

Thermally Stable and Organo-soluble Polyamides Containing Triazine Rings and Ether Linkages in the Main Chain: Synthesis and Characterization

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Abstract In order to synthesize new polyamide (PA) **5** and copolyamides (CoPA) **7a–7d**, a new-type of dicarboxylic acid containing triazine ring was successfully synthesized from 2,4-dichloro-6-methoxy-s-triazine **1** and 4-aminobenzoic acid. New polyamide **5** containing triazine moiety was prepared from 4,4'-(6-methoxy-1,3,5-triazine-2,4-diyl)bis(azanediyldibenzoic acid **3** and 4,4'-diaminodiphenylether by direct polycondensation reaction. Then new series thermally stable copolyamides **7a–7d** with high solubility in common organic solvents were synthesized from the direct polycondensation reaction of diacid **3** as a first monomer, aliphatic-aromatic diacids **6a–6d** as second diacids and 4,4'-diaminodiphenylether. All of the above polymers were fully characterized by ¹H-NMR and FTIR spectroscopy, elemental analysis (CHN), inherent viscosity, solubility tests, gel permeation chromatography (GPC), differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). The resulted CoPAs **7a–7d** have shown good inherent viscosities, solubility and thermal properties.

Keywords: Polyamide; Thermally stable; Organo-soluble; Polycondensation; Triazine.

INTRODUCTION

Polyamides are well recognized as a class of commercially significant thermally stable polymers^[1]. The ordering and varying of backbone functions have profound effects on the final properties such as solubility and thermal characteristics of the heterocyclic polymers. It has been generally recognized that the introduction of ether groups into the polymer chains improves the solubility of heterocyclic polymers while maintaining the thermal stability^[2].

Aromatic polymers that contain aryl ether linkages generally have lower glass transition temperatures, greater chain flexibility, and tractability in compared to their corresponding polymers without these groups in the chain^[3, 4]. The lower glass transition temperatures and also improved solubility are attributed to the flexible linkages that provide a polymer chain with a lower energy of internal rotation^[5].

The introduction of ether groups into polymers such as poly(amide-imide)^[6–10], poly(amide-ester)^[11], polycarbonates^[12] *etc.* endows them with specific properties: a combination of enhanced heat resistance with excellent solubility which is particularly important for rigid aromatic polymers.

Over the past few decades, the increasing and constant requirement of the automotive, aerospace, microelectronic and military industries has provided the driving force for the development of new high

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performance polymers to be used in structural applications. Most frequently, much effort has been paid to the development of aromatic heterocyclic polymers for use as advanced composite matrix, structural adhesives or coatings in high-temperature applications^[13–15]. Among this category of polymers, triazine-containing polymers are drawing much attention for their more excellent thermal resistance, higher tensile strengths and modulus at elevated temperatures, relative to the conventional polymers^[16–18]. Introduction of triazine moiety and the flexibilizing linkages into the main chain of aromatic polyamides afforded polymers with good solubility and high thermal stability. Thus, these polymers can be considered as promising, processable, high-temperature resistant polymer materials^[19, 20].

A triphenyl phosphite activated polycondensation (phosphorylation reaction) technique for the synthesis of polyamides was reported by Yamazaki *et al.*^[21]. We describe here the preparation and basic characterization of a series of thermally stable and organosoluble copolyamides **7a–7d** based on triazine ring as a heterocyclic segment and ether linkages as a flexible group from the reaction of 4,4'-(6-methoxy-1,3,5-triazine-2,4-diyl)bis(oxy)dibenzoic acid **3** with aliphatic-aromatic diacids **6a–6d** and 4,4'-diaminodiphenylether in a medium consisting of *N*-methyl-2-pyrrolidone (NMP)/triphenyl phosphite (TPP)/calcium chloride (CaCl₂)/pyridine (Py) system.

