New Poly(azomethine-urethane)s Including Melamine Derivatives in the Main Chain: Synthesis and Thermal Characterization

İsmet Kaya,[1] Mehmet Yıldırım,[1] Musa Kamacı,[2] Ali Avcı[3]

[1]Faculty of Sciences and Arts, Department of Chemistry, Çanakkale Onsekiz Mart University, 17020 Çanakkale, Turkey
[2]Kamil Özdağ Science Faculty, Department of Chemistry, Karamanoğlu Mehmetbey University, 70100 Karaman, Turkey
[3]Faculty of Sciences and Arts, Department of Chemistry, Celal Bayar University, 45040 Manisa, Turkey

ABSTRACT: Up to date, only a few kinds of poly(azomethine-urethane)s (PAMUs) derived from aromatic hydroxy compounds were obtained and studied with thermal degradation steps. Novel PAMUs were prepared using the hydroxy-functionalized Schiff bases derived from melamine and toluene-2,4-diisocyanate. Schiff base prepolymers were synthesized by the condensation reaction of melamine with 4-hydroxybenzaldehyde and 2-hydroxy-1-naphtaldehyde. Characterization was made by UV–Vis, FTIR, NMR, and SEC techniques. Thermal charac-terizations of the novel PAMUs were carried out by TG-DTA and DSC techniques. Thermal decomposition steps at various temperatures were also clarified and the physical changes of the synthesized PAMUs with exposing to the thermal degradation steps were displayed. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 120: 3027–3035, 2011

Key words: poly(azomethine-urethane); thermal degradation; polyurethane; Schiff base; melamine

INTRODUCTION

Polyurethanes (PU) have a wide range of applications in medical, automotive, and industrial fields. They can be used in several products such as furniture, coatings, adhesives, constructional materials, fibers, paddings, paints, elastomers, and synthetic skins.[1] A lot of studies exist in the development of thermally stable polymers. Several organic polymers have useful structural and mechanical properties, which are limited by their thermo-oxidative stability and high flammability. A typical example of these polymers is PU those are widely used in the aerospace and construction industries. Their thermal stabilities have been studied extensively.[2–4] However, PU are highly combustible.[5]

As well known, polyurethane foams have been employed in many application fields due to their unique properties like high thermal stability, anti-flammable, antibacterial, high durability protective coatings, etc. Polyurethane coatings could be applied onto farming equipments, wood grain surfaces, car trims, boat panels, mobile phones, motor bike parts, trucks, ute beds, floor coatings, hoopers, etc.[6]

One attractive class of macromolecules is poly(azomethine)s. They contain azomethine ($-CH=N-$) linkages in the backbone, and are known to exhibit good thermal stability and many desirable properties such as electrical conductivity, thermal stability, solubility due to the resonance stabilization of poly Schiff's base unit.[7] Polyazomethines (PAMs), with a wide range of applications, have had continual increasing interest due to having a lot of useful properties such as high thermal stability and excellent mechanical strength as well as their semiconductivity and optoelectronic properties.[8,9] On the other hand, PAMs, especially aromatic derivatives suffer from low solubility. To solve this problem, many kinds of PAMs like poly(azomethine ether)s,[10] poly(acrylate-azomethine)s,[11] poly(azomethine carbonate)s,[12] poly(amide-azomethine-ester)s,[13] and poly(azomethine sulfone)[14] have been synthesized so far. Additionally, PAMs containing methoxy substituents had been presented with fine solubility and high thermal stability.[15]

Oligophenols and their azomethine derivatives had been previously synthesized by oxidative poly condensation method and presented in the literature with their several useful properties.[16–20] This method is easy to apply and environmentally harmless due to the usage of water as a medium. Also using of cheap oxidants such as NaOCl, H_2O_2, and air O_2 is another advantage of this method. Several investigations are made on the development of heat-resistant

polymers such as polyesters,[21,22] polyethers,[23] poly siloxanes,[24] etc., containing azomethine linkages in the polymer backbone. A lot of interesting studies on synthesis of PU containing azomethine groups have been reported.[25–30] However, only a few studies have been focused on the effect of the azomethine linkages on thermal stability.[25,26] The previously published poly(azomethine-urethane)s (PAMUs) had fine solubilities as well as good thermal stabilities which degraded upon 200°C.[25] At the present study we synthesized new kinds of the PAMUs including melamine linkages. Melamine has an unusual structure with several number of the azomethine linkage in a ring form. We aimed to investigate the effect of that interesting structure on the properties of the PAMUs. Melamine has been used in production of several resins with formaldehyde, urea, etc. so far.[31,32] However, the usage of melamine in synthesis of azomethine compounds and polymers are very limited and needs to be carry out.[33] On the other hand, the used aldehydes with phenylene and naphthalene structures were chosen to determine the effect of the increasing conjugated and rigid structure on the thermal behaviors by increasing of the aromatic ring number.

At the first part of this study, we synthesized the Schiff base monomers by condensation reaction of melamine with aromatic hydroxyaldehydes like 4-hydroxybenzaldehyde and 2-hydroxy-1-naftaldehyde. Then, we converted these compounds to polyurethane derivatives using toluene-2,4-diisocyanate (TDI) as the comonomer. We characterized the synthesized compounds using FTIR, UV–Vis, ^{1}H and ^{13}C-NMR, and SEC analyses. TG-DTA technique was used to understand and clarify the thermal degradation steps. Thermal degradation steps were also characterized by FTIR analysis of the degraded products. DSC analyses of the PAMUs were also carried out to determine the glass transition temperatures (T_g).

EXPERIMENTAL

Materials

TDI, melamine, 4-hydroxybenzaldehyde, 2-hydroxy-1-naphtaldehyde, dimethylformamide (DMF), dimethylsulfoxide (DMSO), tetrahydrofurane (THF), methanol, acetonitrile, acetone, toluene, ethyl acetate, heptane, hexane, CCl_4, $CHCl_3$, and H_2SO_4 were supplied from Merck Chemical (Germany) and they were used as received.

