-DSDA -Amine-MDI=2-1-1-0
PUI-13:	MDI-PMDA-Amine-MDI=5-4-2-1	PUI-10:	MDI-DSDA -Amine-MDI=3-2-1-0
PUI-14:	MDI-PMDA-Amine-MDI=5-4-3-2	PUI-11:	MDI-DSDA -Amine-MDI=4-3-1-0
PUI- 5 :	MDI-BTDA -Amine-MDI=2-1-1-0	PUI-12:	MDI-DSDA -Amine-MDI=5-4-1-0
PUI- 6 :	MDI-BTDA -Amine-MDI=3-2-1-0	PUI-17:	MDI-DSDA -Amine-MDI=5-4-2-1
PUI- 7 :	MDI-PMDA-Amine-MDI=4-3-1-0	PUI-18:	MDI-DSDA -Amine-MDI=5-4-3-2

Scheme 5

flask of 250 mL and heated at 60°C for 1.5 h. The triethylamine catalyst (0.03 g/L) was then added and the temperature was raised to 80°C. The viscosity was found to increase gradually with the evolution of carbon dioxide. The viscosity was adjusted by adding the solvent. The reaction was

Table I   Elemental Analysis of Ether Amine Monomer

	Elemental Analysis (%)		
	C%	H%	N%
Calcd.	76.79	4.83	5.60
Found	76.71	4.77	5.88

completed after 1 h. A golden viscous solution of polyimide was obtained. The reaction is shown in Scheme 3.

Synthesis of Aromatic Polyurea

Equimolar MDI and diamine monomer were dissolved in NMP (solid content of 15%) into the reaction flask of 250 mL and reacted at room temperature for 1 h. The temperature was then raised to 60°C for another 2 h. A golden viscous solution of polyurea was obtained (see Scheme 4).

Synthesis of Poly(urea-imide) with Heterocyclic Side-Chain Structure

The copoly(urea-imide) with heterocyclic side-chain structure was synthesized by reacting isocyanate-terminated polyimide prepolymer with amine-terminated polyurea prepolymer with heterocyclic side-chain structure at room temperature for 1 h. Then the temperature was raised to 60°C and kept at that temperature for another 2 h. When copolymer was obtained, it was cast into a film by solvent evaporation as shown in the reaction Scheme 5.

Measurements

Infrared (IR) spectra of polyimides, polyurea, and copoly(urea-imides) with heterocyclic side-chain structure were obtained by using a Hitachi Model 260-50 spectrometer over a range of 250–4000 cm^{-1}. The samples were in the form of a film about 10-μm thick. Elemental analysis (C, H, N) was performed by using a Perkin–Elmer 2400 analyzer. Also, the ^{1}H-NMR spectrum was obtained by using the VXR-300/51 NMR spectrometer.

Differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) were performed by using a DuPont 9900 differential scanning calorimeter and a Seiko SSC-5000-type thermogravimetric analyzer at a heating rate of 20°C/min under nitrogen atmosphere to obtain glass

transition temperature (T_g) and weight loss, respectively.

Wide-angle X-ray photographs were taken with Ni-filtered CuKα radiation by using a Rigaku D/max-II-type X-ray diffractometer. All stress–strain data were obtained from an Instron 1122-type testing instrument at an extension rate of 10 mm/min by using a specimen 50-mm long, 10-mm wide, and 0.5-mm thick.

RESULTS AND DISCUSSION

Synthesis of Aryl Ether Diamine and Its Polyurea Prepolymer with Heterocyclic Side-Chain Structure

Aryl ether diamine and its polyurea prepolymer with heterocyclic structure was prepared from phenolphthalein, p-chloronitrobenzene, and MDI. The molecular weight of the aryl ether diamine was 500 (mp, 96–98°C). The elemental analysis shows that the found values are in good agreement with calculated values. The results of elemental analysis are shown in Table I.

The IR spectrum of diamine (Fig. 1) exhibits the —NH stretching vibrations at 3368 cm^{-1}, C—O—C absorption at 1238 cm^{-1}, and C=O stretching at 1760 cm^{-1}. Figure 2 shows the ^{1}H-NMR spectrum of the diamine monomer; all the expected absorptions are observed. The δ values are 6.74 ppm (d, 4H, Ar—NH$_2$), 6.60 ppm (d, 4H, Ar—NH$_2$), 6.85 ppm (d, 4H, Ar—O), 7.20 ppm (d, 4H, Ar—O), 7.62–7.92 ppm (m, 4H, side chain of benzene ring), and 5.00 ppm (s, 4H, amine). These values support the formation of diamine with heterocyclic side-chain structure.

