Synthesis of Novel Azo-Containing Polyureas Derived from 4-[4′-(2-Hydroxy-1-naphthylazo)phenyl]-1,2,4-triazolidine-3,5-dione

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ABSTRACT: 4-[4′-(2-Hydroxy-1-naphthylazo)phenyl]-1,2,4-triazolidine-3,5-dione (**HNAPTD**) (**1**) has been reacted with excess amount of *n*-propylisocyanate in DMF (*N,N*-dimethylformamide) solution at room temperature. The reaction proceeded with high yield, and involved reaction of both N—H of the urazole group. The resulting bis-urea derivative **2** was characterized by IR, ^{1}H-NMR, elemental analysis, UV-Vis spectra, and it was finally used as a model compound for the polymerization reaction. Solution polycondensation reactions of monomer **1** with Hexamethylene diisocyanate (**HMDI**) and isophorone diisocyanate (**IPDI**) were performed in DMF in the presence of pyridine as a catalyst and lead to the formation of novel aliphatic azo-containing polyurea dyes, which are soluble in polar solvents. The polymerization reaction with tolylene-2,4-diisocyanate (**TDI**) gave novel aromatic polyurea dye, which is insoluble in most organic solvents. These novel polyureas have inherent viscosities in a range of 0.15–0.22 g dL^{-1} in DMF at 25°C. Some structural characterization and physical properties of these novel polymers are reported. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 3177–3183, 2001

Key words: polymeric dyes; polyurea; step-growth polymerization; inherent viscosity; 4-[4′-(2-Hydroxy-1-naphthylazo)phenyl]-1,2,4-triazolidine-3,5-dione

INTRODUCTION

Photoresponsive systems are systems that respond to light energy beyond merely absorbing and releasing it as other kinds of energy. Among the photoresponsive materials, azo compounds are of great interest, because of *cis* ⇌ *trans* isomerization about the diazo bond, which is responsible for the immediate optical properties observed, especially the absorption spectra.[1–5]

Photoresponsive polymers, especially azo-containing polymers, have attracted a great deal of interest for both fundamental and applied significance, particularly in view of the application as optoelectronic devices,[6] optical switches,[7] optical data storage,[7,8] halographic grating fabrication,[9] liquid crystals,[10] storage of the light energy (to reappear as fluorescence, phosphorescence, and chemical or even mechanical energy), membrane,[11] as a monolyer for Langmuir-Blodgett films,[12] drug delivery system,[13] colored coating,[14,15] colorant, particularly in textile,[16] and indicator.[17] This wide range of application is significantly due to *cis* ⇌ *trans* isomerization in the unique hindered media of azo polymers that could control the changes in chemical and physical

properties of azo polymers, i.e., conductivity, solubility, metal ion capture capability, membrane potential and permeability, surface wettability, and glass transition temperature,[11] also degradability by azo reductases,[13] wide-range shadow,[18] and color changing by pH.[17]

The use of monomeric dyes causes some disadvantages such toxicity,[19] releasing from and migration in dyed materials,[14,15] Tethering of these dyes in polymers could solve these problems.

Polymeric dyes are prepared by using either inert or reactive monomeric dyes. In the first case, the dye is dissolved in molten polymer and a physical blend of the macromolecule and dye is obtained. In the second case, the dye become part of the macromolecule,[20] as the main chain,[9,20,21] or side chain,[22–25] both via polymerization of monomeric dye,[9,21] or chemical modification of colorless polymers.[11,17,26,27] Polymerization of monomeric dyes could be performed via polycondensation of bifunctional dyes,[9,20,21] or polyaddition of vinyl containing dyes,[10,15,28] Concerted color formation polymerization is also reported.[29]

Recently we have been able to take advantage of acidic N—H in a compound, 1-methyl-2,5-bis(4-phenylurazoyl) pyrrole, and synthesis novel polymers via N-alkylation and N-acylation reactions.[30,31] Polymerization of compound 4-phenylurazole (**PHU**) with phosgene, terephthaloyl chloride, and epichlorohydrin has been reported to give insoluble polymers.[32] Polymerization of compound **PHU** with aliphatic diacid chlorides gave soluble polyamides with inherent viscosity of 0.39 dLg^{-1}.[33] Copolymerization of 4-cyclohexylurazole (**CHU**) with aliphatic diacid chlorides gave soluble polyamides.[34] We have also reported the reaction of **PHU** with diisocyanates.[35] The resulting novel polyureas have inherent viscosities in a range of 0.04–0.23 g/dL.