Syntheses of the prepolymers

Prepolymers (PPs) Ia and Ib were synthesized by simple condensation reaction of melamine with 4-
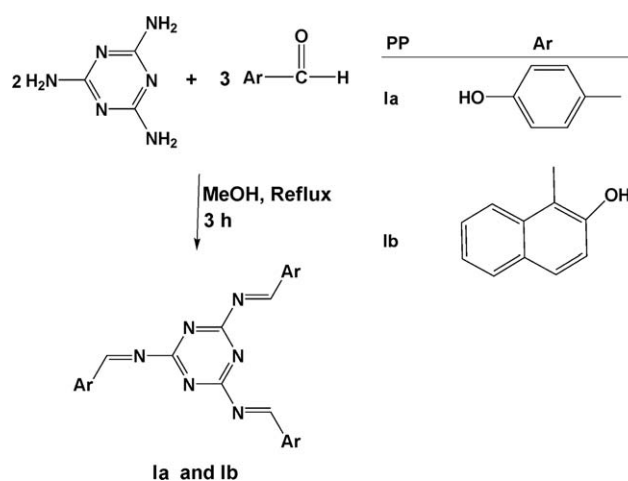

Scheme 1 Syntheses of the prepolymers Ia and Ib.

hydroxybenzaldehyde and 2-hydroxy-1-naphtaldehyde.[33] Reactions were made as follows: Melamine (0.630 g, 0.005 mol) was placed into a 250-mL three-necked round-bottom flask which was fitted with condenser, thermometer, and magnetic stirrer. About 80 mL DMF was added into the flask. Reaction mixture was heated to 140°C and at this temperature melamine was solved. A solution of excess amount of 4-hydroxybenzaldehyde (2.440 g, 0.02 mol) or 2-hydroxy-1-naphtaldehyde (3.440 g, 0.02 mol) in 20 mL. DMF was added into the flask. All reactions were maintained for 3 h under reflux (Scheme 1). Reaction mixture cooled at room temperature and was slowly dropped into 200 mL toluene to precipitate the Schiff bases. Product was filtered and washed with methanol/toluene (1 : 1) to separate the unreacted aldehyde from the condensation products. Then, it was dried in a vacuum desiccator (yields: 37 and 78% for Ia and Ib, respectively).

Syntheses of the poly(azomethine-urethane)s

The synthesized prepolymers with azomethine linkages (Ia and Ib) were used in synthesizing of the PAMUs. Synthesis procedure of the PAMUs is as follows:[25] TDI (1.74 g, 10^{-2} mol) was dissolved in 50 mL THF and added into a 250-mL three-necked round-bottom flask which was fitted with condenser, magnetic stirrer, and inert argon gas supplier. Reaction mixture was heated up to 60°C and equivalent amount of PP (7.62 g of Ia, 10.24 g of Ib) was added into the flask. Reactions were maintained for 6 h, cooled at the room temperature, and kept for 24 h. THF was removed in evaporator. The obtained polymer was washed by methanol (2 × 50 mL), acetonitrile (2 × 50 mL), and water (2 × 100 mL) to remove the unreacted components. The products were dried in a vacuum oven at 80°C for 24 h (Scheme 2)
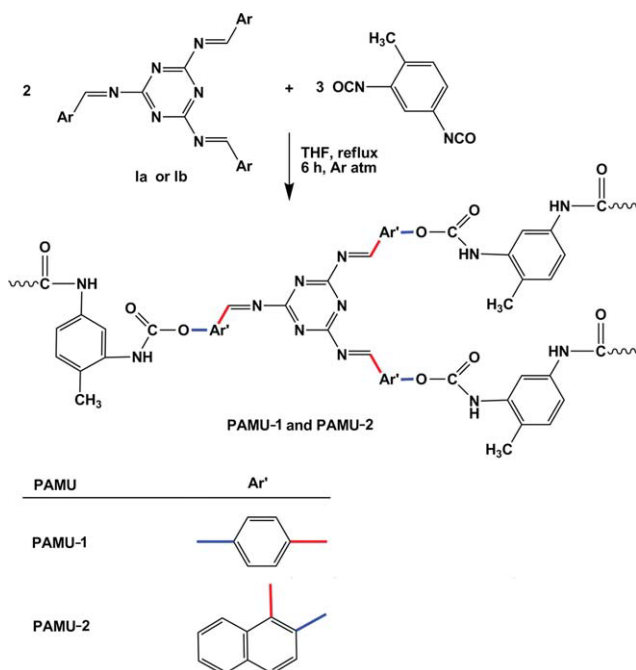
Scheme 2 Syntheses of PAMU-1 and PAMU-2. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

(yields 96 and 71% for PAMU-1 and PAMU-2, respectively).

Characterization techniques

The solubility tests were carried out in different solvents by using 1 mg sample and 1 mL solvent at 25°C. The IR and UV–Vis spectra were measured by Perkin–Elmer FTIR Spectrum one and Perkin–Elmer Lambda 25, respectively. The FTIR spectra were recorded using universal ATR sampling accessory

(4000–550 cm^{-1}). UV–Vis spectra of the synthesized compounds were determined by using DMSO. ^{1}H and ^{13}C-NMR spectra (Bruker AC FT-NMR spectrometer operating at 400 and 100.6 MHz, respectively) were recorded using deuterated DMSO-d$_6$ as a solvent at 25°C. Tetramethylsilane was used as internal standard. Thermal data were obtained using Perkin–Elmer Diamond Thermal Analysis. The TG-DTA measurements were made between 20 and 1000°C (in N$_2$, 10°C/min). DSC analyses were carried out using Perkin–Elmer Pyris Sapphire DSC. DSC measurements were made between 25 and 420°C (in N$_2$, 20°C/min). The number average molecular weight (M_n), weight average molecular weight (M_w), and polydispersity index (PDI) were determined by size exclusion chromatography (SEC) techniques of Shimadzu For SEC investigations, an SGX (100 Å and 7 nm diameter loading material) 3.3 mm i.d. × 300 mm columns was used; eluent: DMF (0.4 mL/min), polystyrene standards were used. Moreover, refractive index detector (RID) and a UV detector were used to analyze the products at 25°C.