EXPERIMENTAL

Materials

Cyanuric chloride (Merck) was recrystallized in hot carbon tetrachloride. 2,4-Dichloro-6-methoxy-s-triazine was synthesized according to the previous literature^[19, 20], 4-aminobenzoic acid, 4,4'-diaminodiphenylether (Merck) were used without further purification. Solvents: *N*-methyl-2-pyrrolidone (NMP, Fluka), pyridine (Acros), triphenyl phosphite (TPP, Merck) and *N,N*-dimethylformamide (DMF, Merck) were used as received. Commercially available calcium chloride (CaCl₂, Merck) was dried under vacuum at 150 °C for 6 h.

Techniques

¹H-NMR and ¹³C-NMR spectra were recorded on a Bruker 300 MHz instrument (Germany) and a Bruker 250 MHz instrument DPX300. Fourier transform infrared (FTIR) spectra were recorded on a Galaxy series FTIR 5000 spectrophotometer (England). Spectra of solid were performed by using KBr pellets. Vibration transition frequencies were reported in wavenumber (cm⁻¹). Band intensities were assigned as weak (w), medium (m), shoulder (sh), strong (s) and broad (br). Inherent viscosities were measured with a standard procedure by using a Technico Regd Trad Mark Viscometer. Weight-average (*M_w*) and number-average (*M_n*) molecular weights were determined by gel permeation chromatography (GPC). The eluents were monitored with a UV detector (JMST Systems, USA, VUV-24) at 254 nm. Poly(vinylpyrrolidone) was used as the standard. Thermal gravimetric analysis (TGA) data for polymers were taken on a Mettler TA4000 System under N₂ atmosphere at a rate of 10 K/min and differential scanning calorimeter (DSC) was conducted with a DSC Mettler 110 (Switzerland) at a heating rate of 10 K/min in a nitrogen atmosphere. Elemental analyses were performed by using a Vario EL equipment.

Monomer Synthesis

Synthesis of 4,4'-(6-methoxy-1,3,5-triazine-2,4-diyl)bis(azanediyldibenzoic acid **3**

To a solution of 6-methoxy-2,4-dichlorotriazine **1** (10.84 mmol) in acetone (50 mL), a mixture of sodium bicarbonate 0.5 g, and 4-aminobenzoic acid **2** (21.69 mmol) in distilled water (50 mL) was added slowly at 5–10 °C with continuous stirring, and the reaction mixture was heated under reflux in an oil bath for 5 h. After that the white precipitate was filtered and dried in room temperature for overnight (yield: 84%). M.P., 356–358 °C. FTIR (KBr): 2600–3700 (m, br), 1707 (s), 1512 (w), 1469 (w), 1388 (m), 1309 (w), 1195 (m), 1068 (m), 788 (m) cm⁻¹. ¹H-NMR (250 MHz, DMSO, δ): 13 (s, 1H), 10.14 (s, 1H), 7.9–8 (d, 2H), 7.24–7.28 (d, 2H), 3.8 (s, 1H). ¹³C-NMR (300 MHz, DMSO, δ): 187.9, 152.2, 142.9, 131.0, 121.0, 120.7, 114.1, 54.2. Elemental Analysis for C₁₅H₁₅N₅O₅ (%): calculated: C, 56.69; H, 3.96; N, 18.37; found: C, 56.61; H, 3.92; N, 18.26.

Polymer Syntheses

Synthesis of polyamide 5

A 50 mL round-bottom flask fitted with a stirring bar was filled with 4,4'-(6-methoxy-1,3,5-triazine-2,4-diyl)bis(azanediyldibenzoic acid **3** (14 mmol), 4,4'-diaminodiphenylether **4** (14 mmol), calcium chloride (0.5 g, 4.5 mmol), triphenyl phosphite (8.7 mL, 28 mmol), pyridine (1 mL) and *N*-methyl-2-pyrrolidone (8 mL). The reaction mixture was heated under reflux in an oil bath at 60 °C for 1 h, 90 °C for 3 h, and 120 °C for 8 h. Then it was poured into 100 mL of methanol and the precipitated product was collected by filtration and washed thoroughly with hot methanol. Finally, the product was dried at 80 °C for 12 h in a vacuum oven until the constant weight. FTIR (KBr): 3348 (w), 1709 (s), 1610 (w), 1516 (m), 1456 (w), 1383 (m), 1309 (m), 1238 (w), 1174 (m), 937 (w), 744 (w), 698 (w) cm^{-1} . $^1\text{H-NMR}$ (250 MHz, DMSO, δ): 11 (s, 1H), 10.33 (s, 1H), 8.28 (d, 2H), 7.99 (d, 2H), 7.41 (d, 2H), 7.11 (d, 2H), 3.95 (s, 1.5 H). Elemental analysis for $\text{C}_{30}\text{H}_{23}\text{N}_7\text{O}_4$ (%): calculated: C, 66.05; H, 4.25; N, 17.97, found: C, 65.85; H, 4.21; N, 17.89.