The aim of this investigation was to examine the step-growth polymerization reactions of **HNAPTD** as a monomer with diisocyanates. In the present article for the first time we wish to report on the successful polycondensation reaction where **HNAPTD** as a novel monomer is used for the synthesis of soluble novel polyurea dyes.

EXPERIMENTAL

Apparatus

Proton nuclear magnetic resonance (^{1}H-NMR, 90 MHz) spectra was recorded on a Varian EM-390 instrument. Tetramethylsilane (TMS) was used as an internal reference. IR spectra were recorded on Shimadzu 435 IR spectrophotometer. Spectra of solids were carried out using KBr pellets. Vibrational transition frequencies are reported in wave numbers (cm^{-1}). Band intensities are assigned as weak (w), medium (m), shoulder (sh), strong (s), and broad (br). Inherent viscosities were measured by a standard procedure using a Cannon Fensk Routine Viscometer (Germany). Thermal Gravimetric Analysis (TGA) data for polymers were taken on a Mettler TGA-4000 (Switzerland) in nitrogen atmosphere at a rate of 10°C/min. Elemental analyses were performed by Research Institute of petroleum Industry, Tehran, I.R. Iran.

Reagents and Monomer

Reagents were purchased from Fluka Chemical CO. (Buchs, Switzerland), Alderich Chemical CO. (Dorset, England), and Riedel-deHaen AG (Germany), and were used without purification. 4-[4'-(2-Hydroxy-1-naphthylazo)phenyl]-1,2,4-triazolidine-3,5-dione (**HNAPTD**) was synthesized according to our previous report.[36]

Preparation of 1,2-bis(n-Propylamidocarbonyl)-4-[4'-(2-Hydroxy-1-naphthylazo)phenyl]-1,2,4-triazolidine-3,5-dione (Model Compound 2)

In a 25-mL round-bottom flask, n-propylisocyanate (1 mL, 1.07 × 10^{-2} mol) was added to a solution of **HNAPTD** 1 (0.200 g, 5.76 × 10^{-4} mol) in 1 mL of DMF. The solution was stirred for 12 h at room temperature. The excess n-propylisocyanate was removed by aspirator and the residue was percipitated in 50 mL of diethyl ether. The resulting solid was filtered off, dried to give 0.26 g (87%) of red solid **2**. This red solid was recrystallized from water–acetone mixture, m.p. 258–260°C; IR(KBr): 3400–3200 (m, br), 2950 (m, sh), 1790 (w), 1740 (s), 1720 (m), 1620 (m), 1510 (s), 1450 (w), 1415 (s), 1255 (s, sh), 1205 (s), 1150 (m, sh), 1070 (w), 1045 (w), 1010 (w), 980 (w), 820 (m), 755 (m), 500 (w) cm^{-1}; ^{1}H-NMR (MDSO-d$_6$, TMS): δ 0.9 (t, 6H, J = 7.5 Hz), 1.5 (m, 4H), 3.2 (t, 4H, J = 7.5 Hz), 3.8 (s, br, 2H), 6.9 (dd, 1H, J_1 = 12.0 Hz, J_2 = 3.0 Hz), 7.75 (m, 8H), 8.6 (d, 1H, J = 9.0 Hz), 15.8 (s, 1H). Anal. Calcd for C$_{26}$H$_{27}$N$_7$O$_5$ C, 60.34%; H, 5.26%; N, 18.94%. Found: C, 59.70%; H, 5.00%; N, 18.00%.