Synthesis of Polyimide, Polyurea, and Copoly(urea-imide) with Heterocyclic Side-Chain Structure

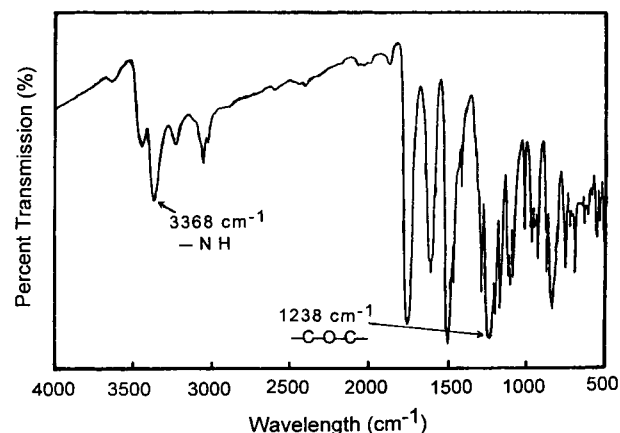MDI and three aromatic dianhydrides (BTDA, PMDA, and DSDA) were used to prepare three

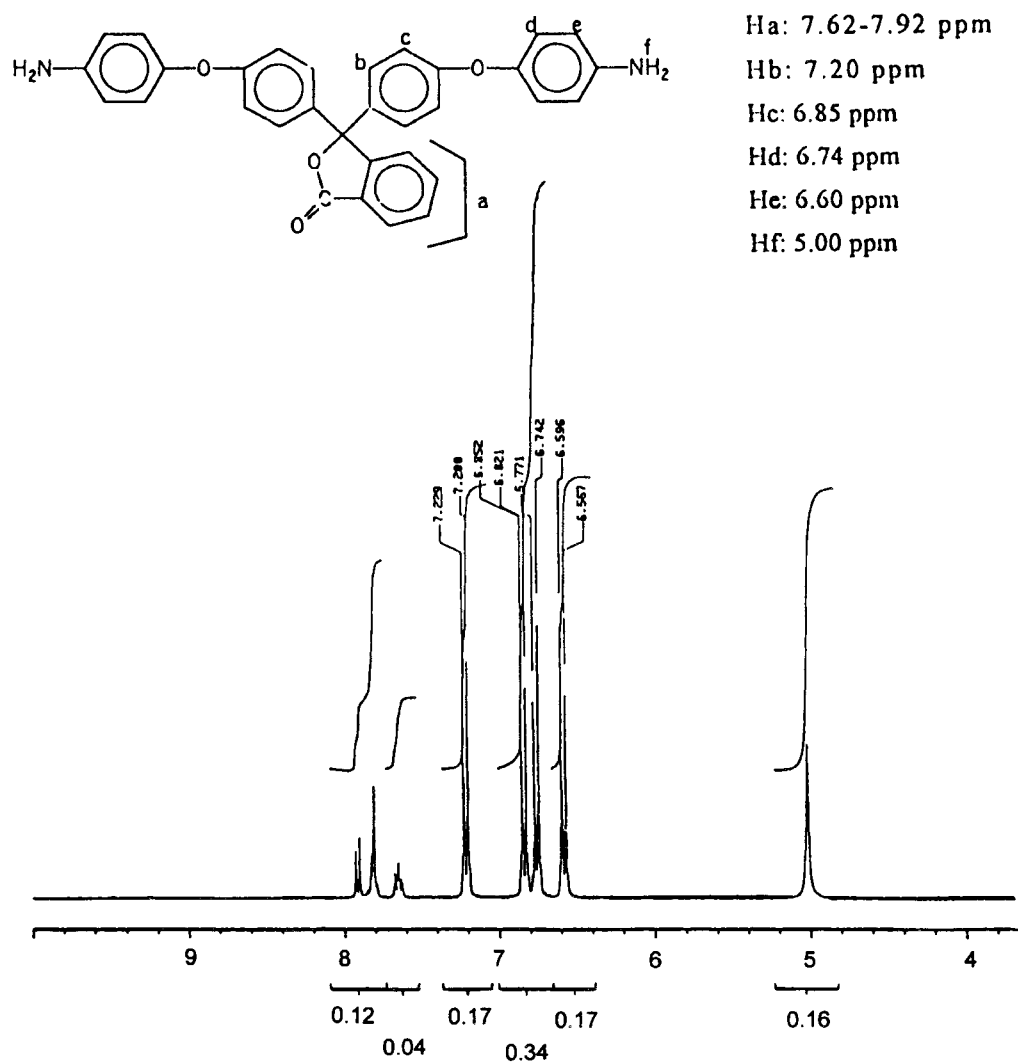**Figure 1**   The IR spectrum of diamine monomer.