RESULTS AND DISCUSSION

Solubilities and structures of the poly(azomethine-urethane)s

Prepolymer Ia and its poly(azomethine-urethane) compound (PAMU-1) are light-yellow colored, while Ib and PAMU-2 are brown colored. Solubility test results showed that the obtained PAMUs are completely soluble in only strongly polar solvents like DMSO, DMF, and H$_2$SO$_4$. They are all insoluble in methanol, ethanol, THF, acetonitrile, ethyl acetate, hexane, heptane, acetone, and chlorinated solvents like chloroform and CCl$_4$.
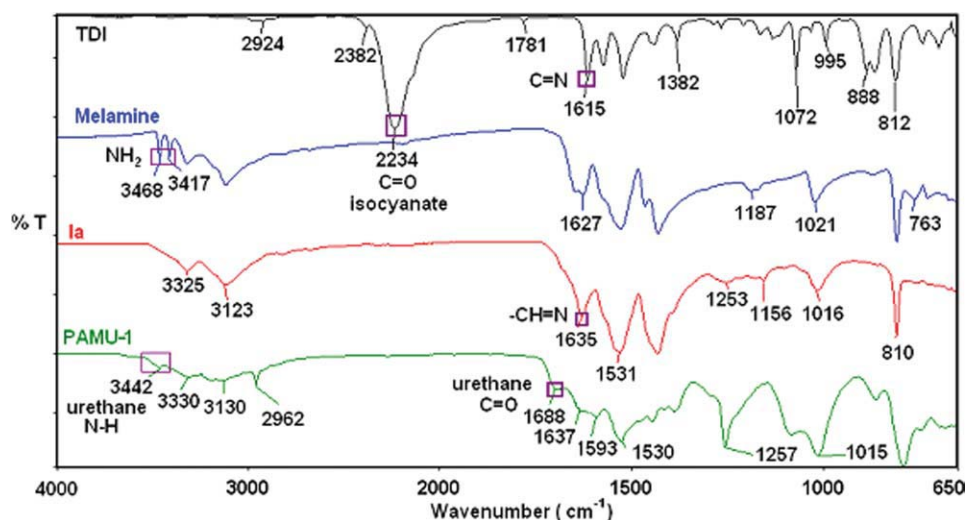

Figure 1 FTIR spectra of TDI, melamine, Ia, and PAMU-1. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
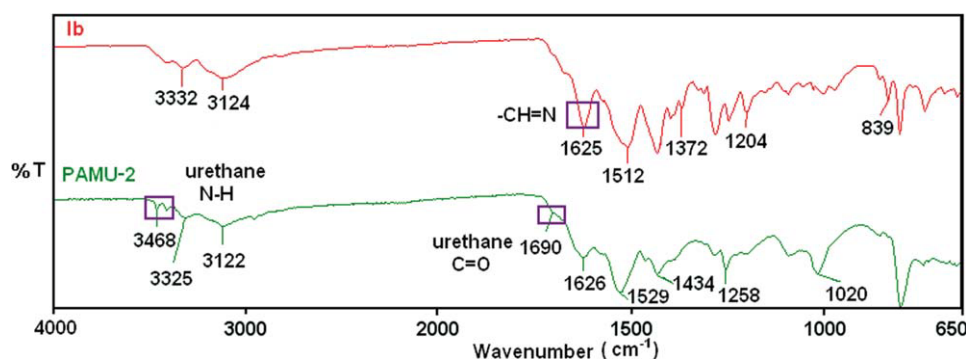
Figure 2 FTIR spectra of Ib and PAMU-2. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

FTIR spectra of TDI, melamine, Ia, PAMU-1 are given in Figure 1 and Ib and PAMU-2 in Figure 2. At the spectrum of TDI characteristic isocyanate C=O peak is observed at 2234 cm^{-1} which agrees with the literature values.[30] However, at the spectra of the PAMUs this peak disappears as a result of the urethane formation. Also, at the spectrum of TDI C=N stretch vibration of isocyanate group is observed at 1615 cm^{-1} which disappears at the other spectra. At the PAMU-1 and PAMU-2 spectra the new peaks appear at 3442 and 3468 cm^{-1}, and 1688 and 1690 cm^{-1} indicating the urethane N—H stretch and urethane carbonyl (C=O) stretch vibrations, respectively. Azomethine bonds (C=N) in the structures of PAMU-1 and PAMU-2 are observed at 1593 and 1626 cm^{-1}, respectively, which are a bit lower than those of their prepolymers (PPs). This is probably due to electron withdrawing effect of the urethane groups in the polymer structures which decreases the electron density of imine carbon and consequently imine vibration, as observed in the previous studies.[30] The peaks at 3330 and 3325 cm^{-1} at the spectra of PAMU-1 and PAMU-2 are attributed to the O—H stretch vibrations of the unreacted

(free) —OH groups of Ia and Ib. Some additional peaks including aliphatic C—H vibration (2962 and 2960 cm^{-1}) and aromatic C=C stretch (1530 and 1529 cm^{-1}) are also shown in Figures 1 and 2. The observed results clearly confirm the polyurethane formation.