Polymerization of HNAPTD with HMDI

In a 25-mL round-bottom flask **HNAPTD** (0.0956 g, 2.75 × 10^{-4} mol) was added to a solution of

HMDI (**3**) (0.0463 g, 2.75 × 10⁻⁴ mol) and pyridine (0.5 mL, 6.45 × 10⁻⁴ mol) in 0.5 mL of DMF. The solution was stirred for 24 h at room temperature, then for 24 h at 80°C, and finally 24 h at 105°C. The viscous solution was precipitated in 50 mL of methanol. The solid was filtered off, dried to give 0.12 g (83%) of red polyurea **PU1.** m.p. >205°C; inherent viscosity (0.5 g/dL DMF, 25°C) = 0.22; IR(KBr): 3400 (m), 2900 (m), 1780 (w), 1730 (s), 1700 (s), 1620 (m), 1550 (m), 1505 (s), 1410 (s, br), 1350 (m), 1205 (m), 1140 (m), 980 (w), 830 (m), 850 (m), 505 (w) cm⁻¹. ¹H-NMR (DMSO, TMS): δ 1.4 (m, br, 4H), 3.3 (m, 4H), 4.4 (m, br, 6H), 6.95 (d, 1H, J = 12.0 Hz), 7.8 (m, 8H), 8.7 (d, 1H, J = 9.0 Hz), 15.9 (s, 1H). Anal. Calcd for $C_{26}H_{25}N_7O_5$: C, 60.58%; H, 4.89%; N, 19.02%. Found: C, 59.50%; H, 4.90%; N, 18.60%. Thermal analysis: T_5 242°C, T_{10} 258°C, char yield at 600°C: 28.0%.

Polymerization of HNAPTD with IPDI

In a 25-mL round-bottom flask **HNAPTD** (0.0912 g, 2.63 × 10⁻⁴ mol) was added to a solution of **IPDI** (**4**) (0.0584 g, 2.63 × 10⁻⁴ mol) and pyridine (0.05 mL, 6.45 × 10⁻⁴ mol) in 0.5 mL of DMF. The solution was stirred for 24 h at room temperature, then for 24 h at 80°C, then 1 mL of DMF was added and the mixture was stirred for 24 h at 105°C. The solution was precipitated in 100 mL of methanol/water (75/25) mixture. The solid was filtered off, dried to give 0.12 g (82%) of red polyurea **PU2,** m.p. >225°C; inherent viscosity (0.5 g/dL DMF, 25°C) = 0.19; IR(KBr): 3300 (m), 2950 (m, sh), 1780 (m), 1730 (s), 1710 (s), 1620 (m), 1550 (m), 1510 (s), 1415 (m, br), 1305 (w, br), 1250 (m), 1230 (m), 1205 (m), 1145 (m), 980 (w), 830 (m), 755 (m) cm⁻¹. Anal. Calcd for $C_{30}H_{31}N_7O_5$: C, 63.26%; H, 5.49%; N, 17.21%. Found: C, 54.40%; H, 4.70%; N, 14.10%. Corrected for 14% moisture intake: C, 63.26%; H, 5.47%; N, 16.40%. Thermal analysis: T_5 233°C, T_{10} 262°C, char yield at 600°C: 24.0%.

Polymerization of HNAPTD with TDI

In a 25-mL round-bottom flask **HNAPTD** (0.2553 g, 7.35 × 10⁻⁴ mol) was added to a solution of **TDI** (**5**) (0.1280 g, 7.35 × 10⁻⁴ mol) and pyridine (0.15 mL, 1.86 × 10⁻³ mol) in 1 mL of DMF. The red solid was precipitated immediately. One milliliter of DMF was added, and stirring was continued for 24 h at 110°C. The solution was precipitated in 100 mL of methanol. The solid was filtered off,

dried to give 0.28 g (73%) of red polyurea **PU3,** m.p. >290°C; inherent viscosity (0.5 g/dL DMF, 25°C) = 0.15; IR(KBr): 3300 (s, br), 1800 (w, br), 1700 (s, br), 1640 (w), 1620 (w), 1600 (w), 1500 (s), 1410 (m, br), 1300 (w), 1200 (m), 1140 (w), 980 (w), 830 (m), 750 (m) cm⁻¹. ¹H-NMR (DMSO, TMS): δ 2.3 (s, 3H), 3.0 (s, 2H), 7.0–8.2 (m, 12H), 8.8 (m, br, 1H), 16.0 (s, 1H). Anal. Calcd for $C_{27}H_{19}N_7O_5$: C, 62.19%; H, 3.67%; N, 18.80%. Found: C, 58.20%; H, 4.40%; N, 17.00%. Corrected for 7% moisture intake: C, 62.58%; H, 3.95%; N, 18.28%. Thermal analysis: T_5 217°C, T_{10} 262°C, char yield at 600°C: 39.5%.

