

Synthesis and Characterization of New Aromatic Poly(amide-imide-urea)s Derived from bis(4-Trimellitimidophenyl) urea and Various Aromatic Diamines

Ahmad Banihashemi*, Bahman Tamami, and Amir Abdolmaleki

Department of Chemistry, College of Sciences, Shiraz University, Shiraz-71454, I.R. Iran

Received 16 August 2003; accepted December 2003

ABSTRACT

New aromatic poly(amide-imide-urea)s with enhanced thermal stability and high inherent viscosities were synthesized from a new bis(4-trimellitimidophenyl) urea (**I**) and aromatic diamines. The bis(4-trimellitimidophenyl) urea (**I**) was readily obtained by the condensation reaction of bis(4-aminophenyl) urea with trimellitic anhydride. Polymers were prepared by direct polycondensation reaction using triphenyl phosphite in *N*-methyl-2-pyrrolidone (NMP)/pyridine solution containing dissolved CaCl₂. The obtained poly(amide-imide-urea)s showed high thermostability. Their decomposition temperatures at 10% weight loss in nitrogen atmosphere were above 515 C and the anaerobic char yield at 800 C ranged from 45 to 65%. Almost all the poly(amide-imide-urea)s showed high glass transition temperature above 205 C by differential scanning calorimetry (DSC) measurements. These polymers were readily soluble in various organic solvents and could be cast into transparent, tough, and flexible films.

Key Words:

aromatic polyimides;
poly(amide-imide-urea);
aromatic diamines;
direct polycondensation;
thermal stability.

INTRODUCTION

Aromatic polyimides are well accepted as high-performance polymer materials for their excellent mechanical strength and high thermal stability and balanced mechanical and electrical properties [1-5]. Polyimides are mainly used in the aero-

space and electronics industries in the form of film and mouldings, but high melting points and insolubility in organic solvents limited their applications [6-9]. The initial method of overcoming the intractability of the polyimides utilized the poly(amic

(*) To whom correspondence should be addressed.
E-mail: banihashemi@chem.susc.ac.ir

acid) stage, which could be fabricated into certain end-use forms by virtue of its good solubility in aprotic solvents [10]. Subsequent cyclodehydration of the poly(amic acid) resulted in the polyimide form of the end-use article. While this process has been employed successfully for thin films, the problems associated with the necessary removal of volatiles, make this technique unsatisfactory for other polyimides products such as thick laminated composites and compression moulded materials. To overcome these problems, some copolymers have been proposed in particular, one of them being poly(amide-imide)s (PAIs), whose amide groups can improve solubilities, and also stable in contrast to poly(amic acid)s solutions. Furthermore, poly(amide-imide)s have lighter colour than corresponding polyimides. This class of polymers having bulky and kinked units in their backbone seems to be a good compromise between thermostability and processability. Although poly(amide-imide)s are known as soluble, processable high-temperature polymers, their glass transition temperature (T_g) values were somewhat lower than those corresponding polyimides. Aromatic PAIs can be prepared using performed imide-containing aromatic dicarboxylic acid and various aromatic diamines [11-16].

Urea containing polymers have strong hydrogen bonding and high crystallinity. A wide variety of urea containing polymers has been synthesized in academic and industrial laboratories, but despite their generally good properties, economic factors have limited the commercial development of them [17]. The introduction of urea unit into aromatic polymer backbone has been proven to improve the crystallinity due to strong hydrogen bonding [18]. Various polymers containing ureas group such as polyamide [19], polyimide [20], and polyurethane-urea [21] also have been described in the literature.

In 1974 Yamazaki and co-workers [22,23] reported a procedure for the synthesis of aromatic polyamides, which involved complexing the dicarboxylic acid with triphenyl or diphenyl phosphite and pyridine in a solvent consisting of NMP containing LiCl. Recently, there has been a great deal of scientific activity to achieve a balance between mouldability, mechanical properties and thermal stability of these polymers. New materials were chiefly obtained by copolymerization [24-28]. In our earlier studies [11], a number of

poly(amide-imide)s were successfully synthesized from the direct polycondensation of imide ring containing diacids and aromatic diamines by using the phosphorylation reaction. The present article describes a series of aromatic poly(amide-imide-urea)s with good thermal stability and high T_g , which were synthesized from bis(4-trimellitimidophenyl) urea, and various aromatic diamines by using triphenyl phosphite and pyridine as condensing agents. Solubility, physical property, and thermal properties of the polymers will also be investigated.