UV–Vis spectra of the synthesized PPs and PAMUs are also comparatively given in Figure 3. As seen in Figure 3, lower conjugations of the synthesized PAMUs cause a bit shift in absorption edges into lower wavelengths resulting in higher band gaps with compared those of the prepolymers.[34] According to Figure 3 aromatic bands are observed at 286 and 259 nm for Ia and Ib prepolymers due to benzene $\pi \to \pi^*$ transitions, respectively. Similarly, at the spectra of PAMU-1 and PAMU-2 the same transitions are observed at 265 and 271 nm, respectively. On the other hand, the absorption peaks of Ib and PAMU-2 are placed at higher wavelengths due to higher conjugated structure of naphthalene rings. Additionally, the urethane linkage containing unpaired electrons on oxygen and nitrogen atoms enabled many resonance structures resulting in pseudo conjugation.[35]
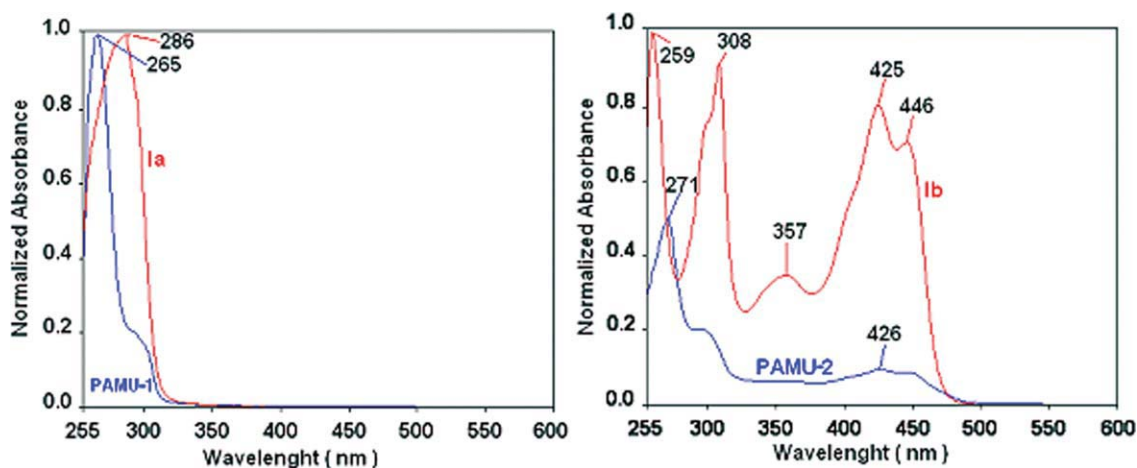

Figure 3 UV–Vis spectra of the synthesized PPs and PAMUs. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
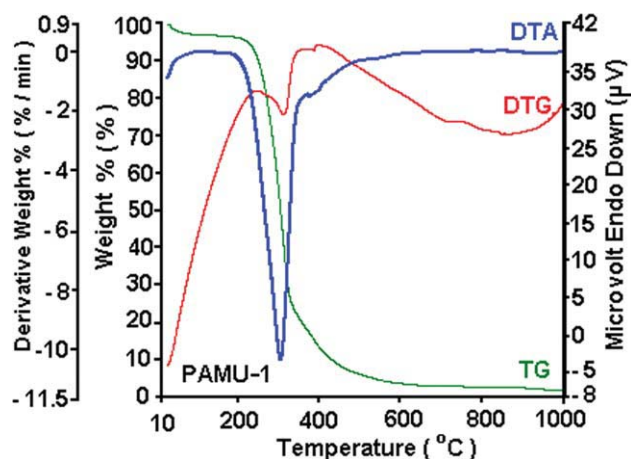
Figure 4 ^{1}H-NMR (a) and ^{13}C-NMR (b) spectra of PAMU-2.

The ^{1}H-NMR spectra of all synthesized compounds are obtained and the peak assignments are given below. Also, ^{1}H and ^{13}C-NMR spectra of PAMU-2 are shown in Figure 4. According to Figure 4(a) urethane and imine protons (—N*H*CO and —N=C*H*) are observed at 9.87 and 8.57 ppm, respectively. The peak at 13.62 ppm is attributed to the

unreacted (free) —OH groups of Ib which states at 13.57 ppm at the spectrum of Ib. Similarly, the —OH peak of Ia at 10.92 ppm does not completely disappear after the polyurethane formation indicating the presence of the free —OH groups. This result is supported by the FTIR results, as emphasized above. Methyl groups are observed at 2.54 and 2.51 ppm for PAMU-1 and PAMU-2, respectively. ^{13}C-NMR spectrum of PAMU-2 also confirms the structure by the peaks observed at 163.42, 152.91, and 17.94 ppm which could be attributed to the urethane, imine, and —CH$_3$ carbon resonances, respectively.

PAMU-1

Ia: ^{1}H-NMR (DMSO): δ ppm, 10.92 (s, —O*H*), 9.29 (s, —C*H*=N—), 7.76 (d, Ar—Ha), 6.93 (d, Ar—Hb). PAMU-1: ^{1}H-NMR (DMSO): δ ppm, 10.92 (s, —O*H* [free]), 9.80 (s, —N*H*), 9.17 (s, —C*H*=N—), 7.73 (d, 2H, Ar—Ha), 7.47 (s, Ar—Hc), 6.91 (d, 2H, Ar—Hb), 6.60 (d, Ar—Hd), 6.42 (d, Ar—He), 2.54 (s, —CH$_3$). Ib: ^{1}H-NMR (DMSO): δ ppm, 13.57 (s, —O*H*), 8.82 (s, —C*H*=N—), 7.90 (d, Ar—Hf), 7.86 (d, Ar—Hc), 7.80 (d, Ar—Hb) 7.32 (t, Ar—He), 7.10 (t, Ar—Hd), 6.71 (d, Ar—Ha).