Synthesis of copolyamides 7a–7d

Into a 100 mL round bottomed flask was placed a mixture of 4,4'-(6-methoxy-1,3,5-triazine-2,4-diyl)bis(azanediyldibenzoic acid **3** (0.326 mmol), diacids **6a–6d** (0.326 mmol), 4,4'-diaminodiphenylether **4** (0.652 mmol), 0.10 g of calcium chloride, 0.84 mL of triphenyl phosphite, 0.2 mL of pyridine and 1.8 mL NMP. The mixture was heated for 1 h at 60 °C, 2 h at 90 °C and then refluxed at 120 °C for 7 h until a viscose solution was formed. Then it was cooled to room temperature and 30 mL of methanol was added to the reaction mixture. The precipitate was formed, filtered off and washed with methanol. The resulting polymers **7a–7d** were dried under vacuum.

Copolyamide **7a**: FTIR (KBr): 3061 (w), 3034 (w), 2933 (w), 1709 (s), 1591 (s), 1525 (m), 1496 (m, sh), 1381 (m), 1315 (m), 1249 (w), 1149 (m), 1105 (m), 696 (w) cm^{-1} . Elemental analysis for $\text{C}_{61}\text{H}_{47}\text{N}_9\text{O}_9$ (%), calculated: C, 69.77; H, 4.51; N, 12.00; found: C, 68.71; H, 4.45; N, 11.54.

Copolyamide **7b**: FTIR (KBr): 3306 (w), 2930 (w), 1707 (s), 1604 (w, sh), 1533 (m), 1494 (m), 1377 (m, br), 1234 (w), 1170 (m), 1030 (w), 931 (w), 758 (m), 696 (m) cm^{-1} . Elemental analysis for $\text{C}_{62}\text{H}_{49}\text{N}_9\text{O}_9$ (%), calculated: C, 69.96; H, 4.64; N, 11.85; found: C, 68.85; H, 4.51, N, 11.79.

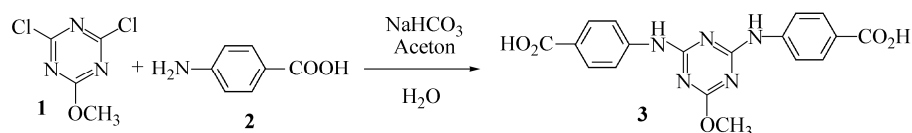
Copolyamide **7c**: FTIR (KBr): 3348 (w), 1710 (s), 1604 (w), 1498 (s), 1383 (m), 1309 (w), 1222 (m), 1170 (w, sh), 744 (w), 698 (w) cm^{-1} . Elemental analysis for $\text{C}_{63}\text{H}_{51}\text{N}_9\text{O}_9$ (%), calculated: C, 70.18; H, 4.77; N, 11.69; found: C, 60.12; H, 4.23; N, 11.41.

Copolyamide **7d**: FTIR (KBr): 3315 (w), 2901 (w), 1710 (s), 1664 (s, sh), 1595 (m), 1537 (m), 1479 (m), 1383 (m), 1303 (m), 1244 (m), 1151 (m), 1097 (w), 746 (w), 696 (m) cm^{-1} . Elemental Analysis for $\text{C}_{64}\text{H}_{53}\text{N}_9\text{O}_9$ (%), calculated: C, 70.38; H, 4.89; N, 11.54; found: C, 69.54; H, 4.56; N, 11.41.