Ha: 7.62-7.92 ppm
Hb: 7.20 ppm
Hc: 6.85 ppm
Hd: 6.74 ppm
He: 6.60 ppm
Hf: 5.00 ppm

Figure 2 The ^{1}H-NMR spectrum of diamine monomer.

polyimides. The polyurea containing heterocyclic side-chain structure was synthesized from MDI and aryl ether diamine.

Eighteen poly(urea-imide) copolymers with heterocyclic side-chain structure were synthesized from four different molar ratios of MDI, three dianhydride, and aryl ether diamine (or amine-terminated polyurea prepolymer). The inherent viscosity of polyimide, polyurea, and poly(urea-imide) copolymers are listed in Table II. It is found that the intrinsic viscosities of polymers are > 0.80 dL/g. Especially, some copolymers have a value of > 1.00. The content of urea and imide group in the copolymer is given in Table II.

All of these polymers can be cast into transparent, tough, and flexible films. Figure 3 shows the infrared spectra of polyimide and polyurea; it is found that the characteristic absorption at 1720, 1780, 1360, and 720 cm^{-1} indicate an existence of an imide group. The absorption peaks at 1670–1630, 3500–3350, and 1260–1200 cm^{-1} indicate the presence of urea and aryl ether group, respectively. Figures 3–6 show the infrared spectra of poly(urea-imide) copolymers. The characteristic absorption at 1720, 1780, 1360, and 720 cm^{-1} indicate the presence of an imide group; the absorption peaks at 1670–1630 and 3500–3350 cm^{-1} indicate the presence of a urea group, and the absorption peak at 1260–1200 cm^{-1} indicates the vibration of an ether group. Hence it can be concluded that the polyurea with heterocyclic side-chain structure was introduced into the main chain of polyimide.

Table II The Inherent Viscosity of Polymers

Code	η_{inh} (dL/g)	Imide Content (%)	Urea Content (%)
PU	1.09	0	100
PIP	0.71	100	0
PUI-1	0.80	33.6	66.4
PUI-2	0.81	50.3	49.7
PUI-3	0.86	60.3	39.7
PUI-4	0.91	67.0	33.0
PUI-13	0.97	50.3	49.7
PUI-14	1.08	40.3	59.7
PIB	0.89	100	0
PUI-5	0.81	39.2	60.8
PUI-6	0.88	56.3	43.7
PUI-7	1.01	65.9	34.1
PUI-8	0.89	72.1	27.9
PUI-15	0.84	56.3	43.7
PUI-16	1.02	46.2	53.8
PID	0.85	100	0
PUI-9	0.83	40.9	59.1
PUI-10	0.89	58.1	41.9
PUI-11	0.87	67.5	32.5
PUI-12	0.93	73.5	26.5
PUI-17	0.91	58.1	41.9
PUI-18	0.98	48.0	52.0

Figure 4 The infrared spectra of PMDA series poly(urea-imides).