EXPERIMENTAL

Materials

All chemicals were purchased from Merck and Fluka. Trimellitic anhydride (above 98%) and triphenyl phosphite (TPP) (above 97%) were used as received. Reagent-grade calcium chloride was dried under vacuum at 180 C before use. *N*-Methyl-2-pyrrolidone (NMP), *N,N'*-dimethylacetamide (DMAc), *N,N'*-dimethylformamide (DMF) and dimethylsulphoxide were purified by distillation under reduced pressure over calcium hydride and stored over 4A molecular sieves. According to a literature [8], 4-phenyl-2,6-bis(4-aminophenyl)pyridine was prepared by the condensation of *p*-nitroacetophenone, benzaldehyde and ammonium acetate, followed by the reduction of the intermediate dinitro compound. Bis(4-aminophenyl) urea (BAU) was synthesized through the reaction of 4-nitroaniline with urea under microwave irradiation [29].

Apparatus

^1H NMR and ^{13}C NMR spectra were recorded on a Bruker Advance DPX-250 250MHz spectrometer in $\text{DMSO}-d_6$ at 25 C with frequencies of 250.13 and 62.89 MHz for the ^1H and ^{13}C spectra, respectively.

Tetramethylsilane was used as an internal standard. FTIR Spectra were recorded on a Shimadzu FTIR-8300 spectrometer. The resolution of the spectrometer was 4 cm^{-1} . The spectra of solids were obtained using KBr pallets. Melting points (mp) were determined with a Buchi 535 melting point apparatus at a heating rate of 1 C/min. Inherent viscosities were measured by a standard procedure using a Cannon Fensk routine viscometer in DMF at a concentration of 0.5 gL^{-1} at 30 C.

Thermogravimetric analysis (TGA) data for polymers were obtained using a Mettler TA4000 system under a nitrogen atmosphere (valve $T/t_{1/2}$) at a rate of 10 C min^{-1} with range FS of 10 mg. Elemental analysis were performed with a Perkin-Elmer 2400 CHN analyzer. Ultraviolet λ_{max} values were obtained in DMSO (99.99%) at a concentration of 0.1 mg mL^{-1} at 25 C using a Shimadzu Multispec-1501 spectrometer.

Synthesis of the Monomers

Synthesis of bis(4-Trimellitimidophenyl) urea (I)

BAU (4.84g, 20 mmol) was dissolved in 15 mL of dried DMAc in a 50 mL flask. After BAU was dissolved completely (8.07g, 42 mmol) trimellitic anhydride was added to it in one portion. The mixture was stirred at room for 1 h. Toluene (10 mL) was then added, and the mixture was heated at the reflux for about 3 h until about 0.35 mL of water was distilled off azeotropically in a Dean-Stark trap. After cooling, methanol was added and the precipitated product was isolated by filtration, purified by recrystallization from DMF-methanol solution, and dried in vacuum, to give

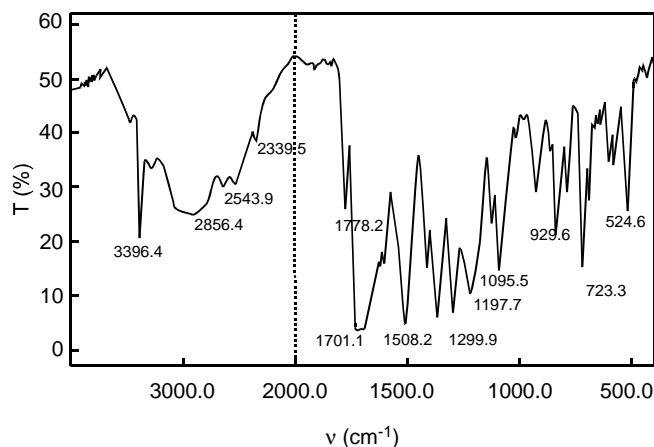


Figure 1. IR Spectrum of bis(4-trimellitimidophenyl) urea (I).

