

# Synthesis and Characterization of Novel Polyamides Containing Azobenzene Units and Tetrahydropyrimidinone and Tetrahydro-2-thioxopyrimidine Derivatives by Microwave Assisted Polycondensation

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## ABSTRACT

Rapid and highly efficient synthesis of polyamides were achieved under microwave irradiation by using a domestic microwave oven from the polycondensation reactions of 4,4'-azodibenzoyl chloride [4,4'-azobenzenedicarboxylic acid] with seven different derivatives of tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine compounds in the presence of a small amount of a nonpolar organic medium that acts as a primary microwave absorber. Suitable organic media were *o*-cresol. The polycondensation proceeded rapidly, and compared with the conventional melt polycondensation and solution polycondensation it was almost completed between 8-12 min giving a series of polyamides with inherent viscosities about 0.22-0.42 dL/g. The resulting polyamides were obtained in high yield and they were thermally stable. All of the above compounds were fully characterized by means of FTIR spectroscopy, elemental analyses, viscometry and solubility test. Thermal properties of the polyamides were investigated using thermal gravimetric analysis (TGA) method.

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### Key Words:

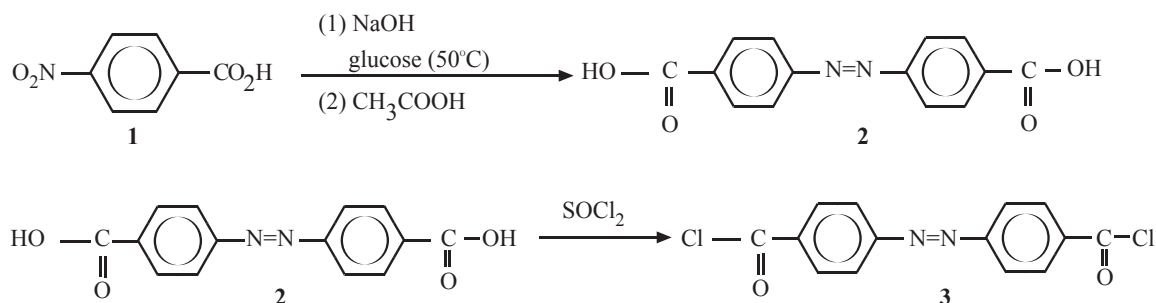
4,4'-Azodibenzoyl chloride;  
tetrahydropyrimidinone and  
tetrahydro-2-thioxopyrimidine com-  
pounds;  
microwave assisted polycondensation;  
inherent viscosity.

## INTRODUCTION

Aromatic polyamides have already been noted for their high temperature resistance and excellent mechanical properties. They are also considered as difficult processable materials because of their insolubility in common organic solvents and too high

glass transition temperatures [1-2]. Many approaches have been investigated in attempting to improve the solubility of aromatic polyamides including the addition of pendant groups to polymeric backbone [3-4] and incorporation of bulky [5-6] or

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Scheme I

flexible [7-9] and heterocycles [10-12] unit within the parent chain.

Recently, there has been growing interest in applying microwave energy to synthetic organic chemistry [13-14], sample preparation for analysis [15], extraction of natural products from plants [16], waste treatment [17] and polymer synthesis [18-19]. The reactions are very fast and are completed within a short period time. Recently we have used microwave irradiation for the synthesis of poly(amide-imide)s and poly(ester-imide)s [20-30].

This paper reports a rapid and highly efficient method for the synthesis of new polyamides with azobenzene moieties from the polycondensation reaction of 4,4'-azodibenzoyl chloride (3) with seven different derivatives of tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine compounds (4a-g) as a heterocycles unit under microwave irradiation. Monocyclic six-member ring nitrogen heterocycles such as tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine compounds are an extremely important class of compounds that occur in a wide variety of natural products such as purine, caffeine, nucleic acids (DNA, RNA), uric acid, and synthetic products [31].

## EXPERIMENTAL

### Apparatus

A Samsung domestic microwave oven (2450 MHz, 900W) was used for carrying out polycondensation reactions.