RESULTS AND DISCUSSION

Model Compound Studies

HNAPTD (**1**) was allowed to react with excess *n*-propylisocyanate in DMF solution, and gave 1,2-bis(*n*-propylamidocarbonyl)-1-[4′-(1,2,4-triazolidine-3,5-dione-4-yl)phenylazo]-2-naphthol (**2**) in high yield (Scheme 1). The compound (**2**) was characterized by IR, ¹H-NMR, elemental analysis, and UV-Vis spectra. The IR spectrum of (**2**) showed two peaks at 1790 and 1740 cm⁻¹ for the carbonyl groups. These are characteristic patterns for the urazole moiety. The ¹H-NMR spectrum of **2** showed a broad mutipilet at 3.8 ppm for the two NH groups, a triplet at 3.2 ppm for the two methylene groups attached to the amide groups, a multiplet at 1.5 ppm for the other two methylene groups attached to the methyl groups, a triplet at 0.9 ppm for the two methyl groups, a multiplet at 6.9–8.6 ppm for aromatic protons, and a singlet at 15.8 ppm for hydroxyl group of β-naphthol moiety. It is interesting to mention that the OH group on naphthyl system does not react with excess *n*-propylisocyanate. This could be explained in terms of steric hindrance and intraomolecular H-bonding of OH group with nitrogen atom of the azo group, which does not make the OH group readily available for the reaction with *n*-propylisocyanate.
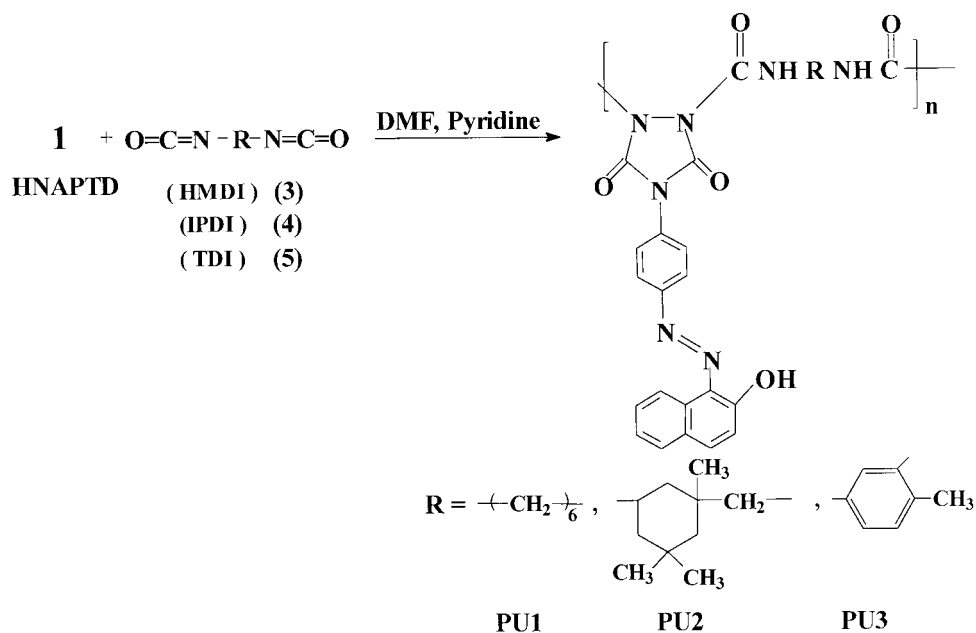
Polymerization Reactions

Because 1,2-bis(*n*-propylamidocarbonyl)-4-[4′-(2-Hydroxy-1-naphthylazo)phenyl]-1,2,4-triazolidine-3,5-dione (**2**) as a model compound was synthesized in high yield and purity, we became interested to perform this type the reaction for the formation of the novel azo-containing polyureas.