11.57g of bis(4-trimellitimidophenyl) urea (**I**) as yellow solid mp $> 300\text{ C}$ (98% yield). IR (KBr): 3396 (NH urea), 2543-3500 (acid-OH), 1778 (imide, symmetric C=O stretching), 1730 (acid C=O stretching and asymmetric imide C=O stretching) (Figure 1). ^1H NMR (DMSO- d_6 , δ , ppm): 9.07, 8.45-8.43, 8.30, 8.08-8.05, 7.65-7.62, 7.40, 7.36. ^{13}C NMR (DMSO- d_6 , δ ,

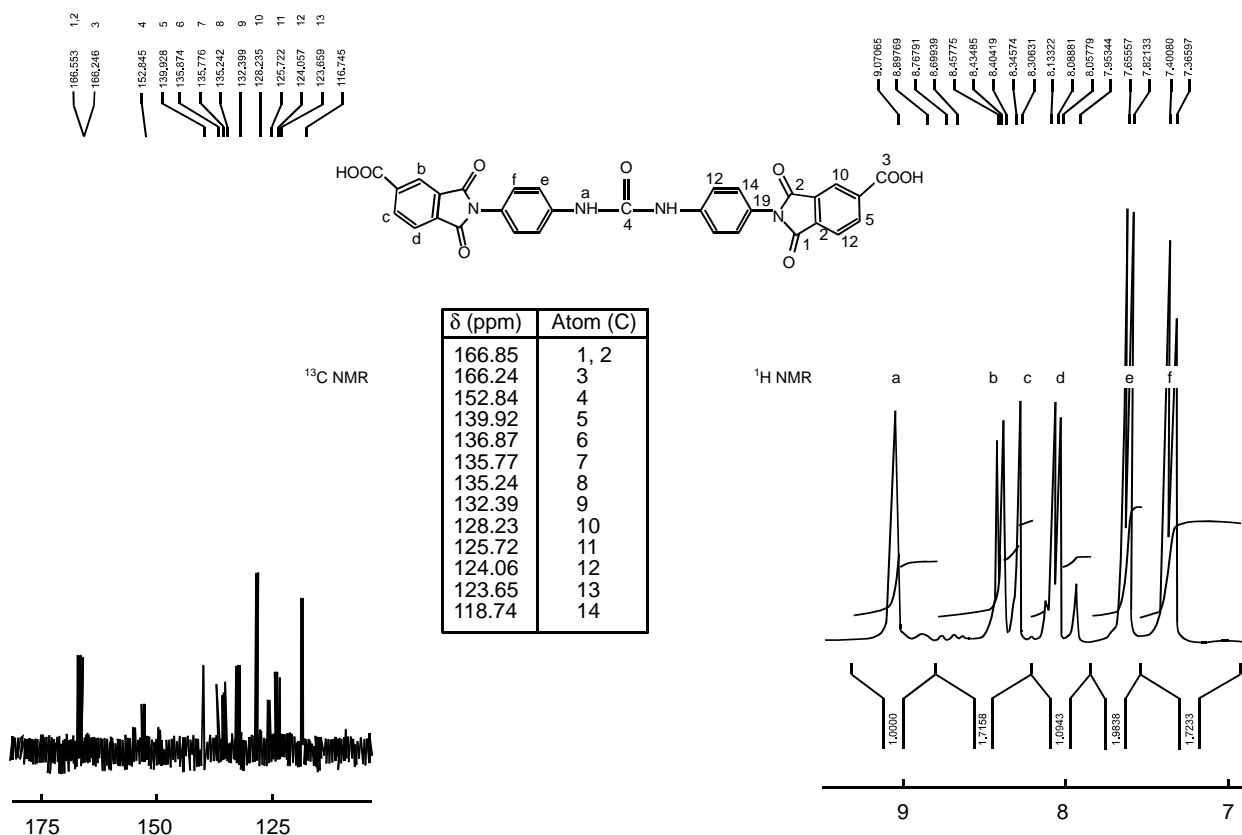


Figure 2. NMR Spectra of bis(4-trimellitimidophenyl) urea (I).

Table 1. Synthesis and some physical properties of poly(amide-imide-urea)s.

Polymer	η_{inh}^a	λ_{max}^b	Yield (%)
PAIUa	1.12	290	98
PAIUb	1.21	282	99
PAIUc	1.06	300	97
PAIUd	1.28	282	95
PAIUe	1.11	288	98
PAIUf	1.14	280	96
PAIUg	1.23	300	98
PAIUh	1.14	298	99
PAIUj	1.29	300	96

^(a) Measured in DMF at a concentration of 0.5 g dL⁻¹ at 30°C; ^(b) obtained in DMSO at a concentration of 0.1 mg mL⁻¹.

ppm): 166.85, 166.24, 152.84, 139.92, 136.87, 135.77, 135.24, 132.39, 128.23, 125.72, 124.06, 123.65, 118.74 (Figure 2).