PAMU-2: ^{1}H-NMR (DMSO): δ ppm, 13.62 (s, —O*H* [free]), 9.87 (s, —N*H*), 8.57 (s, —C*H*=N—), 7.86 (d, Ar—Hf), 7.81 (d, Ar—Hc), 7.63 (d, Ar—Hb), 7.49 (s, Ar—Hg), 7.29 (t, Ar—He), 7.05 (t, Ar—Hd), 6.85 (d, Ar—Hh), 6.63 (d, Ar—Ha), 6.45 (d, Ar—Hi), 2.51 (s, —CH$_3$).

According to the SEC chromatograms, the calculated number-average molecular weight (M_n), weight average molecular weight (M_w), and polydispersity index (PDI) values of the synthesized PAMUs measured using both RID and UV detectors are given in

TABLE I
SEC Analyses Results of the Synthesized PAMUs

				Molecular weight distribution parameters											
	Total			Fraction I				Fraction II				Fraction III			
Compounds	M_n	M_w	PDI	M_n	M_w	PDI	%	M_n	M_w	PDI	%	M_n	M_w	PDI	%
PAMU-1[a]	1990	2190	1.100	2000	2200	1.100	99	900	950	1.009	1	–	–	–	–
PAMU-1[b]	1600	2290	1.431	1700	2500	1.485	86	950	1000	1.009	14	–	–	–	–
PAMU-2[a]	2160	2230	1.032	7600	8850	1.161	1	2500	3300	1.332	1	2100	2150	1.025	98
PAMU-2[b]	2450	2800	1.143	7900	9100	1.153	9	2100	2800	1.326	2	1900	2150	1.140	89

[a] Determined by refractive index (RI) detector.
[b] Determined by UV detector.

Figure 5 TG-DTG-DTA curves of PAMU-1. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
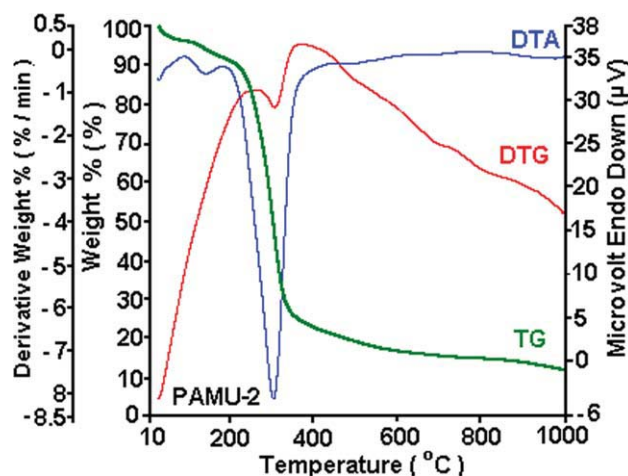

Figure 6 TG-DTG-DTA curves of PAMU-2. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. According to the total values, PAMU-1 and PAMU-2 have quite low molecular weights indicating that the synthesized materials are oligomers. Also, the average molecular weight of PAMU-2 is a bit higher than that of PAMU-1. The obtained results show that the synthesized PAMUs contain only a few repeated units.

Thermal characterization

TG-DTG-DTA curves of PAMU-1 and PAMU-2 are given in Figures 5 and 6, respectively. Thermal degradation values are also summarized in Table II. According to the obtained thermograms, both of the synthesized PAMUs decompose in a single step that ranges from 230 to 420°C. Table II indicates that PAMU-1 has higher onset temperature that extends from 235 to 371°C. Also, the peak degradation temperature and char residue of PAMU-2 at 1000°C are higher than those of PAMU-1. These differences probably depend on the prepolymer structures. The prepolymer structure of PAMU-1 includes phenylene rings bounded with melamine while PAMU-2 includes thermally-stable naphthalene rings. Thus, PAMU-2 has large side chains and consequently highly resist against thermal degradation. Moreover, the char residues at 1000°C are 2 and 12%, respectively.

On the other hand, physical changes of the synthesized PAMUs with exposing to the thermal degradation steps are displayed using a "Metler Toledo MP70." The obtained thermodegradation photographs of PAMU-1 and PAMU-2 at various temperature points are given in Figures 7 and 8, respectively. Figure 7 indicates a clear color change from light to dark colors with increasing temperature. At the temperature of 240°C the color is yellow, while at the 280°C the color occurs darker indicating the thermal degradation step of PAMU-1, as given in Figure 5 and Table II. Similarly, as emphasized above thermal degradation start point of PAMU-2 is 229°C. As seen in Figure 8 upon this temperature, the degradation of PAMU-2 clearly observed as a color change.

DSC traces of the synthesized PAMUs are given in Figure 9. According to the obtained DSC curves, the glass transition temperatures (T_g) are calculated as 244 and 207°C for PAMU-1 and PAMU-2, respectively. The broad peaks until 170°C could be attributed to the absorbed solvent removal.[36]

Thermal degradation

It is known that the thermal degradation of PUs occurs in a two- to three-step process.[37–42] The first

TABLE II
TG-DTG-DTA and DSC Thermal Data of the Synthesized PAMUs

	First degradation temperature (°C)			TG-DTG-DTA		DSC	
Compounds	T_{on}[a]	T_{max}[b]	Percentage of weight loss	Char at 1000°C (%)	Endothermic peak temperature (°C)	T_g (°C)	ΔC_p (J/g K)
PAMU-1	241	267	78	2	314, 388	235	0.054
PAMU-2	229	310	71	12	309	207	0.171