1
HNAPTD

+   CH$_3$CH$_2$CH$_2$N = C = O
Excess

DMF

CH$_3$CH$_2$CH$_2$NH C ... C NHCH$_2$CH$_2$CH$_3$

2
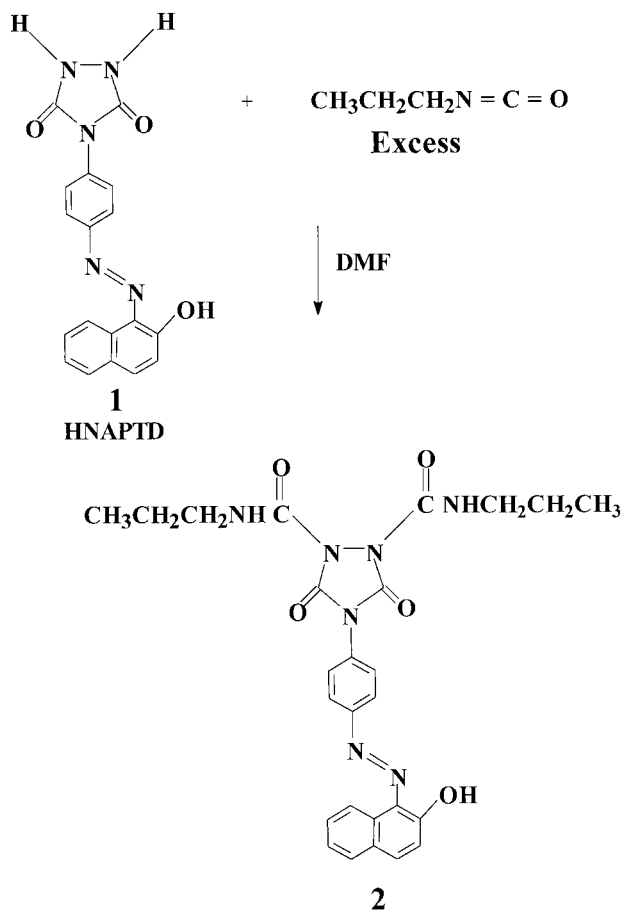
Scheme 1

Thus **HMDI** (**3**), **IPDI** (**4**), and **TDI** (**5**) were selected as diisocyanates. The reaction of monomer **1** with these diisocyanates was performed via solution polymerization. The reactions were carried out in DMF solution in the presence of pyridine as a catalyst and the resulted polyureas **PU1–PU3** were obtained as red solids in high yield (Scheme 2). The resulting polymers were characterized by IR, ^{1}H-NMR, elemental analysis, UV-Vis, and TGA. The IR spectrum of polymer **PU1** showed two peaks at 1780 and 1730 cm^{-1} for the carbonyl groups, which are characteristic patterns for the urazole moiety; also a strong peak at 1700 cm^{-1} for the urea carbonyl group. The ^{1}H-NMR spectrum of polymer **PU1** showed a broad multiplet