Analysis (wt%): calculated for C₃₁H₁₈N₄O₉: C, 63.05; H, 3.07; N, 9.48; found: C, 62.80; H, 3.12; N, 9.31.

Synthesis of the Poly(amide-imide-urea)s

A typical example of polymerization was as follows. A mixture of (1 mmol, 0.590 g) of bis(4-trimellitimidophenyl) urea (**I**), (1 mmol, 0.108 g) of diamine (d), (1 g) of calcium chloride, (2.6 mmol) of triphenyl phosphite, (2 mL) of pyridine and (5.0 mL) NMP was heated with stirring at 105 – 5 C for 3 h under nitrogen. The viscosity of reaction solutions increased after 1 h and an additional 5.0 mL of NMP was added to the reaction mixture. After cooling, the reaction mixture was poured

into a large amount of methanol with constant stirring to isolate polymer (PAIUd). The precipitated polymer was separated by filtration and washed with methanol (35 mL) and hot water (100 mL) and dried at 120 C under vacuum for 24 h. All poly(amide-imide-urea)s were synthesized in a similar procedure. The yields were quantitative. Inherent viscosities, λ_{max} values, yield and the results of elemental analyses of obtained polymers are summarized in Tables 1 and 2.

RESULTS AND DISCUSSION

Monomer Synthesis

As shown in Scheme I, the monomer was obtained by the condensation of bis(4-aminophenyl) urea (BAU) with 2 mol equiv of trimellitic anhydride in refluxing DMAc. The condensation reaction between the amines and anhydride groups, and the subsequent cyclodehydration reaction, were carried out in the heterogeneous solution. The structure of the monomer was confirmed by means of FTIR and NMR spectroscopy and elemental analysis. FTIR Spectra of bis(4-trimellitimidophenyl) urea (**I**) are presented in Figure 1. The spectra showed absorption bands around 3396 (urea-NH), 2543-3500 (acid-OH), 1778 (imide, symmetric C=O stretching), 1730 (acid C=O stretching and asymmetric imide C=O stretching) and 1371 cm⁻¹(imide ring vibration, axial), confirming the presence of imide ring, urea and carboxylic acid group in the structure. The bis(4-trimellitimidophenyl) urea (**I**) is also confirmed by NMR spectra (Figure 2). In the ¹H NMR spectrum, the proton of urea H_a appears in the most downfield

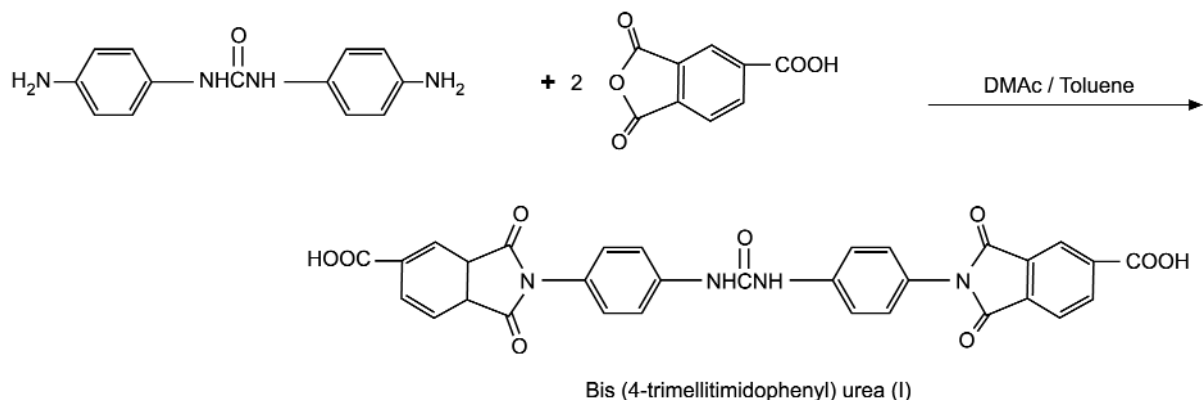
**Scheme I**

Table 2. Elemental analysis of poly(amide-imide-urea)s.