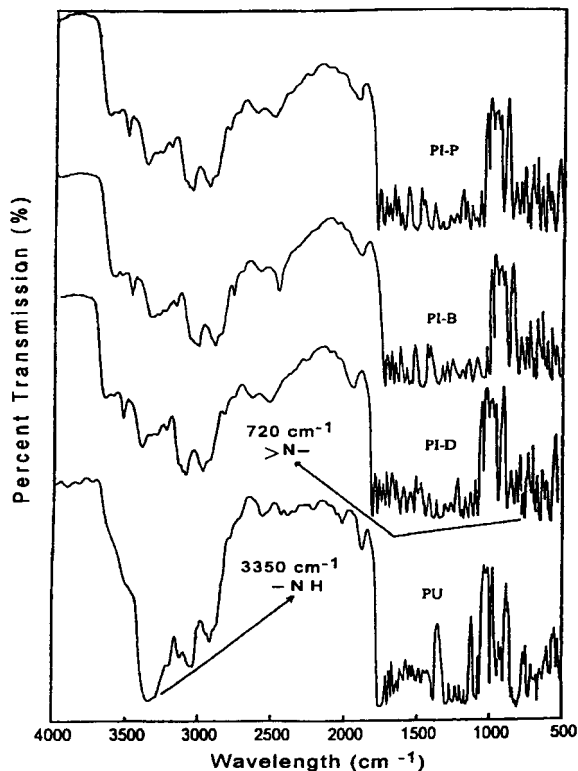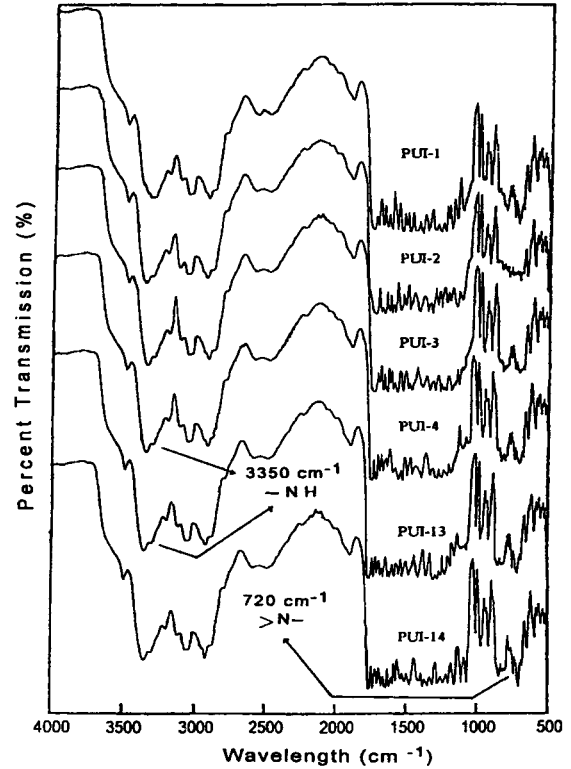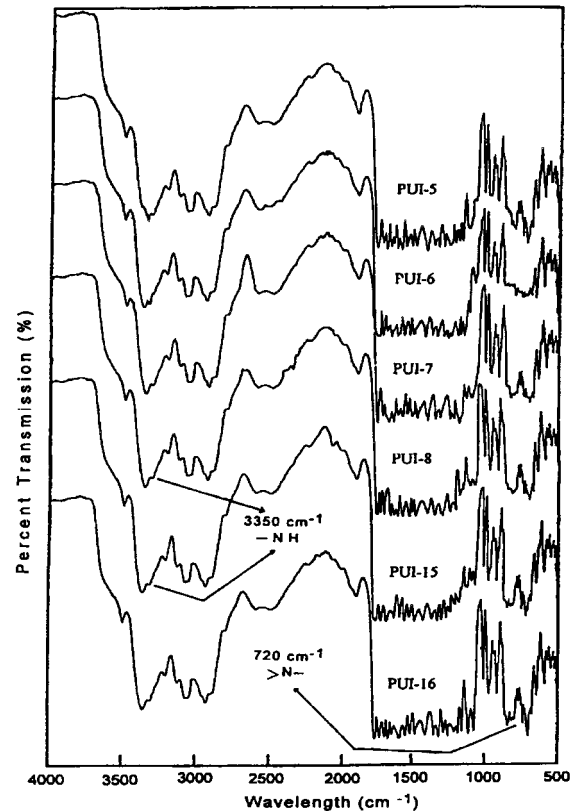**Figure 3** The infrared spectra of polyimides.

Figure 5 The infrared spectra of BTDA series poly(urea-imides).

Table III   Solubility of Polymers

Code	NMP	DMI	DMSO	DMF	DMAC	*m*-Cresol	Sulfolane
				Solvent[b]			
			Solubility of Polymer[a]				
PU	++	++	++	++	++	++	++
PIP	++	X	X	X	X	+	+
PUI-1	++	+	++	++	++	++	+
PUI-2	++	+	++	++	+	++	+
PUI-3	++	+	++	++	+	++	+
PUI-4	++	+	++	++	+	++	+
PUI-13	++	+	++	++	+	++	+
PUI-14	++	+	++	++	++	++	+
PIB	++	+	X	X	X	+	+
PUI-5	++	++	++	++	++	++	+
PUI-6	++	++	++	++	++	++	+
PUI-7	++	++	+	+	++	++	+
PUI-8	++	++	+	+	++	++	+
PUI-15	++	++	+	+	++	++	+
PUI-16	++	++	++	++	++	++	+
PID	++	+	+	X	X	+	++
PUI-9	++	++	++	++	++	++	++
PUI-10	++	++	++	++	++	++	++
PUI-11	++	++	++	++	++	++	++
PUI-12	++	++	++	+	+	++	++
PUI-17	++	++	++	++	++	++	++
PUI-18	++	++	++	++	++	++	++

[a] ++, soluble in room temperature; +, soluble at 80°C; +−, partially soluble at 80°C; −, completely insoluble at 80°C.
[b] NMP: *N*-methyl-2-pyrrolidone; Py: pyridine; DMF: *N,N*-dimethylformamide; Me: methanol; DMAc: *N,N*-dimethylacetamide; DMI: 1,3-dimethyl-2-imidazolidone; DMSO: dimethyl sulfoxide.