RESULTS AND DISCUSSION

Synthesis of Monomer 4,4'-(6-Methoxy-1,3,5-triazine-2,4-diyl)bis(azanediyldibenzoic Acid **3**

Dicarboxylic acid **3** containing triazine ring was synthesized according to the reported procedure^[19, 20]. In order to synthesize thermally stable polyamides diacid **3** containing triazine ring as a heterocyclic unit was synthesized from one mole 6-methoxy-2,4-dichlorotriazine **1** and two mole of 4-aminobenzoic acid **2** in a solution of sodium bicarbonate (Scheme 1).



Scheme 1 Synthesis route of dicarboxylic acid **3**

The chemical structure and purity of dicarboxylic acid **3** was confirmed with elemental analysis, FTIR, ^1H -NMR, and ^{13}C -NMR spectroscopy. The measured results in elemental analyses of this compound were closely corresponded to the calculated ones, demonstrating that the expected compound was obtained.

The FTIR spectrum of dicarboxylic acid **3** shows a broad peak at $2600\text{--}3600\text{ cm}^{-1}$, which was assigned to the OH related to carboxylic acid groups in this compound, and a strong peak at 1707 cm^{-1} related to carbonyl in carboxylic groups (Fig. 1a). Also ^1H -NMR spectrum of dicarboxylic acid **3** showed doublet of doublet peaks around $\delta = 7.24$ and 7.97 assigned to the H(d) and H(c) related to aromatic protons and a singlet peak at $\delta = 10.14$ was assigned to the H(b) related to NH groups in this compound. A broad singlet peak around $\delta = 13.01$ was related to carboxylic acid protons (Fig. 2). Also ^{13}C -NMR spectrum of dicarboxylic acid **3** showed eight different carbon atoms (Fig. 3).

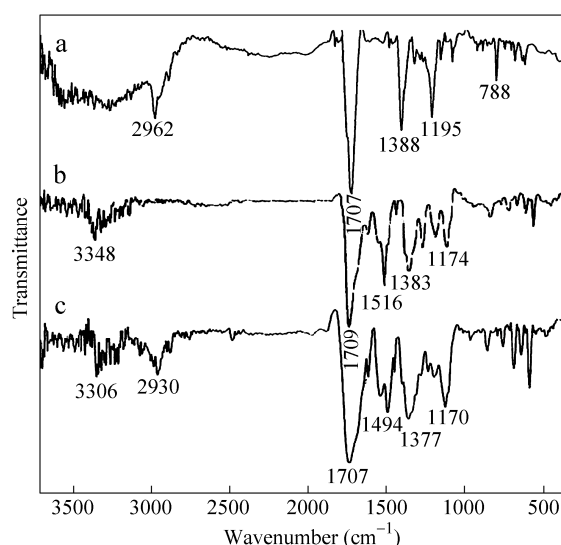


Fig. 1 FTIR spectra of (a) dicarboxylic acid **3**, (b) polyamide **5** and (c) copolyamide **7b**