Polymer designation	Moisture uptake ^a	Formula \bar{M}_w		Elemental analysis ^b		
				C (%)	H (%)	N (%)
PAIUa	4.85	$(C_{44}H_{28}N_8O_8)_n$ (796.75) _n	Calcd	66.32	3.54	14.06
			Found	66.18	3.30	14.36
PAIUb	4.36	$(C_{54}H_{33}N_7O_7)_n$ (891.89) _n	Calcd	72.72	3.72	10.99
			Found	72.30	3.74	10.65
PAIUc	4.21	$(C_{43}H_{26}N_6O_9S)_n$ (802.78) _n	Calcd	64.33	3.26	17.93
			Found	64.12	3.36	18.10
PAIUd	4.59	$(C_{37}H_{22}N_6O_7)_n$ (662.61) _n	Calcd	67.06	3.34	12.68
			Found	67.25	3.48	12.62
PAIUe	4.32	$(C_{44}H_{26}N_6O_8)_n$ (766.72) _n	Calcd	68.92	3.25	10.96
			Found	68.73	3.26	10.59
PAIUf	4.05	$(C_{44}H_{28}N_6O_7)_n$ (752.74) _n	Calcd	70.20	3.45	11.16
			Found	69.91	3.50	10.87
PAIUg	3.73	$(C_{43}H_{26}N_6O_8)_n$ (754.71) _n	Calcd	68.43	3.47	11.13
			Found	67.96	3.73	10.67
PAIUh	3.54	$(C_{44}H_{26}N_6O_7)_n$ (750.72) _n	Calcd	70.39	3.49	11.19
			Found	70.06	3.71	10.67
PAIUi	3.29	$(C_{43}H_{26}N_6O_7)_n$ (738.71) _n	Calcd	69.91	3.54	11.37
			Found	69.46	3.78	11.19

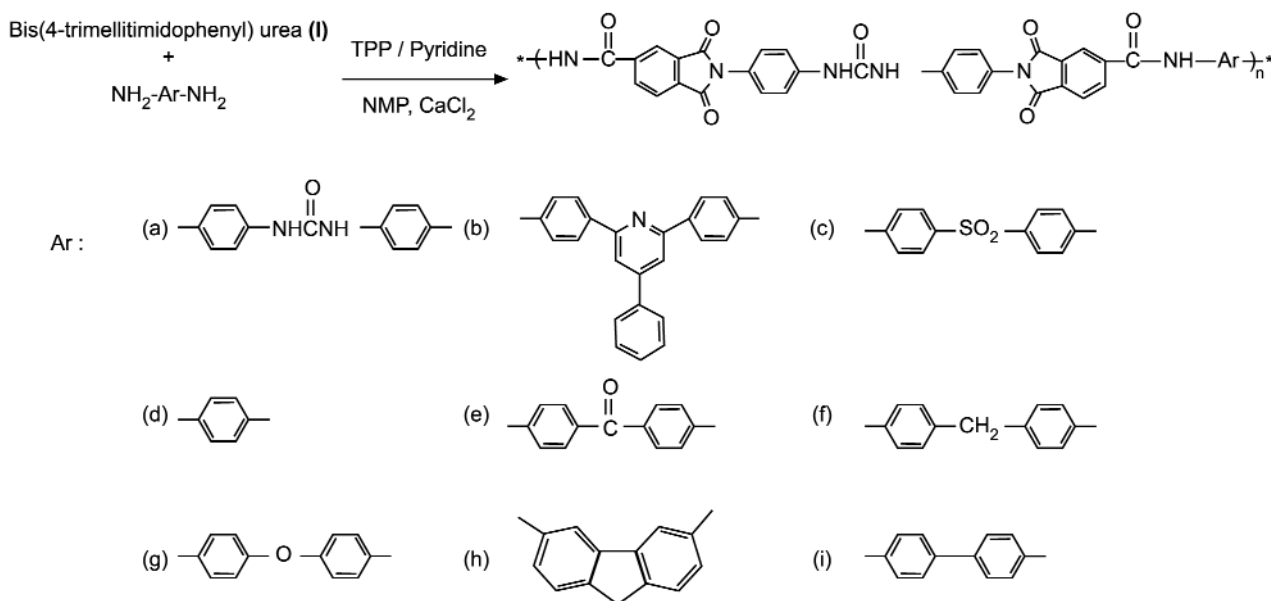
^(a) Moisture uptake(%) = $(W - W_0)/W_0 \times 100\%$; W = weight of polymer sample after standing at room temperature, and W_0 = weight of polymer sample after dried in vacuum at 100°C for 10 h; ^(b) For C and N: corrected value = found value \times (100% + moisture uptake%). For H: corrected value = found value \times (100% - moisture uptake%).