peak at 4.4 ppm for two NH and two methylene groups attached to the NH groups. Broad multiplet peaks at 3.3 and 1.4 ppm are related to the other four methylene groups attached to the N—H group, respectively. Also, a singlet at 15.9 ppm is related to the hydroxyl group of β-naphthol moeity. The elemental analysis results are also in good agreement with calculated percentages for carbon, hydrogen, and nitrogen contents in the polymer repeating unit. The IR spectrum of polymer **PU2** showed two strong peaks at 1780 and 1730 cm^{-1} for the carbonyl groups, which are related to the urazole moiety, and a strong peak at 1710 cm^{-1} for the urea carbonyl group. This polyurea absorb moisture; therefore, the elemen-
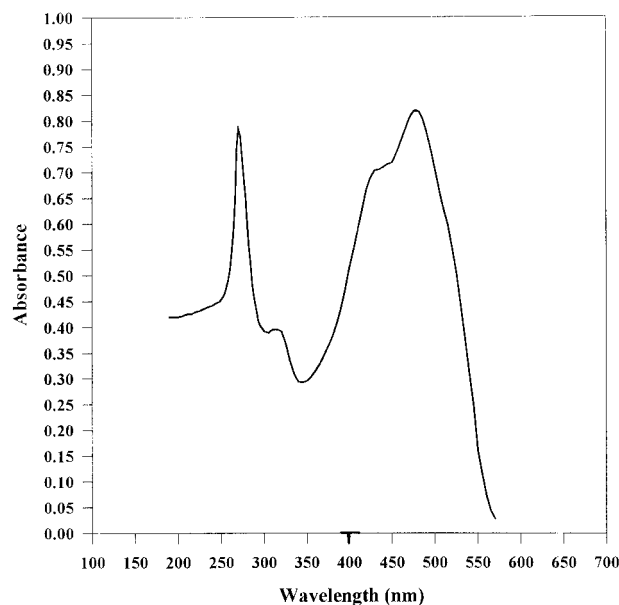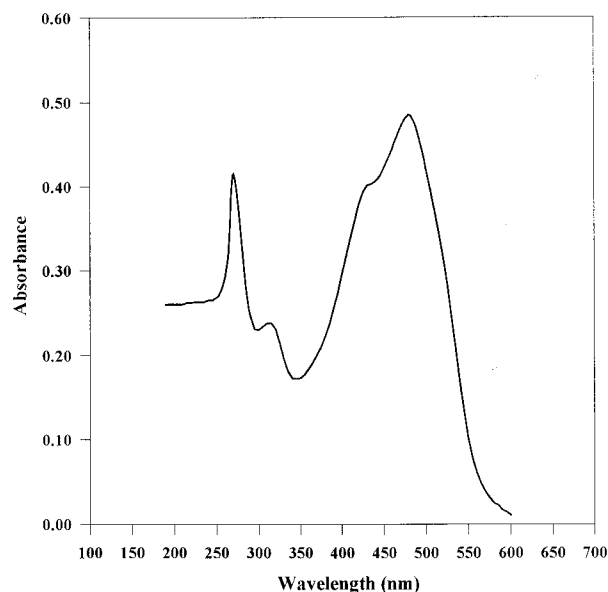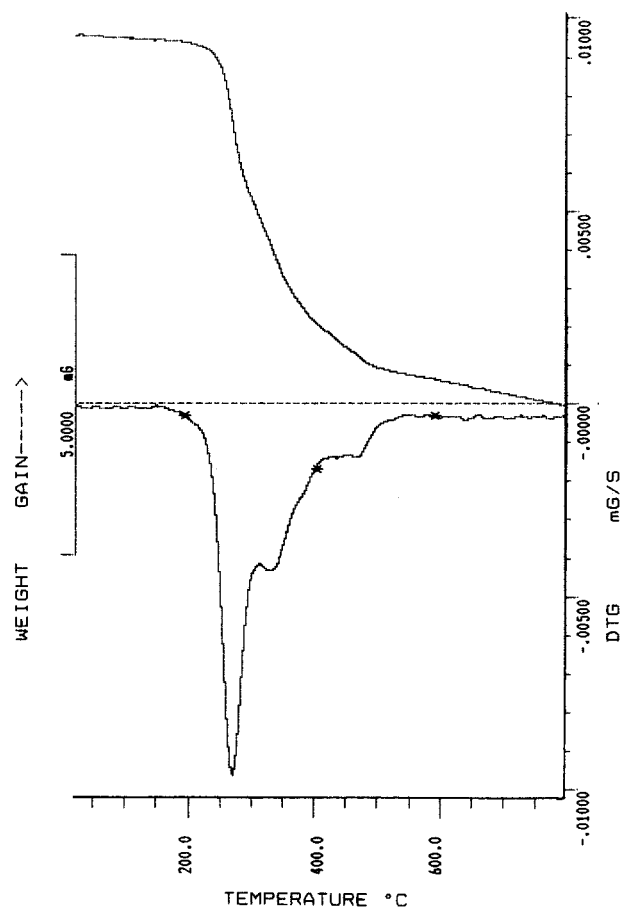
1   + O=C=N – R–N=C=O
HNAPTD
（HMDI）(**3**)
(IPDI) (**4**)
(TDI) (**5**)