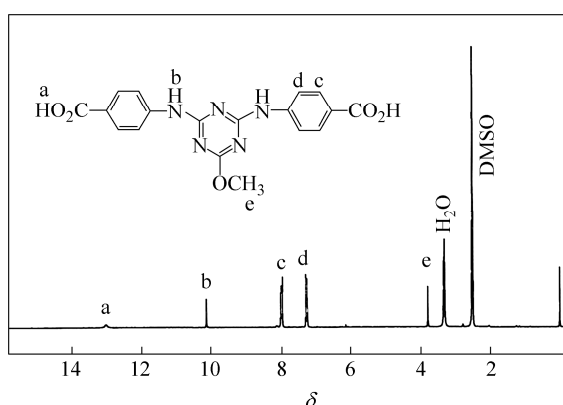


Fig. 2 ^1H -NMR spectrum of dicarboxylic acid **3**

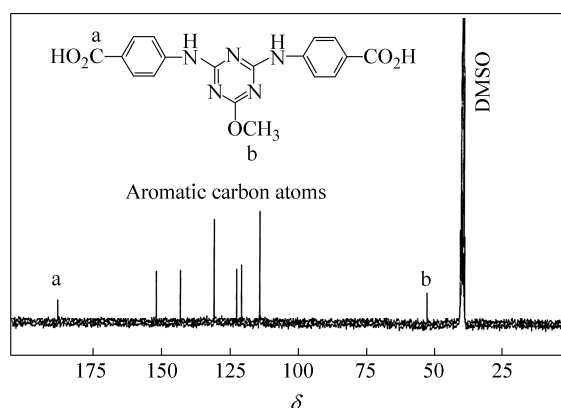
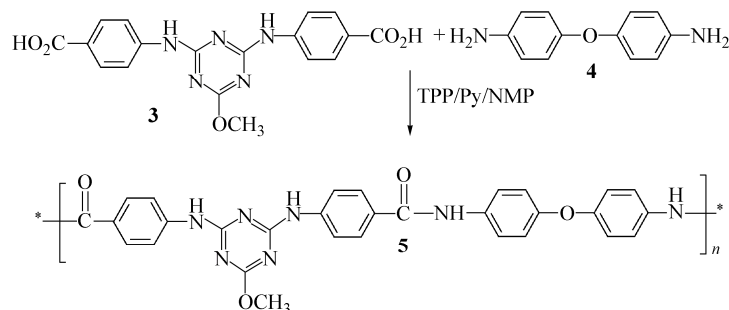


Fig. 3 ^{13}C -NMR spectrum of dicarboxylic acid **3**

Synthesis of Polymers

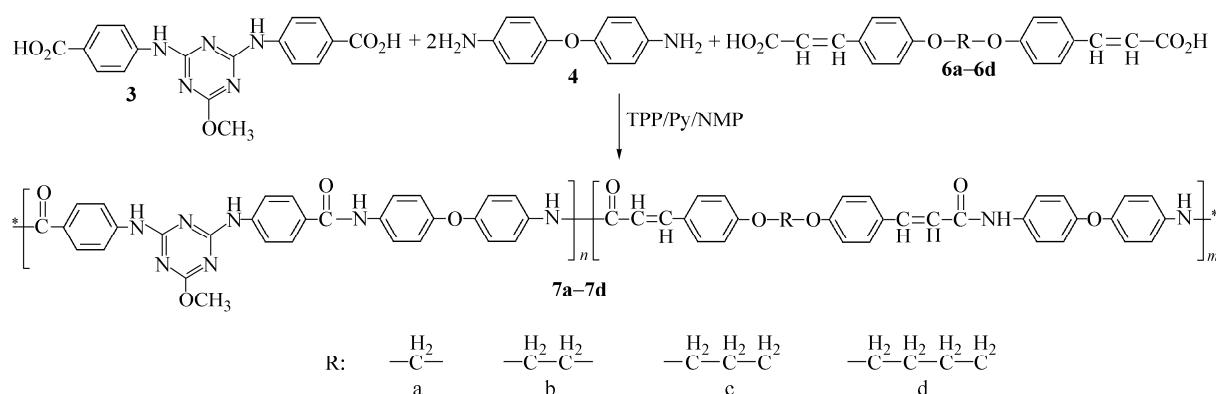
We synthesized a new polyamide **5** containing triazine ring as heterocyclic segment in the main chain as a model. A triphenyl phosphite (TPP)-activated polycondensation (phosphorylation reaction) technique for the synthesis of polyamides was reported by Yamazaki *et al.*^[21] Polyamide **5** was synthesized from the reaction of diacid **3** as a new diacid monomer with 4,4'-diaminodiphenylether **4** by direct polycondensation in a medium

consisting of *N*-methyl-2-pyrrolidone (NMP)/triphenyl phosphite (TPP)/calcium chloride (CaCl₂)/pyridine (Py) system (Scheme 2).



Scheme 2 Synthesis of polyamide 5

The main objective of our work is synthesis and characterization of four aliphatic-aromatic copolyamides **7a–7d** containing triazine ring and ether linkage and comparison their properties such as solubility and thermal stability with those of polyamide **5**. In order to synthesis copolyamides **7a–7d**, we used from aliphatic aromatic diacids **6a–6d**, as second diacid, containing ether linkages which were synthesized according to our previous works^[22–24]. Copolyamides **7a–7d** were synthesized from diacid **3** as the first diacid monomer, diacids **6a–6d** as the second diacids monomers and 4,4'-diaminodiphenylether **4** by direct polycondensation in a medium consisting of *N*-methyl-2-pyrrolidone (NMP)/triphenyl phosphite (TPP)/calcium chloride (CaCl₂)/pyridine (Py) system (Scheme 3).