region, the protons $H_{b,c,d}$ adjacent to the carbonyl acid or imide ring (*ortho*-oriented protons) resonated at the farthest downfield region, due to the inductive effect of the carboxyl group and the resonance of the imide ring; the H_e *ortho*-oriented to urea group at the upfield region, due to the shielding effect. The H_f appeared at higher field in accordance to the rest of proton in that were less affected by other groups. The protons of carboxylic acid groups in the 1H NMR spectra were not observed. This was probably due to the fast exchange of these protons with a trace amount of moisture associated with the solvent. The ^{13}C NMR spectra of bis(4-trimellitimidophenyl) urea (**I**) shows 13 signals, including 2 signals of carbonyl of acid and imide($C_{1,2,3}$) and 11 signals of carbon of benzene(C_{4-13}). It will give off 15 signals for carbon of benzene, but the actual measurement showed 14 signals due to

overlap of carbons (Figure 2). The carbons C_1, C_2 and C_3 of imide carbonyl groups and carboxylic acid presented in the downfield at 166.85-166.24 ppm. It was revealed that urea carbon presented at 152.84. These spectra are in agreement with the predicted structures of the monomer.

Polymer Synthesis

New PAIUs were prepared from bis(4-trimellitimidophenyl) urea (**I**) and various aromatic diamines by a phosphorylation reaction with TPP as a promoter in NMP in the presence of pyridine and calcium chloride. Structure and codes of the synthesized polymers are shown in (Scheme II). All polymerizations proceeded homogeneously throughout the reaction. In some cases, an additional 2-6 mL of NMP was added gradually to the reaction mixture after about 1 h to maintain a



Scheme II

degree of agitation. The solubility of polymers and the state of stirring significantly affected the inherent viscosity of the resulting poly(amide-imide-urea)s. In general, the molecular weight of obtained polymer in this polycondensation reaction is highly dependent on the monomer concentration during the initial period of reaction and the amount of inorganic chlorides. High molecular weight polymers were obtained by the use of a higher initial reactant concentration and the addition of a proper amount of supplemental NMP into the viscous reaction medium before the formation of swollen gel. In this study, no attempts were made to maximize molecular weight. Viscosities of the polymerization mixtures were normally very high, and the nature of the precipitated polymers implied high molecular weight products. As shown in Table 1, these polymers were obtained as yellowish materials in quantitative yields with inherent viscosities of up to 1.29 dL/g. The composition and structure of these resulting poly(amide-imide-urea)s were characterized by their IR and NMR spectra and elemental analysis. The typical NMR and IR spectra are shown in Figures 3 and 4. The ^1H NMR spectrum of PAIUa is divided into two parts: the first is due to N-H protons and appears in the most downfield region 9.80 ppm, and the second is due to the aromatic protons in the region 7.34-8.43 ppm. The FTIR spectrum of polymer PAIUa exhibited characteristic absorp-

tion bands for the imide ring at 1776, 1724 (asymmetrical and symmetrical C=O stretching vibration), 1373 (C-N stretching vibration), 1093, and 731 cm^{-1} (imide ring deformation). The results of the elemental analysis of all the poly(amide-imide-urea)s are listed in Table 2. In all cases, the carbon values found were lower than the calculated ones for the proposed structures. This was possibly caused by the hygroscopic nature of the amide groups of these polymers. The moisture uptakes