DMF, Pyridine →

C NH R NH C
... n

R = ⫴CH$_2$⫴$_6$,   CH$_3$ ... CH$_2$— ,   — CH$_3$
CH$_3$ CH$_3$

PU1   **PU2**   **PU3**

Scheme 2

Table I   UV-Vis Spectra of Model Compound 2 and Polymers PU1–PU3 in DMF

Code	Color	λ_{max}(A, ε M^{-1} cm^{-1})
Compound 2	Orange	270 (0.780,18,353)
		314 (0.400, 9,412)
		430 (0.700,16,471)
		480 (0.840,19,765)
PU1	Red	270 (0.415,16,468)
		313 (0.240, 9,524)
		430 (0.400,15,873)
		480 (0.485,19,246)
PU2	Red	270 (0.425,12,143)
		314 (0.250, 7,143)
		430 (0.385,11,000)
		479 (0.455,13,000)
PU3	Red	272 (1.210,31,510)
		434 (0.460,11,979)
		482 (0.556,14,479)

Figure 2   UV-Vis spectrum of **PU1** in DMF.

tal analysis results are required to make correction relative to moisture intake. The corrected elemental analysis results for 14% water content are in good agreement with calculated percentages for carbon, hydrogen, and nitrogen contents in the polymer repeating unit.

The IR spectrum of polymer **PU3** showed two strong peaks at 1800 and 1740 cm^{-1} (which appears as a shoulder on the strong peak at 1700 cm^{-1}) for the carbonyl groups, which are related

Figure 1   UV-Vis spectrum of model compound **2** in DMF.

Figure 3   TGA and DTG thermograms of polymer **PU1** under nitrogen atmosphere.
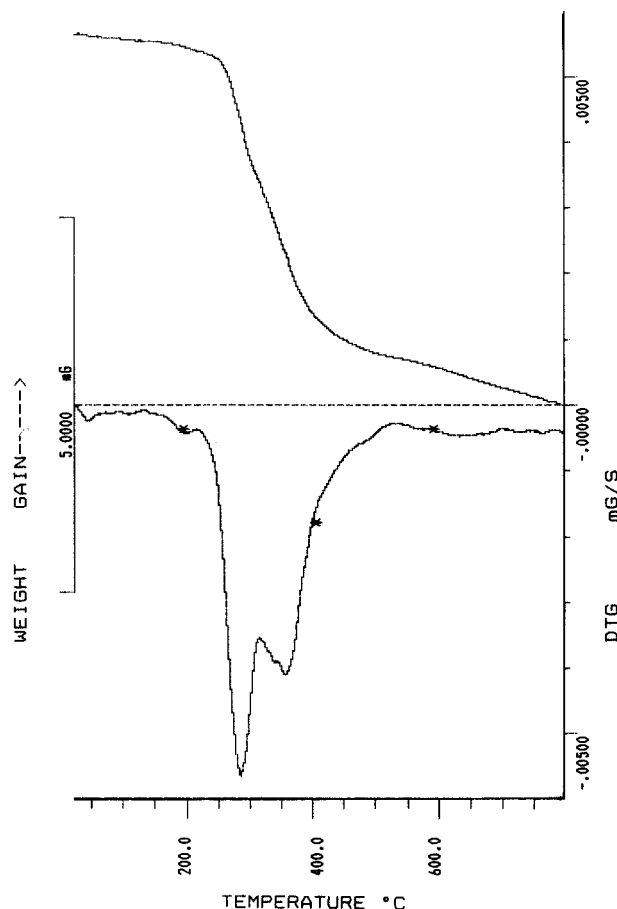
Figure 4 TGA and DTG thermogram of polymer **PU2** under nitrogen atmosphere.

to urazole moiety, and also a strong peak at 1700 cm^{-1} for the urae carbonyl group. The ^{1}H-NMR spectrum of polymer **PU3** showed a singlet at 3.0 ppm for two NH groups, a singlet at 2.3 ppm for the tolulene methyl group, a multiplet at 7.0–8.2 ppm, and a broad peak at 8.8 ppm for aromatic hydrogenes. A singlet at 16.0 ppm is related to the hydroxyl group of β-naphthol moiety. The corrected elemental analysis results for 7% moisture content are in good agreement with calculated percentages for carbon, hydrogen, and nitrogen contents in the polymer repeating unit. It is interesting to note that the water molecule may incorporate through H-bonding between two molecules via the azo group in the polymer matrix. We have observed such a phenomena in our previous work.[36]

The UV-Vis spectra of the azo dye model compound **2** and polyureas **PU1, PU2,** and **PU3** were recorded in DMF, and the data are shown in Table I. It is apparent that the wavelength of

maximum absorptions are related to the azo groups in the compounds, and they are observed within 311–468 nm (Figs. 1–2). All of these compounds show almost similar UV-Vis spectra pattern.