### Materials

All chemicals were purchased from Fluka Chemical Co. (Switzerland), Aldrich Chemical Co. (Milwaukee, WI) and Merck Chemical Co. (Germany).

### Techniques

Fourier transform infrared (FTIR) spectra were recorded on Galaxy series FTIR 5000 spectrophotometer (England). Spectra of solids were performed using KBr pellets. Vibrational transition frequencies are reported in wavenumber (cm<sup>-1</sup>). 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 Technico Regd. Trade Mark Viscometer. Thermal gravimetric analysis (TGA) data for polymers were taken on a Mettler TA4000 System under N<sub>2</sub> atmosphere at a rate of 10°C/min. Elemental analyses were performed by the Research Institute of Petroleum Industry, Tehran, Iran.

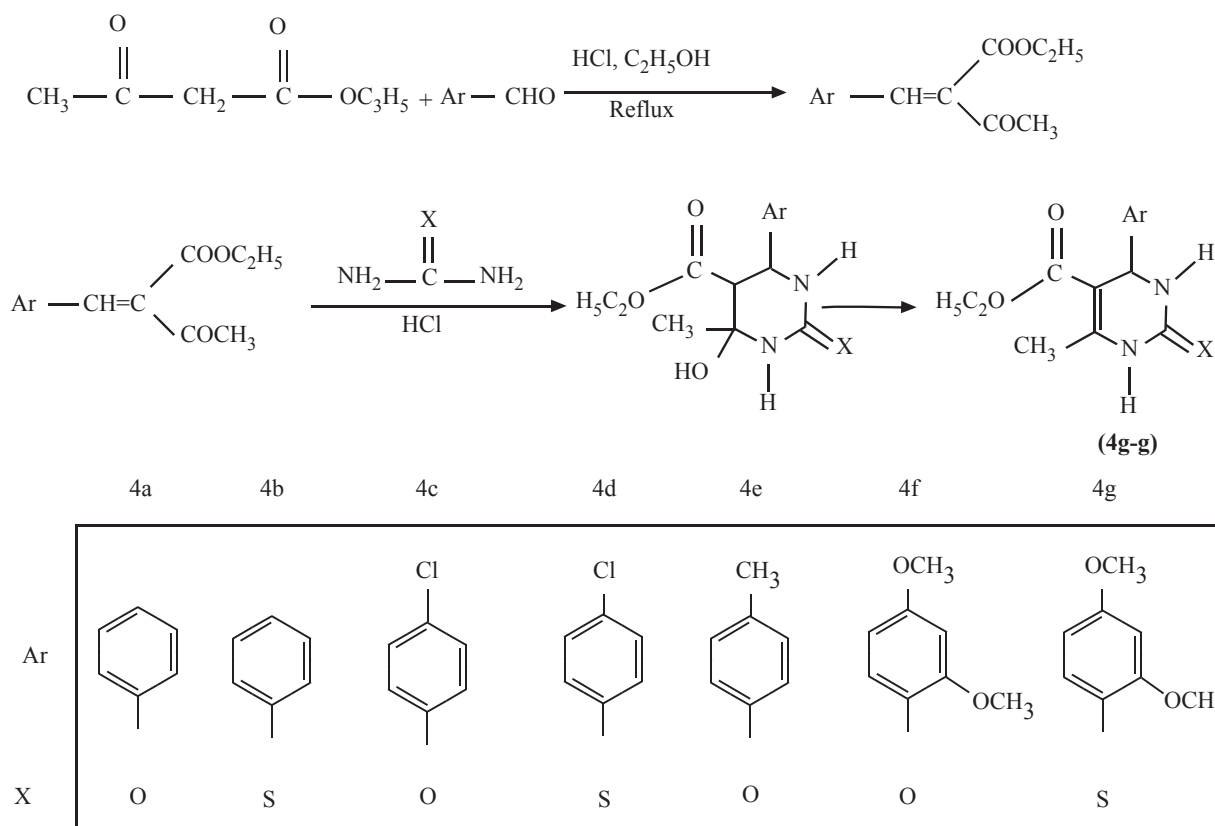
### Monomer Synthesis

#### 4,4'-Azodibenzoyl Chloride 3

4,4'-Azodibenzoyl chloride 3 was prepared according to a typical procedure that was shown in scheme I [32-33].