Scheme 3 Synthesis of copolyamides **7a–7d**

The chemical structures and purities of all polymers were confirmed with elemental analysis, FTIR and ¹H-NMR spectroscopy. The measured results in elemental analyses of these compounds were closely corresponded to the calculated ones, demonstrating that the expected compounds were obtained. Figure 1(b) displays the FTIR spectrum of polyamide **5**. A peak at 3348 cm⁻¹ is related to amidic NH groups in polymer backbone and strong peaks at 1709 cm⁻¹ is related to carbonyls in amide groups. Also ¹H-NMR spectrum of polyamide **5** showed two doublet of doublet peaks in aromatic region related to aromatic protons and two singlet peaks at δ = 11.0 and 10.33 were assigned to the H(a) and H(b) related to NH groups in this compound. A peak at δ = 3.98 is related to methoxy's protons (Fig. 4).

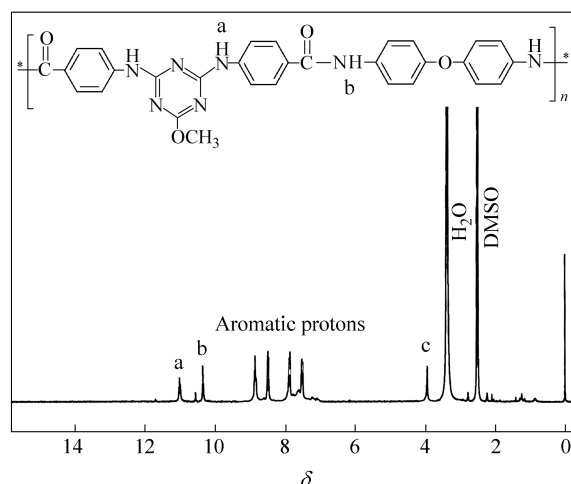


Fig. 4 ^1H -NMR spectrum of polyamide **5**

Figure 1(c) displays the FTIR spectrum of copolyamide **7b**. A peak at 3306 cm^{-1} is related to amidic NH groups in polymer backbone and a strong peak at 1707 cm^{-1} is related to carbonyls in amide groups. Also ^1H -NMR spectrum of copolyamide **7b** showed three singlet peaks at $\delta = 10.18$, 10.50 and 10.82 were assigned to the H(a), H(b) and H(b) related to NH groups (aromatic amine and two kinds amide groups) in this compound. Two peaks appeared at $\delta = 4.15$ and 4.41 are related to methyl and methylene groups, respectively (Fig. 5).