The polymers **PU1** and **PU2** are soluble in organic solvents such as DMF, dioxane, DMSO, DMAc, and Acetic acid, and are insoluble in solvents such as water, methanol, acetone, cyclohexane, and chloroform. The polymer **PU3** is soluble in DMAc and DMSO, but in insoluble in most organic solvents.

Thermal Properties

The thermal behavior of polyureas **PU1, PU2,** and **PU3** were measured by thermaogravimetric analysis (TGA) at a rate of 10°C/min in nitrogen atmosphere (Figs. 3–5). An examination of the data reveals that all of the above polyureas are thermally stable up to 200°C in nitrogen atmo-
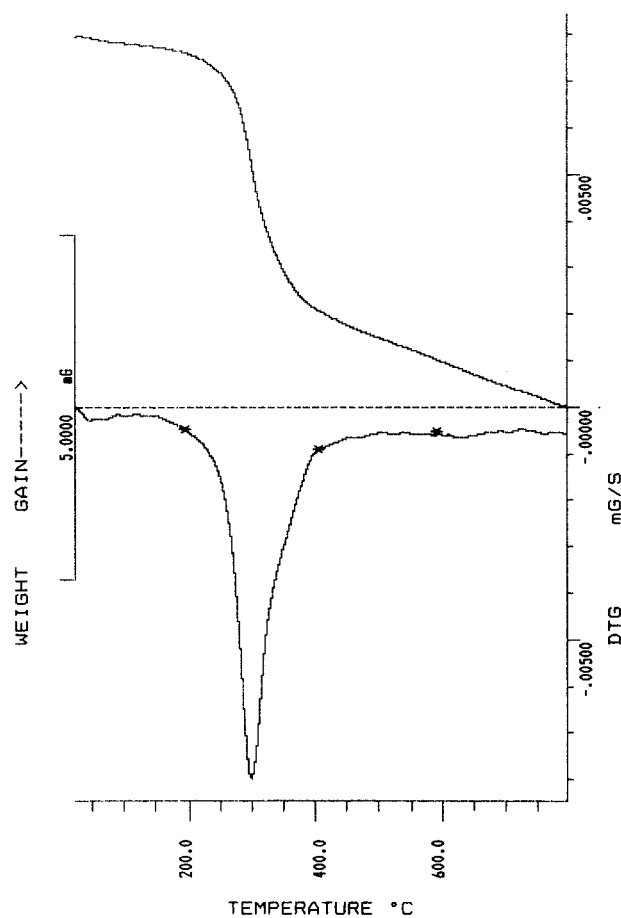

Figure 5 TGA and DTG thermogram of polymer **PU3** under nitrogen atmosphere.

sphere. The polymers **PU1, PU2,** and **PU3** show 5% weight loss at 242, 233, and 217°C, respectively.

CONCLUSIONS

This investigation has shown that **HNAPTD** (**1**) is an interesting monomer for the polycondensation reactions. This compound has two acidic N—H groups, and it can be readily reacted with *n*-propylisocyanate. This reaction gives only N—H substitution adduct in quantitative yield and high purity. Thus, compound (**1**) can act as a bifunctional monomer, and its polymerization reaction with aliphatic and aromatic diisocyanates gave novel polyureas with azonaphthalene containing urazole linkages having inherent viscosities of 0.15–0.22 dL/g, which roughly correspond to molecular weight of 10,000 to 15,000 g/mol. This is the first report on the synthesis of polymeric dyes containing urazole linkages. Because these polyureas have an azo functional group, they have potential to be used as photoresponsive materials.

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