#### 4,4'-Azodibenzoic Acid [4,4'-Azodibenzenedicarboxylic Acid] 2

4-Nitrobenzoic acid 1 (13 g, 0.079 mol) was heated in sodium hydroxide solution (50 g in 250 mL) at 50°C. A solution of glucose (100 g in 150 mL of water) was added slowly at this temperature with occasional shaking. The reaction mixture was then cooled to ambient temperature and aerated for 8 h with vigorous stirring until orange crystals were formed. The mixture was acidified with dilute acetic acid. The liberated diacid was filtered, washed with water and dissolved in hot potassium carbonate solution to obtain orange coloured solution. This solution was concentrated to obtain orange crystals of potassium salt of diacid. On acidifying with dilute acetic acid, 18 g (86 %) of rose coloured 4,4'-azodibenzoic acid 2 was obtained, mp > 300°C, FTIR (KBr): 3600-2900 (m, br), 1669 (s, br), 1614 (s),



Scheme II

1579 (s), 1410 (s), 1300-1200 (s, br), 1100-1000 (s, br), 868 (s), 777 (s)  $\text{cm}^{-1}$ . Analysis: Calculated for  $\text{C}_{14}\text{H}_{10}\text{N}_2\text{O}_4$ : C, 62.24; H, 3.70; N, 10.36; found: C, 62.4; H, 3.8; N, 10.

#### 4,4'-Azodibenzoyl Chloride 3

Into a 100 mL round-bottomed flask were placed (4 g,  $1.48 \times 10^{-2}$  mol) of compound **3**, 15 mL of thionyl chloride and 0.2 mL triethylamine as a base. The mixture was heated in an oil bath up to  $60^\circ\text{C}$ , until the suspension mixture was converted to a clear solution. Then, the solution was stirred for over night at room temperature. Unreacted thionyl chloride was removed under reduced pressure and the residue was washed with dry n-hexane two times, to leave 4.3 g (95%) of red colour crystals with mp  $177\text{-}179^\circ\text{C}$ . FTIR (KBr): 1776 (s), 1732 (m, br), 1595 (m), 1408 (m), 1195 (s, br), 885 (s), 804 (m), 646 (m)  $\text{cm}^{-1}$ . Analysis: Calculated for  $\text{C}_{14}\text{H}_8\text{N}_2\text{O}_2\text{Cl}_2$ : C, 54.72; H, 2.60; N, 9.12; found: C, 54.9; H, 2.8; N, 8.9.

#### Tetrahydropyrimidinone and Tetrahydro-2-thioxopyrimidine Compounds 4a-g

Tetrahydropyrimidinone and tetrahydro-2-thioxopy-

rimidine compounds **4a-g** were prepared according to the Biginelli condensation [34].

#### Polymer Synthesis

Polyamides **5a-g** were synthesised by a typical procedure shown in Scheme III.