Solubility Properties of Polymers

Table III indicates the solubility of all polymers. As indicated, the solubility of copolymers is in between that of polyimide and polyurea. In general, all polymers are soluble in NMP and concentrated sulfuric acid at room temperature. However, the copolymer with polyurea and heterocyclic structure are soluble in a wide range of organic solvents such as NMP, dimethylimidazole, DMAc, DMSO, DMF, *m*-cresol, and sulfolane. The solubility is related to their chemical structure. Those copolymers with sulfonyl and high urea content display good solubility. Thus, the solubility of copolymer from PMDA is inferior to that of BTDA, but the DSDA copolymer with sulfonyl structure shows good solubility. From that, it is manifested that the addition of the sulfone group and heterocyclic polyurea in the polyimide chain does increase their solubility.

Wide-Angle X-Ray Diffraction of Polymers

The WAXD patterns of polyimides, polyureas, and copolymers were measured by the Rigaku diffractometer and are typically shown in Figures 7 and 8. As seen in the figures, it is found that all the polymers are amorphous in nature. This may be caused by the presence of heterocyclic side-chain structure in polymers that decrease their structural regularity.

Thermal Properties of Polymers

The thermal properties of all polymers were evaluated by DSC and TGA. The thermal behavior data of all polymers are listed in Table IV. With the presence of aromatic and heterocyclic ring structures of polyimide, they exhibit an excellent thermooxidative stability and also show a high T_g. The T_g's of three polyimides PI-P, PI-B, and PI-D range from 314 to 351°C. Among PI-P, PI-B, and PI-D, the polyimide PI-D has the lowest T_g because of its sulfonyl structure. However, the T_g of copolymers lies between 205 and 240°C. The lowering of T_g is believed to be due to the urea and ether structure in the polyimide. The relationship between the T_g and the imide content of alternating copolymer is shown in Figure 9. It is found
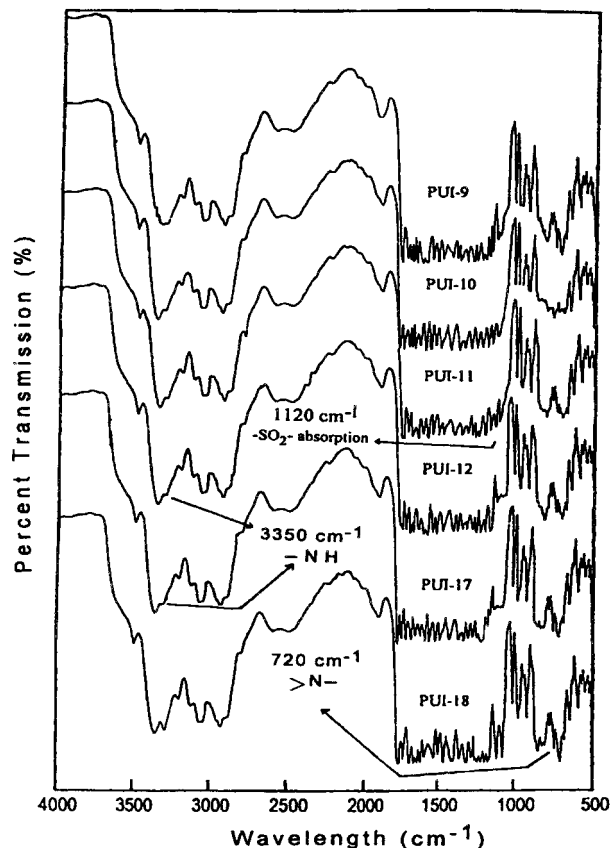
Figure 6 The infrared spectra of DSDA series poly(urea-imides).

Figure 7 X-ray patterns of polyimides.

Table IV   The Thermal Properties of Polymers

Code	T_g (°C)	Imide Content (%)	Urea Content (%)
PU	204	0	100
PI-P	351	100	0
PUI-1	228	33.6	66.4
PUI-2	238	50.3	49.7
PUI-3	236	60.3	39.7
PUI-4	240	67.0	33.0
PUI-13	224	50.3	49.7
PUI-14	205	40.3	59.7
PI-B	314	100	0
PUI-5	209	39.2	60.8
PUI-6	223	56.3	43.7
PUI-7	237	65.9	34.1
PUI-8	239	72.1	27.9
PUI-15	216	56.3	43.7
PUI-16	213	46.2	53.8
PI-D	319	100	0
PUI-9	223	40.9	59.1
PUI-10	226	58.1	41.9
PUI-11	230	67.5	32.5
PUI-12	235	73.5	26.5
PUI-17	212	58.1	41.9
PUI-18	206	48.0	52.0

that the T_g of copolymer decreases with the increasing amount of polyurea.