[a] Thermal degradation onset temperature.
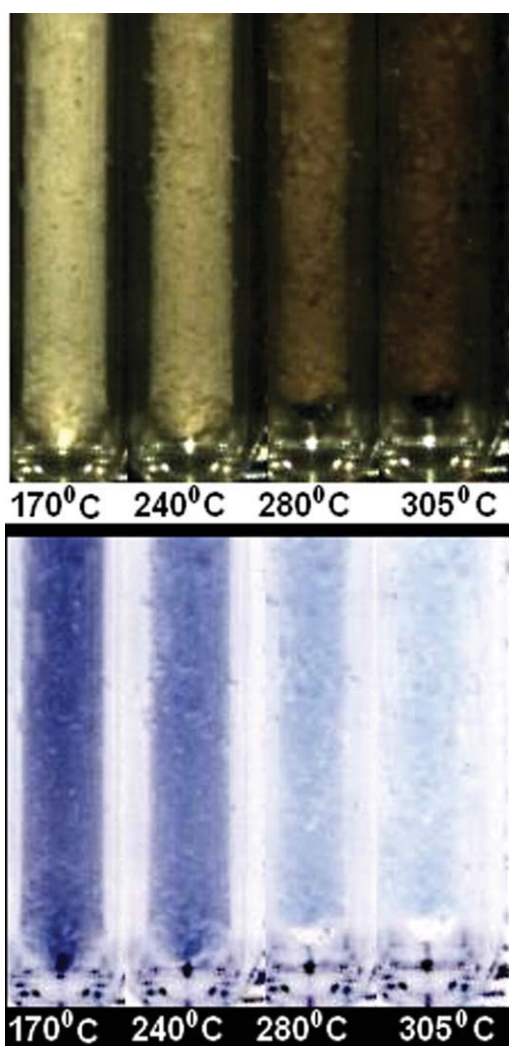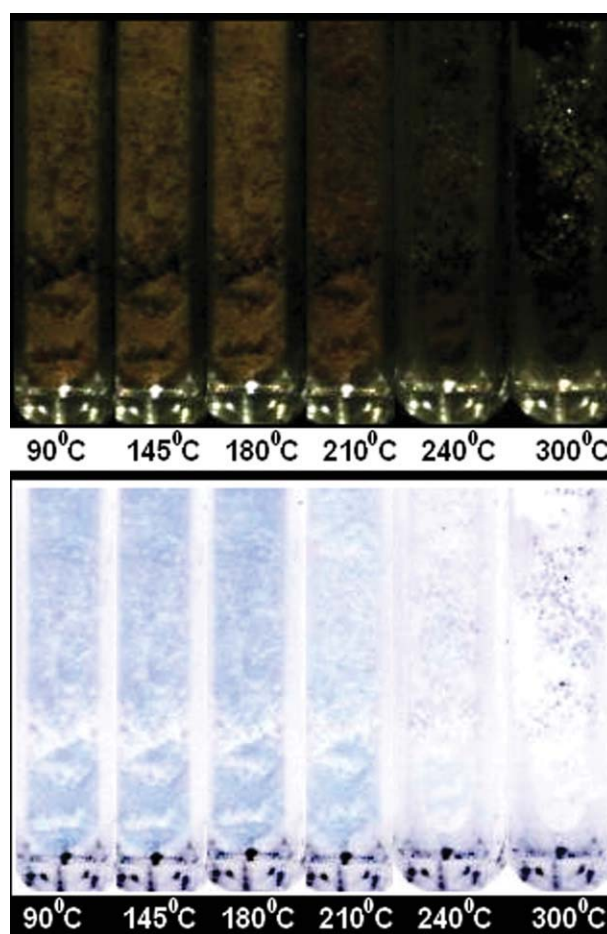[b] Maximum weight loss temperature.

Figure 7 Thermodegradation photographs of PAMU-1 in various steps (top) and the reverse-colored photographs (bottom). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

step is due to degradation of the hard segment, which results in the formation of isocyanate and alcohol, primary or secondary amine, olefin, and carbon dioxide. The second and third steps correspond to the thermal decomposition of the soft segment. The thermal stability of PUs depends primarily on the polymerization↔depolymerization equilibria of the functional groups in the polymer molecule.[43] The isocyanate formed during thermal decomposition may be dimerized to carbodiimide. Carbodiimide can then react with urethane groups to form a crosslinked structure. The obtained results could be also confirmed by FTIR spectra of the thermodegradation products.

To explain the thermodegradation steps of the synthesized PAMUs, FTIR spectra of the products after heating until various temperatures are obtained and given in Figures 10 and 11, respectively. At the

Figure 8 Thermodegradation photographs of PAMU-2 in various steps (top) and the reverse-colored photographs (bottom). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

spectra of PAMU-1, characteristic corbodiimide ($-N=C=N$) peak is observed at 1707 cm^{-1} for 370°C, and 1712 cm^{-1} for both 420 and 500°C, which agree with the literature values.[43] Similarly, at the spectra of PAMU-2, characteristic corbodiimide ($-N=C=N$) peak appears at 1710 cm^{-1} for both 420
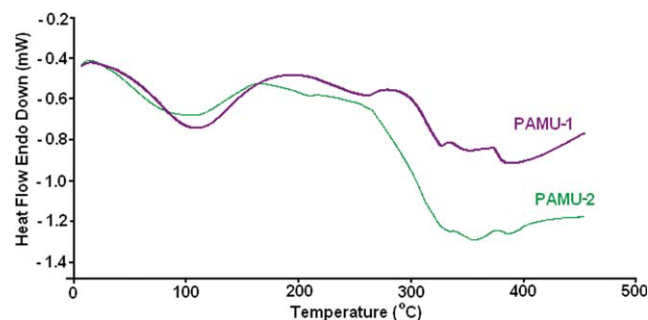