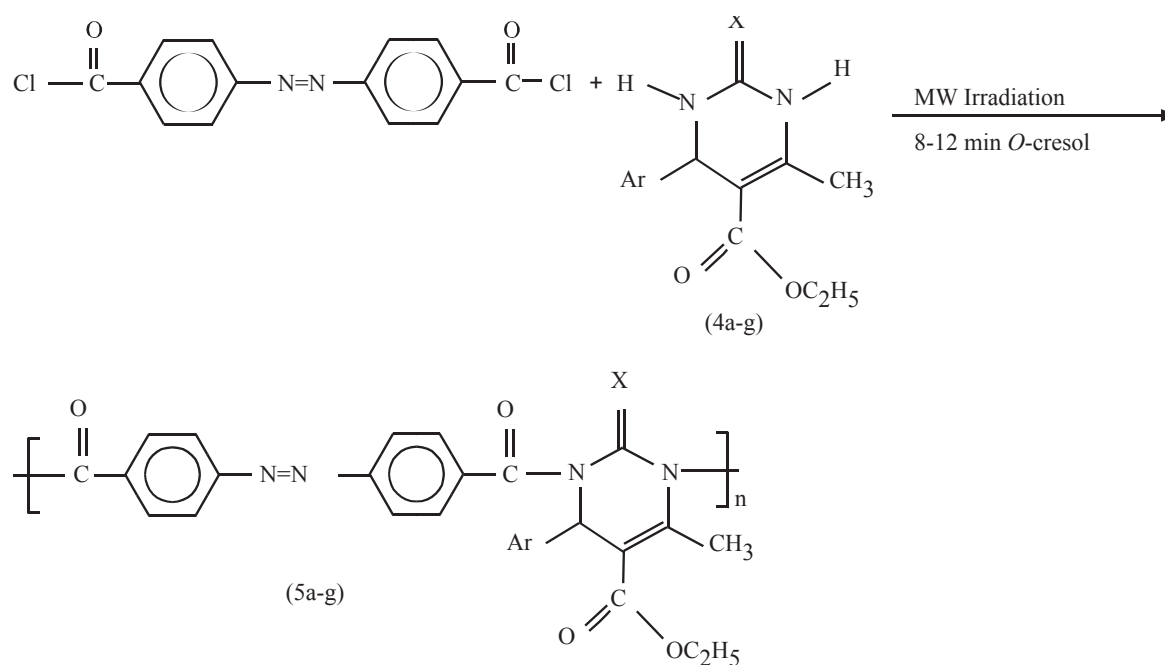
An equimolar mixture of 4,4'-azodibenzoyl chloride **3** (0.4 g,  $1.3 \times 10^{-3}$  mol) and tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine compounds **4a-g** ( $1.3 \times 10^{-3}$  mol) were placed in a reaction vessel and the mixture was ground until fine powder was formed. The polyamides **5a-g** were then synthesized by microwave assisted polycondensation reactions by placing in microwave oven at full power for 8-12 min. The reaction mixture was poured into 50 mL of methanol. The resulting polymer was filtered off, and dried at  $80^\circ\text{C}$  for 8 h under the vacuum.

#### Polymer 5a

FTIR (KBr): 1732 (s, br), 1705 (s, br), 1619 (w), 1508 (m), 1410 (m), 1261 (s, br), 1220 (s), 1170 (s), 1111 (m), 1109 (m), 1070 (s), 770 (w), 754 (m)  $\text{cm}^{-1}$ .

#### Polymer 5b

FTIR (KBr): 1732-1700 (s, br), 1601(w), 1489 (m),



Scheme III

1410 (m), 1261 (s, br), 1221 (m), 1172 (s), 1111(m), 1068 (s), 1010 (w), 871 (w), 770 (w), 754 (m)  $\text{cm}^{-1}$ .

#### Polymer 5c

FTIR (KBr): 1732-1710 (s, br), 1649 (m), 1531 (s), 1489 (m), 1411 (m), 1261 (s, br), 1220 (s), 1172 (s), 1109 (m), 1070 (s), 860 (w), 785 (m), 754 (m)  $\text{cm}^{-1}$ .

#### Polymer 5d

FTIR (KBr): 1732-1710 (s, br), 1601 (m), 1504 (w), 1489 (m), 1410 (m), 1261 (s, br), 1221 (s), 1172 (s), 1111 (m), 1068 (s), 1010 (w), 869 (w), 773 (m), 754 (m)  $\text{cm}^{-1}$ .

#### Polymer 5e

FTIR (KBr): 1732 (s, br), 1705 (s, br), 1647 (m), 1494 (m), 1465 (w), 1263 (s, br), 1224 (s, br), 1172 (s), 1111(m), 1087 (m), 1068 (s), 871 (w), 754 (m)  $\text{cm}^{-1}$ .

#### Polymer 5f

FTIR (KBr): 1732-1710 (s, br), 1601 (w), 1491 (m), 1410 (m), 1262 (s, br), 1221 (s), 1172 (s), 1111(m), 1068 (s), 1010 (w), 871 (w), 770 (w), 754 (m)  $\text{cm}^{-1}$ .