Table V and Figures 10–13 summarize the thermogravimetric data of all polymers. As indicated, the polyimides containing rigid aromatic ring and heterocyclic ring structure have excellent thermal stability and thermooxidative properties, with no significant weight loss up to 510°C in nitrogen and air and also 10 wt % loss at 540 and 530°C (Figs. 10 and 12), respectively. However, the polyurea and poly(urea-imide) copolymer shows two-stage degradation, and the 10 wt % loss of these copolymers at 324–350°C in air and at 326–352°C in nitrogen (Figs. 11 and 13). With the increase of polyurea content, the 10 wt % loss temperature is much lower. This suggests that the urea linkage does deteriorate the thermooxidative stability.

Mechanical Properties of Polymers

The mechanical properties of polymers are shown in Table VI. Most of the polymers have high strength and modulus, but low elongation. This indicates that the polymers are medium-tough
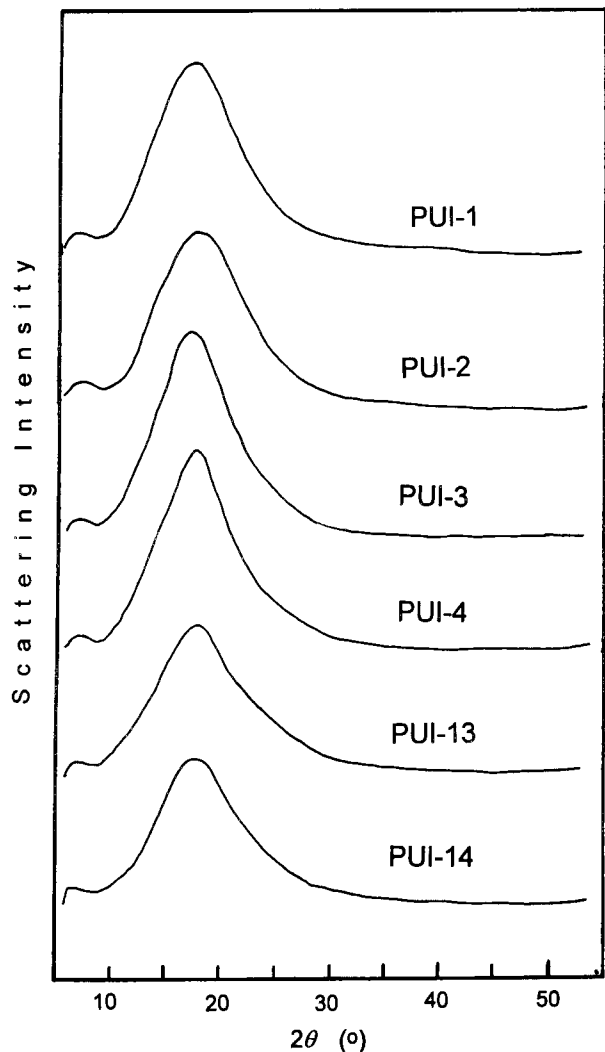
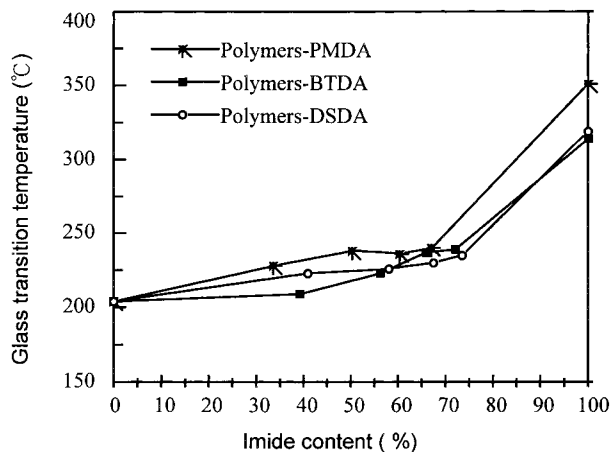**Figure 8** X-ray patterns of PMDA series poly(urea-imides).

Figure 9 The relationship between T_g's and imide content (alternating copolymer).
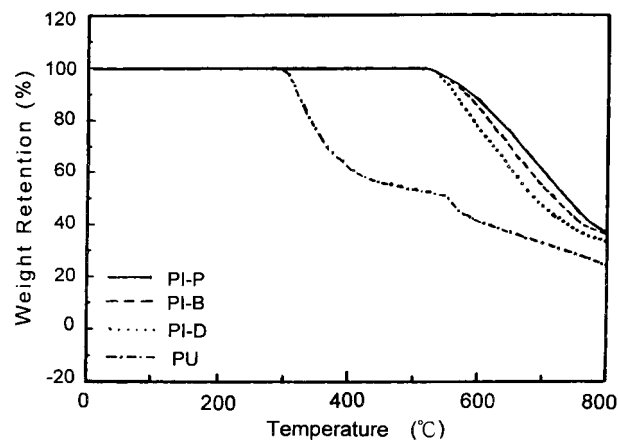