Figure 9 DSC curves of the synthesized PAMUs. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
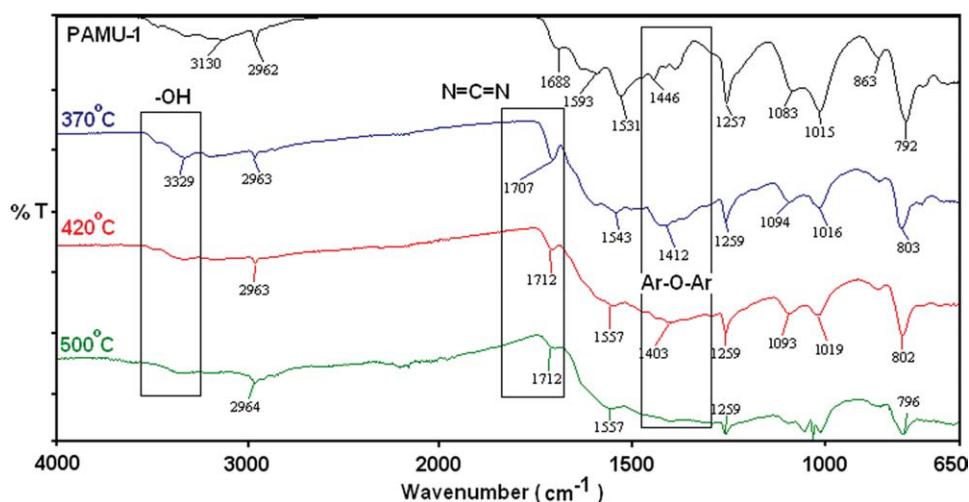
Figure 10 FTIR spectra of PAMU-1 after heating until various temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

and 435°C. However, this peak appears neither at 180°C nor 500°C. The decreasing of the carbodiimide peak intensities at higher temperatures (e.g., 500°C) indicates the degradation of the N=C=N bond to form new volatile products at high temperatures. Additionally, after the depolymerization step with heating up to higher temperatures —OH functionalized Ia and Ib are expected to form new ether-bridged structures with dehydration.[41] As seen in Figures 10 and 11, when PAMU-1 and PAMU-2 are heated up to 370 and 420°C, respectively, new broad absorption bands appear at 1403–1412 and 1332–1357 cm^{-1} indicating the Ar—O—Ar ether bond formation. Also, the intensity of O—H stretch vibrations at 3329 and 3324 cm^{-1} decrease due to the dehydration (Scheme 3).

CONCLUSION

Novel poly(azomethine-urethane)s were synthesized using melamine-based azomethine bounded-phenol compounds. The obtained PAMUs had highly conjugated and aromatic structures. Thermal characterizations were carried out by TG-DTA and DSC techniques. Thermal degradation steps of the new PAMUs were clarified using the FTIR spectra of the degraded forms at various temperatures. Physical changes of the PAMUs were displayed at various temperatures showing that the colors of the PAMUs change from light to dark forms as a result of the thermal degradation. The synthesized PAMUs were found to be thermally stable up to 230–240°C, which were higher than those of the previously synthesized kinds. The PAMU containing naphthalene rings was more thermally
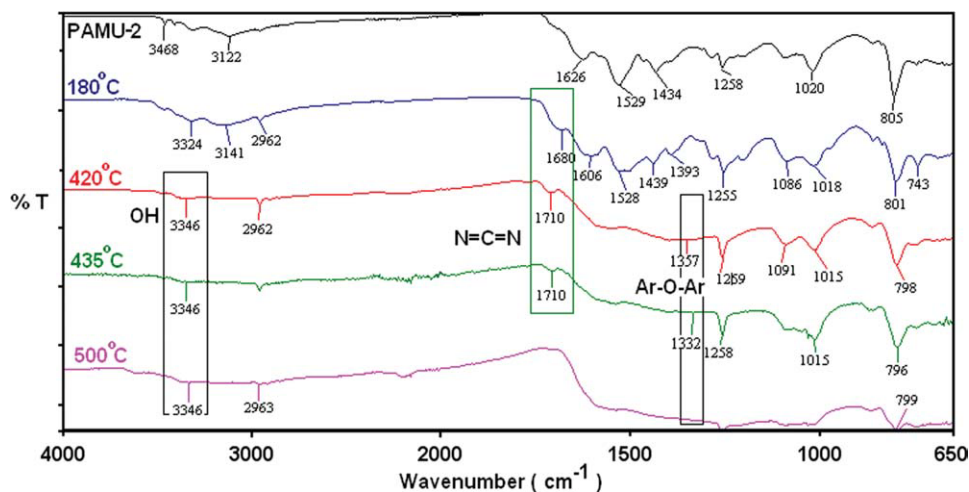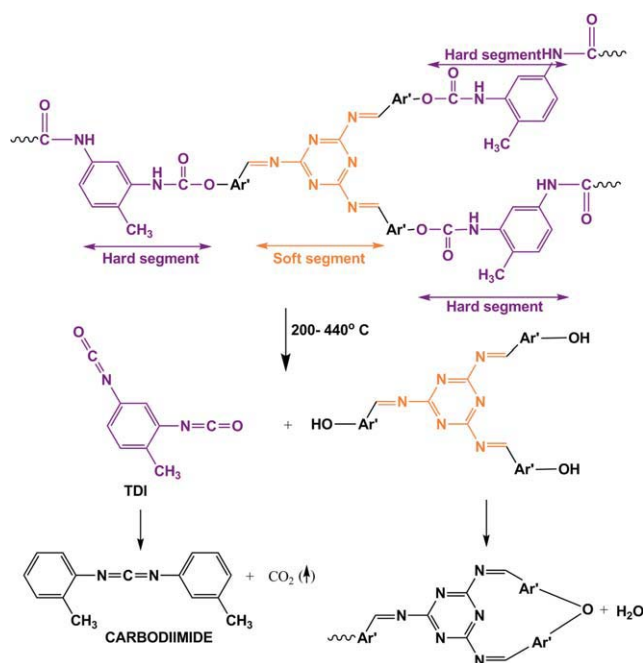

Figure 11 FTIR spectra of PAMU-2 after heating until various temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Scheme 3 Thermal degradation steps of the synthesized PAMUs. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

stable than the PAMU containing phenylene rings. Resultantly, melamine linkage provided fine thermal stability to the synthesized PAMUs.