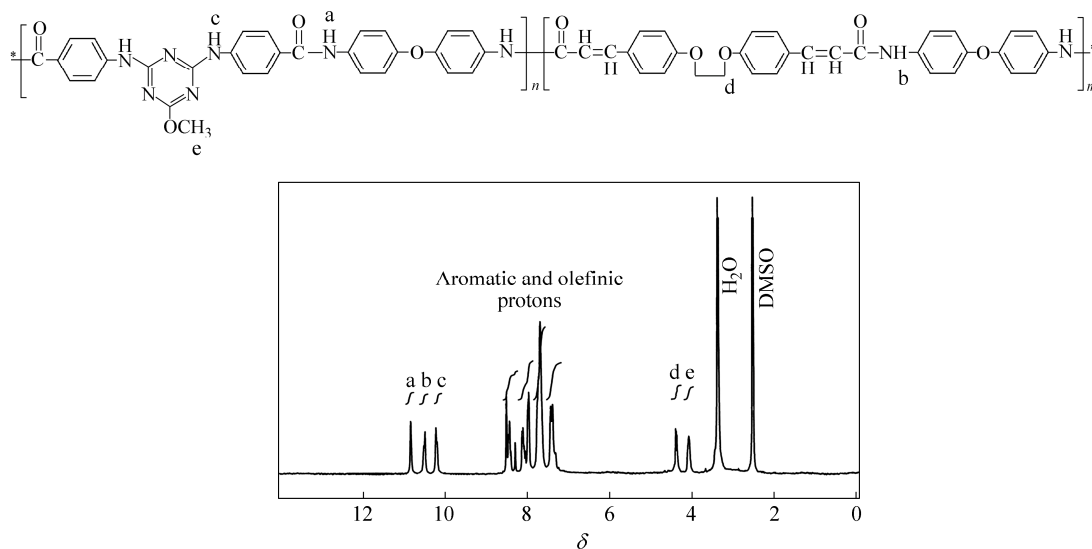


Fig. 5 ^1H -NMR spectrum of copolyamide **7b**

The entire polycondensation reaction readily proceeded in a homogeneous solution, tough and stringy precipitates formed when the viscous PAs and CoPAs solutions were obtained in good viscosities and yields. The syntheses and some physical properties of these new polymers are given in Table 1. Also, these synthesized polymers exhibited number-average molecular weights (M_n) and weight-average molecular weights (M_w) in the range of $1.7\text{--}2.4 \times 10^4$ and $3.6\text{--}4.9 \times 10^4$, as measured by GPC, relative to PVP (poly(vinylpyrrolidone)) standards (Table 1).

Table 1. Some physical properties, number-average and weight-average molecular weights of polymers

Polymer	Yield (%)	η_{inh} (dL/g) ^a	M_n (10 ⁴) ^b	M_w (10 ⁴) ^b	PDI ^c
PA 5	95	0.71	2.0	4.4	2.2
CoPA 7a	92	0.65	1.7	3.6	2.1
CoPA 7b	90	0.68	2.4	4.9	2.0
CoPA 7c	87	0.61	1.9	4.9	2.5
CoPA 7d	89	0.63	1.7	4.5	2.6

^a Measured at a concentration of 0.5 g/dL in DMF at 25 °C;^b Measured by GPC in DMF, Poly(vinylpyrrolidone) was used as standard;^c Polydispersity Index

Solubility of Polymers

One of the main objectives of this study was producing modified polyamides with improved solubility. The solubility of polymers was investigated as 0.01 g of polymeric sample in 2 mL of solvent. Due to the presence triazine ring as heterocyclic group and rigid aromatic ring in the structure of polyamide **5**, the solubility of this polymer was relatively low in common organic solvent. Polyamide **5** was soluble at room temperature in some aprotic solvent such as *N*-methyl-2-pyrrolidinone (NMP), *N,N*-dimethylformamide (DMF) and soluble on heating at 60 °C in *N,N*-dimethylacetamide (DMAc) and insoluble on heating at 60 °C in tetrahydrofuran (THF), pyridine (Py) and cyclohexanone and as well as protic solvents. Whereas, the presence of more ether linkage and aliphatic chains in backbone of CoPAs **7a–7d**, led to enhances solubility of polymers, especially CoPA **7c** and **7d**. All of these CoPAs were easily soluble at room temperature in aprotic polar solvents such as *N*-methyl-2-pyrrolidinone (NMP), *N,N*-dimethylacetamide (DMAc), *N,N*-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), soluble and partially soluble on heating at 60 °C in tetrahydrofuran (THF), pyridine (Py) and cyclohexanone, and insoluble in protic solvents such as acetone, chloroform, ethanol and methanol (Table 2). In comparison to the solubility of the synthesized polymers containing triazine ring in previous works^[19, 20], the copolymers **7a–7d** had better solubility in common solvents such as pyridine and THF.