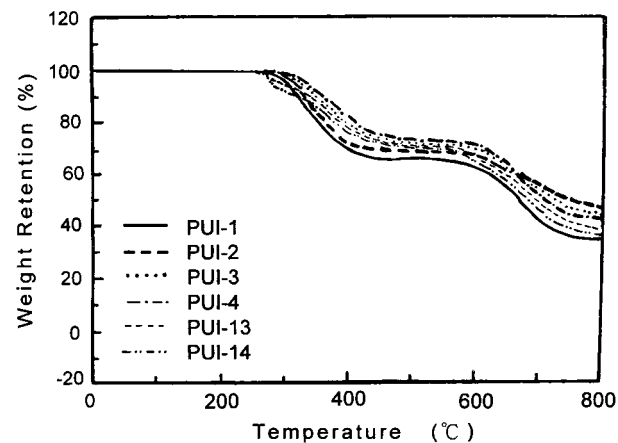**Figure 10** The weight loss of polyimides and polyureas in nitrogen.

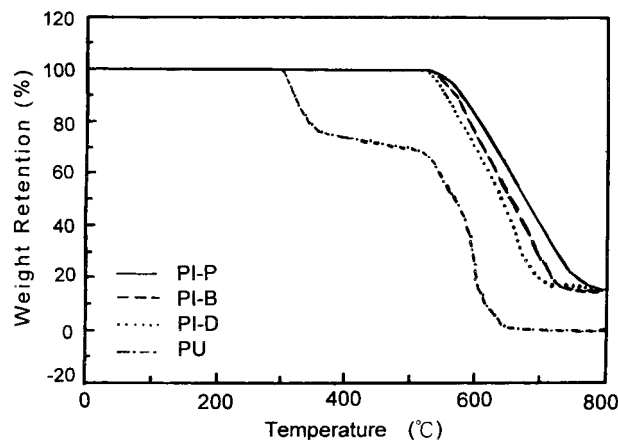**Figure 11** The weight loss of PMDA series poly(urea-imides) in nitrogen.

Figure 12 The weight loss of BTDA series poly(urea-imides) in air.

Table V   The Degradation Temperature of Polymers

Code	Thermal Degradation Temperature (°C) T_{d10}[a]		Thermal Degradation Temperature (°C) T_{d50}[b]		Imide Content [%]	Urea Content [%]
	[Air]	[N$_2$]	[Air]	[N$_2$]		
PU	323	325	567	549	0	100
PI-P	575	583	671	736	100	0
PUI-1	327	326	604	675	33.6	66.4
PUI-2	333	336	608	719	50.3	49.7
PUI-3	339	344	656	712	60.3	39.7
PUI-4	350	352	663	689	67.0	33.0
PUI-13	329	331	616	674	50.3	49.7
PUI-14	327	325	612	662	40.3	59.7
PI-B	570	577	658	718	100	0
PUI-5	325	337	602	610	39.2	60.8
PUI-6	332	341	608	640	56.3	43.7
PUI-7	337	344	610	690	65.9	34.1
PUI-8	345	340	645	715	72.1	27.9
PUI-15	335	334	620	624	56.3	43.7
PUI-16	326	330	612	655	46.2	53.8
PI-D	552	560	634	692	100	0
PUI-9	325	334	590	621	40.9	59.1
PUI-10	330	333	604	633	58.1	41.9
PUI-11	337	341	609	644	67.5	32.5
PUI-12	339	341	615	691	73.5	26.5
PUI-17	330	338	612	640	58.1	41.9
PUI-18	324	329	609	608	48.0	52.0

[a] 10 wt % loss temperature at a heating rate of 20°C/min with TGA measurement.
[b] 50 wt % loss temperature at heating rate of 20°C/min with TGA measurement.