References

1. Howard, G. T. Int Biodeter Biodegr 2002, 49, 245.
2. Gaboriaud, F.; Vantelon, J. P. J Polym Sci Polym Chem Ed 1982, 20, 2063.
3. Ballistreir, A.; Foti, S.; Maraavigna, P.; Montaudo, G.; Scamperrino, E. J Polym Sci Polym Chem Ed 1980, 18, 1923.
4. Barendregt, R. B.; Van Den Berg, P. J. Thermochim Acta 1980, 38, 181.
5. Reed, C. S.; Allcock, H. R.; Coleman, M. M. Soc Plast Eng Annu Tech Conf 2000, 40, 465.
6. http://www.fatcow.com.au/c/Unique-Coating-Solutions/Polyurethane-Coatings-from-Unique-Coating-Solutions-p20937.
7. Segal, C. L. High Temperature Polymers; Marcel Dekker: New York, 1967.
8. Wang, C. G.; Shieh, S.; LeGoff, E.; Kanatzidis, M. G. Macromolecules 1996, 29, 3147.
9. Grigoras, M.; Catanescu, C. O. J Macromol Sci Part C Polym Rev 2004, 44, 131.
10. Marin, L.; Cozan, V.; Bruma, M.; Grigoras, V. C. Eur Polym Mater 2006, 42, 1173.
11. Tanaka, H.; Shibahara, Y.; Sato, T.; Ota, T. Eur Polym J 1993, 2, 1525.
12. Sun, S. J.; Chang, T. C.; Li, C. H. Eur Polym J 1993, 29, 951.
13. Li, C. H.; Chang, T. C. J Polym Sci Chem 1991, 29, 361.
14. Cozan, V.; Butuc, E.; Stoleru, A.; Rusa, M.; Rusu, M.; Ni, Y. S.; Ding, M. X. J Macromol Sci Pure Appl Chem A 1995, 32, 1243.
15. Shukla, U.; Rao, K. V.; Rakshit, A. K. J Appl Polym Sci 2003, 88, 153.
16. Ragimov, A. V.; Mamedov, B. A.; Gasanova, S. G. Polym Int 1997, 43, 343.
17. Mamedov, A.; Vidadi, Y. A.; Alieva, D. N.; Ragimov, A. V. Polym Int 1997, 43, 126.
18. Vural, S.; Mart, H.; Demir, H. Ö.; Siros, O.; Muradoğlu, V.; Koc, M. C. Bull Chem Soc Ethiopia 2006, 20, 219.
19. Sahmetlioğlu, E.; Arıkan, U.; Toppare, L.; Yuruk, H.; Mart, H. J Macromol Sci Pure Appl Chem 2006, 43, 1523.
20. Mart, H. Des Monomers Polym 2006, 9, 551.
21. Banerjee, S.; Saxena, C. J Polym Sci Part A: Polym Chem 1996, 34, 3565.
22. Gaina, C.; Gaina, V.; Sava, M. High Perform Polym 2001, 13, 4.
23. Carter, K. R.; Hedrick, J. L. Macromolecules 1994, 27, 3426.
24. Issam, A. M.; Atto, A. T.; Sara, F. Y. Iraq Pat. 2508, 1993.
25. Buruiana, E. C.; Olaru, M.; Simionescu, B. C. Eur Polym J 2002, 38, 1079.
26. Tamareselvely, K.; Venkatarao, K.; Khothandaraman, H. Makromol Chem 1990, 191, 1231.
27. Tanaka, M.; Nakaya, T. Makromol Chem 1986, 187, 2345.
28. Tang, J. C.; Chang, T. C. Eur Polym J 1994, 30, 1059.
29. Stoica, G.; Stanciu, A.; Cozan, V.; Stoleriu, A.; Timpu, D. J Macromol Sci Pure Appl Chem 1998, 35, 539.
30. Reddy, K. R.; Raghu, A. V.; Jeong, H. M. Polym Bull 2008, 60, 609.
31. Bal, A.; Güçlü, G.; Acar, İ; İyim, T. B. Prog Org Coat 2010, 68, 363.
32. Singru, R. N.; Gurnule, W. B. J Term Anal Calorim 2010, 100, 1027.
33. Kaya, İ.; Yildirim, M. Synth Met 2009, 159, 1572.
34. Xiang, J.; Rondonuwu, F. S.; Kakitani, Y.; Fujii, R.; Watanabe, Y.; Koyama, Y. J Phys Chem B 2005, 109, 17066.
35. Issam, A. M.; İsmail, J. J Appl Polym Sci 2006, 100, 1198.
36. Kaya, İ.; Bilici, A. J Macromol Sci Pure Appl Chem 2006, 43, 719.
37. Pielichowski, K. Polym J 1997, 29, 848.
38. Lage, L. G.; Kawano, Y. J Appl Polym Sci 2001, 79, 910.
39. Lee, H. K.; Ko, S. W. J Appl Polym Sci 1993, 50, 1269.
40. Grassie, N.; Mendoza, G. A. P. Polym Degrad Stabil 1985, 11, 359.
41. Day, M.; Cooney, J. D.; MacKinnon, M. Polym Degrad Stabil 1995, 48, 341.
42. Zhang, Y.; Shang, S.; Zhang, X.; Wang, D.; Hourston, D. J. J Appl Polym Sci 1995, 58, 1803.
43. Chattopadhyay, D. K.; Webster, D. C. Prog Polym Sci 2009, 34, 1068.