Table 2. Solubility of PA **5** and CoPAs **7a–7d**

Solvent	PA 5	CoPAs			
		7a	7b	7c	7d
DMSO	++	++	++	++	++
DMF	++	++	++	++	++
NMP	++	++	++	++	++
DMAc	+	++	++	++	++
THF	–	+	+	+	+
Pyridine	–	+	+	++	++
Cyclohexanone	–	+-	+-	+	+
CHCl ₃	–	–	–	–	–
Acetone	–	–	–	–	–
EtOH	–	–	–	–	–
MeOH	–	–	–	–	–
H ₂ O	–	–	–	–	–

Solubility: ++: soluble at room temperature; +: soluble on heating at 60 °C;

+-: partially soluble on heating at 60 °C; -: insoluble on heating

Thermal Properties

The thermal properties of the prepared polymers were investigated by means of TGA as well as DSC. Thermogravimetric analysis was performed on polyamide **5** and CoPA **7a–7d** under inert atmosphere. Thermograms of these materials are described in Fig. 6, and thermal data are summarized in Table 3.

The initial thermal decomposition temperatures for CoPAs were found between 320–350 °C. However, the pristine PA **5** shows initial weight loss at 370 °C. Also, residual weights for CoPAs at 800 °C ranged from 26.9% and 34.4% in nitrogen respectively. According to these data, it can be concluded that incorporation more ether linkages and aliphatic chains in structure of copolymers, in comparison to PA **5**, reduces thermal stability, despite the increase solubility.

Differential scanning calorimetry (DSC) was used to determine the glass-transition temperature values (T_g) of the samples obtained with a heating rate of 10 K/min under nitrogen. The DSC results are summarized in Table 3. The glass transition temperature (T_g) determined in the second heating runs of DSC measurements of the investigated PA **5** is at 215 °C and for CoPAs is ranged between 208 °C and 195 °C.

Thus, we can speculate that incorporation of flexible groups into the polymer chain serves to moderate the thermal stability and decrease the T_g values of the CoPAs in comparison to PA **5**.

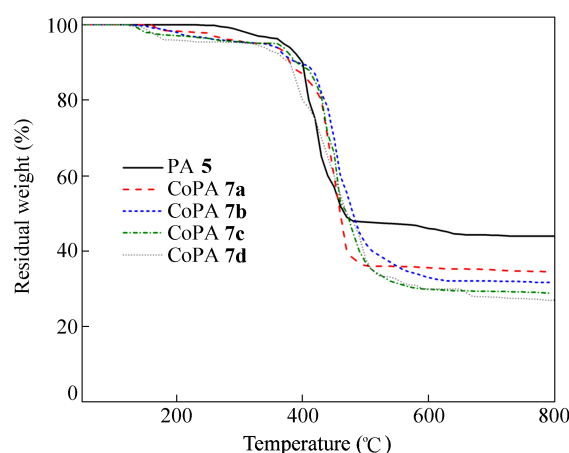


Fig. 6 TGA curves of PA **5** and CoPAs **7a–7d**

Table 3. Thermal behavior of PA **5** and CoPAs **7a–7d**

Polymer	T_g^a	T_5^b (°C)	T_{10}^b (°C)	Char yield ^c
PA 5	215	370	400	44.0
CoPA 7a	208	350	380	34.4
CoPA 7b	205	340	390	31.7
CoPA 7c	205	360	390	28.8
CoPA 7d	195	320	380	26.9

^a Glass transition temperature was recorded at a heating rate of 10 K/min in a nitrogen atmosphere;

^b Temperature at which 5% or 10% weight loss was recorded by TGA at a heating rate of 10 K/min under N₂;

^c Weight percentage of material left after TGA analysis at a maximum temperature of 800 °C under N₂

CONCLUSIONS

In this investigation we successfully synthesized some aliphatic-aromatic copolyamides containing triazine ring and ether linkages with excellent solubility in organic solvents and good thermal properties. For this reason, a new dicarboxylic acid containing triazine moiety was synthesized and with other monomers was used for synthesis of polyamide **5** and four copolyamides **7a–7d**. Structures and properties of these polymers were fully characterized by ¹H-NMR, FTIR, elemental analysis, inherent viscosity, and solubility tests, GPC, TGA and DSC. Comparison between PA **5** and CoPAs **7a–7d** showed that incorporation of more ether linkages and aliphatic chains as flexible groups in polymer backbone enhanced solubility in organic solvents and moderated thermal properties. These properties could make these polymers attractive for practical applications such as processable high-performance engineering plastics.

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