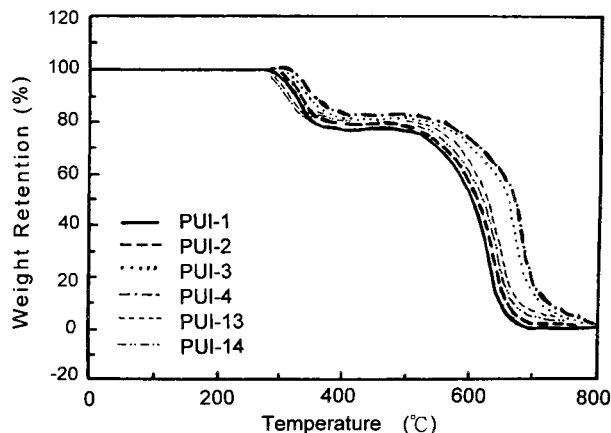materials. Figures 14–15 typically show the relationship of stress and strain in polyimide, polyurea, and PMDA series of copolymer films. As seen in those figures, the tensile strength of the poly(urea-imide) copolymers increase with the increase of imide content, but up to some extent, it be-
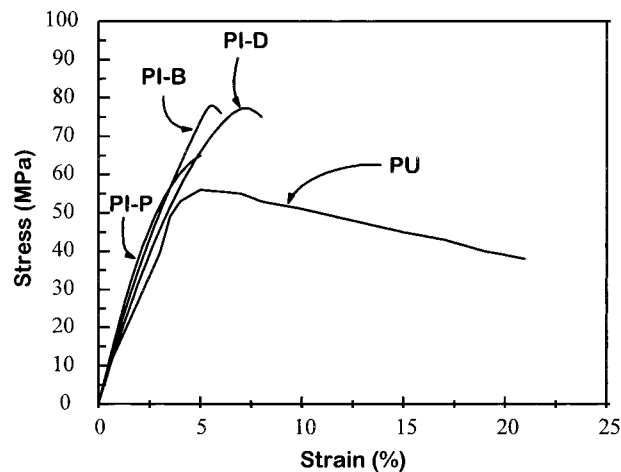

Figure 13 The weight loss of DSDA series poly(urea-imides) in air.

Figure 14 The stress–strain relationship of polyimides and polyureas.

comes even higher than that of polyimide, regardless of the presence of heterocyclic bulky structure. This could be due to the higher molecular weight of copolymer, and also the formation of hydrogen bonding of polyurea structure in copolymer.

Moreover, as shown in Table VI, the breaking elongation of copolymers are increased, but the initial modulus of copolymers is decreased with the increase of urea content due to their heterocyclic bulky structure in the polyurea part of copolymer. However, the formation of hydrogen bonding is considered to be a complex factor. In Table VI, the copolymer of PMDA series exhibits the lowest initial modulus among these three series. It is attributed to its lack of polar carbonyl group or sulfonyl group as in BTDA or in DSDA series.

CONCLUSIONS

During this work, we used a one-step method to synthesize three polyimides, one polyurea, and 18 poly(urea-imide) copolymers. The copoly(urea-imides) showed increase in their molecular

Table VI   The Mechanical Properties of Polymers

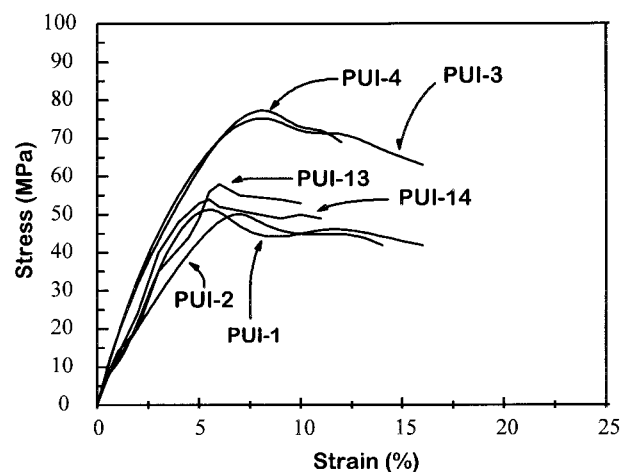| Polymer Code	Tensile Strength (MPa)	Elongation at Break (%)	Initial Modulus (GPa)
PU	38	21	1.91
PIP	65	5	2.15
PUI-1	42	16	1.23
PUI-2	42	14	1.47
PUI-3	63	16	1.86
PUI-4	69	12	1.96
PUI-13	53	10	1.53
PUI-14	49	11	1.39
PIB	76	6	1.95
PUI-5	47	10	1.85
PUI-6	52	7	1.80
PUI-7	65	7	1.92
PUI-8	71	7	1.98
PUI-15	53	7	1.90
PUI-16	42	9	1.94
PID	75	8	1.73
PUI-9	50	7	2.02
PUI-10	58	10	1.94
PUI-11	75	7	1.99
PUI-12	79	6	1.88
PUI-17	53	9	1.88
PUI-18	49	12	1.84

Figure 15 The stress–strain relationship of PMDA series poly(urea-imides).

weights and improvement in their solubility. The solubility of copolymers is found at 0.80–1.08 dL/g and can be cast into a transparent and flexible film. All of these copolymers are amorphous and readily soluble in a wide range of polar solvents because of the presence of the urea and the heterocyclic side-chain structure. The tensile strength of copolymers is increased, but the elongation at break is decreased with the increase of imide content. The strength of copolymer can be even higher than that of pure polyimide, as the imide content up to some extent, regardless of the presence of heterocyclic bulky structure. From the thermal analysis, the T_g of these copolymers are lower than that of polyimide and show a two-stage degradation. The urea linkage in copolymer is therefore considered to be deteriorating their thermooxidative properties.

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