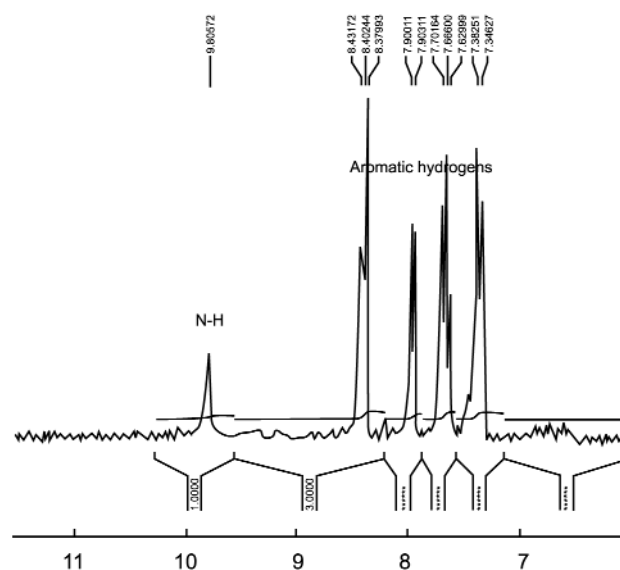


Figure 3. ^1H NMR spectrum of TPMIA1 in DMSO-d_6 at 300 MHz.

Table 3. Solubility of the poly(amide-imide-urea)s^a.

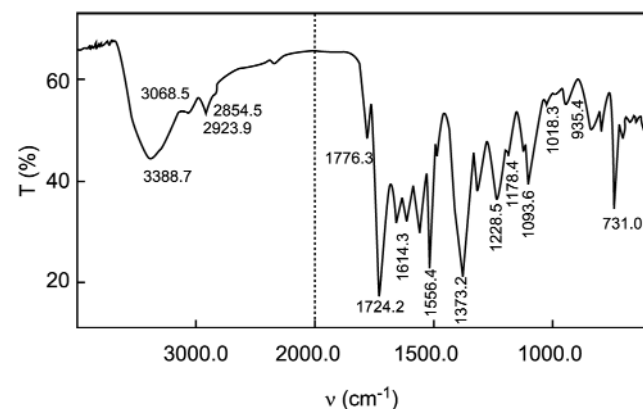
Polymer designation	Solvent ^b						
	Conc.H ₂ SO ₄	NMP	DMAc	DMF	DMSO	<i>m</i> -Cresol	Pyridine
PAIUa	++	++	++	++	++	+	+
PAIUb	++	++	++	++	++	+	+
PAIUc	++	++	++	++	++	+	+
PAIUd	++	+	+	+	+	-	-
PAIUe	++	++	++	++	++	+	+
PAIUf	++	++	++	++	++	+	+
PAIUg	++	+	+	+	+	-	-
PAIUh	++	++	++	++	++	+	+
PAIUi	++	+	+	+	+	-	-

(^a) Concentration: 5mg mL⁻¹; (++) : Soluble at room temperature, (+) : Soluble after 60°C heating, (-) : insoluble; (^b) NMP : *N*-methyl-2-pyrrolidone, DMAc: dimethylacetamide, DMF: dimethylformamide, DMSO: dimethylsulphoxide.

ranged from 3.29-4.85%; these values were calculated from weight change of the vacuum-dried polymer samples after they were exposed in air at room temperature (ca. 27 C) for 1 h. When the found values were corrected by the elimination of the amount of absorbed water, the correction values were in good agreement with the calculated values. The absorptions of amide groups appeared at 3388 (N-H stretch) and 1651 cm⁻¹ (C=O stretch). The inherent viscosities (η_{inh}) and ultraviolet maximum wavelengths (λ_{max}) for the polymers are given in Table 1.

Solubility

The solubility of poly(amide-imide-urea)s was tested qualitatively in various solvents. The results are shown

**Figure 4.** IR Spectrum of PAIUa.

in Table 3. All polymers had excellent solubilities in aprotic polar solvents such as NMP, DMAc, DMF, and DMSO. In addition to highly polar solvents, they were also soluble or swelling in less polar solvents like *m*-cresol and pyridine; however polymers PAIUc, PAIUd, PAIUe, PAIUf PAIUg, and PAIUi with *p*-phenylene and flexible linkage connecting para-aromatic ring structure, respectively, had somewhat limited solubility. This may be due to the semicrystalline nature of these polymers. All the polymers are insoluble in com-

Table 4. Thermal behaviour of poly(amide-imide-urea)s.

Polymer des.	DSC	TGA	
	T _g ^a (°C)	T ₁₀ ^b (%)	Char yield ^c (%)
PAIUa	208	600	55
PAIUb	215	580	53
PAIUc	205	570	65
PAIUd	216	610	53
PAIUe	212	560	57
PAIUf	211	515	51
PAIUg	205	565	53
PAIUh	224	520	45
PAIUi	233	615	59

(^a) From the second heating traces of DSC measurements conducted with a heating rate of 10°C/min; (^b) Temperature at which 10% weight loss was recorded by TG at heating rate of 10°C/min under nitrogen atmosphere; (^c) Char yield in TGA at 800°C under nitrogen atmosphere.

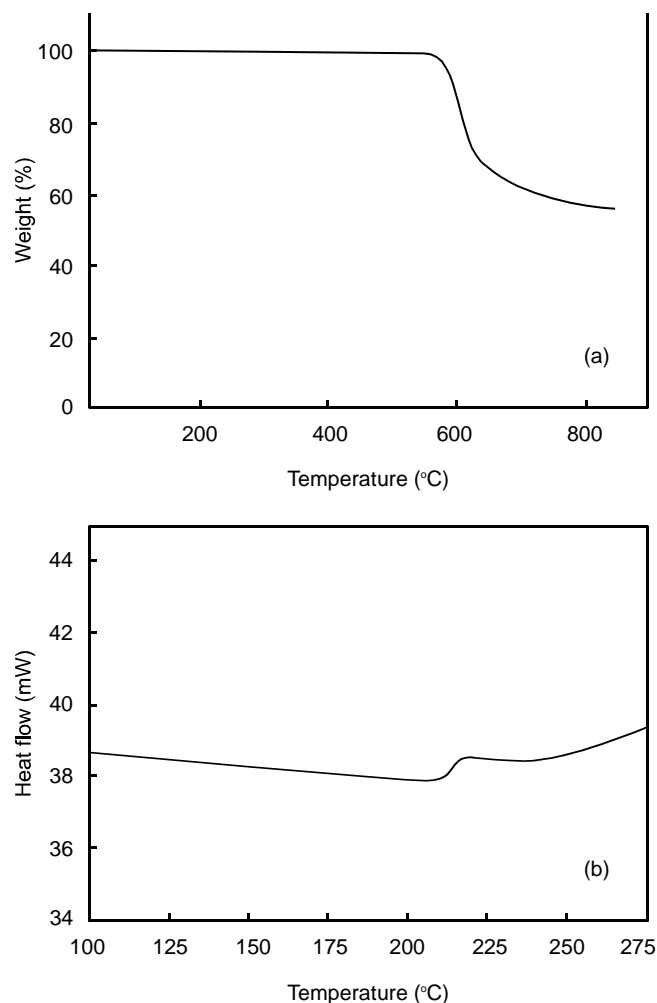


Figure 5. DSC (a), TGA (b) Thermogram of PAIUa at heating rate of $10^{\circ}\text{C}\cdot\text{min}^{-1}$.

mon organic solvents such as tetrahydrofuran, chloroform, acetone and benzene.

Thermal Properties

The thermal properties of all the poly(amide-imide-urea)s were evaluated by thermogravimetry (T_g) and differential scanning calorimetry (DSC). The thermal behaviour data of all polymers are listed in Table 4.

DSC measurements were conducted with a heating rate of $10\text{ C}/\text{min}$ in nitrogen. The T_g values of the poly(amide-imide-urea)s were in the range of $205\text{--}233\text{ C}$, depending on the structure of diamine component. Polymers containing the mono-phenyl group in diamine, which had higher rigidity, exhibited higher T_g values. Polymer having the polar sulphone groups in its backbone also showed high T_g value. The thermal sta-

bility of the poly(amide-imide-urea)s was characterized by TG analysis conducted at heating rate $10\text{ C}/\text{min}$. The temperature of 10% weight loss (T_{10}) in nitrogen was determined from original thermograms. The T_{10} values of these poly(amide-imide-urea)s remained in the range of $515\text{--}615\text{ C}$. They left more than 45% char yield at 800 C (Figure 5).

CONCLUSION

A series of wholly aromatic PAIUs were synthesized by direct polycondensation of bis(4-trimellitimidophenyl) urea (**I**) with various aromatic diamines using TPP and pyridine as condensing agent. These polymers showed excellent solubilities and were characterized by excellent thermal stability as well as high glass transition temperatures. The yellow polymers had η_{inh} and λ_{max} values of $1.06\text{--}1.29\text{ dL/g}$ and $280\text{--}300\text{ nm}$, respectively. Finally, these PAIUs exhibit better processing characteristics than polyamides, polyimides and polyureas with analogous structures.

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