#### Polymer 5g

FTIR (KBr): 1732-1710 (s, br), 1602 (w), 1489 (m), 1410 (m), 1260 (s, br), 1220 (s), 1170 (s), 1111 (m),

1109 (m), 1070 (s), 770 (w), 754 (m)  $\text{cm}^{-1}$ .

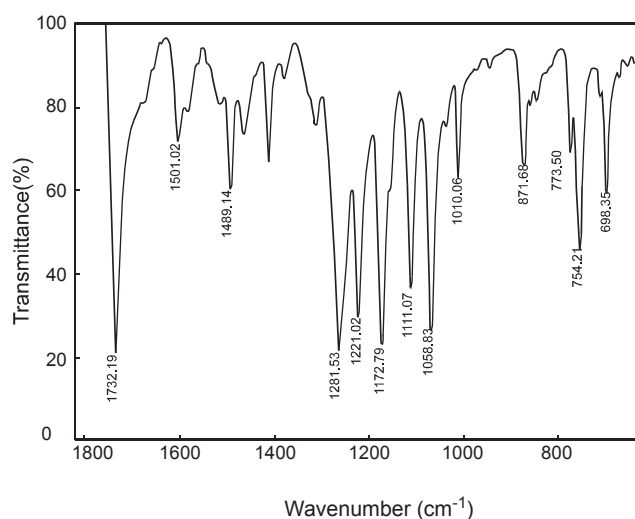
## RESULTS AND DISCUSSION

### Monomer Synthesis

4-Aryl-3,4-dihydro-2(1H)-pyrimidinone esters of type **4a-g** (Biginelli compounds) represent a heterocyclic system with remarkable pharmacological efficiency [35]. The most straightforward protocol to synthesize these compounds (**4a-g**) involves the one-pot condensation of a  $\beta$ -keto ester, with an aryl aldehyde and urea or thiourea derivative under strongly acidic condition (Scheme II)[34].

### Polymer Synthesis

Polyamides **5a-g** were synthesized by microwave assisted polycondensation reactions of an equimolar mixture of monomer **3** with seven different derivatives of tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine compounds **4a-g** in *o*-cresol as organic solvent by using a domestic microwave oven as shown in Scheme III. The *o*-cresol was used as polar organic medium that acts as a primary microwave absorber and as a solvent for both of the starting monomers and the resulting polymers, as it also allows to induce effective homogeneous heating of the reaction mixture and



**Figure 1.** FTIR Spectrum of PAIs **5a**.

thereby subsequent polycondensation reaction leading to the polymer formation. The optimum period of reaction time (irradiation time) was found between 8-12min and below this time will give polymers with lower inherent viscosities and above this time the materials will degrade.

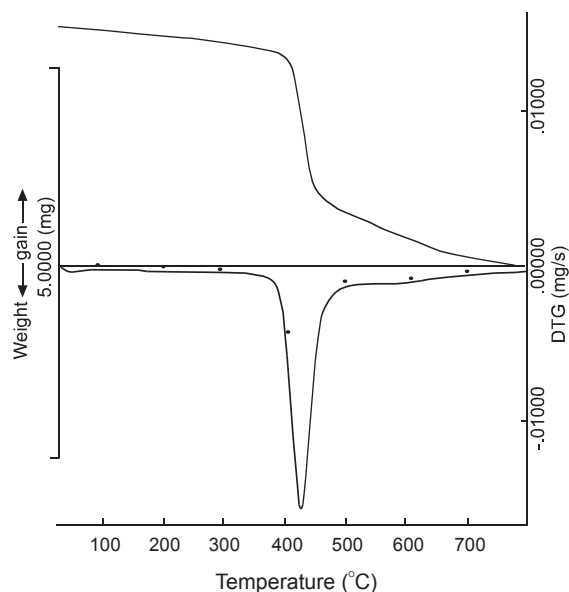
### Polymer Characterization

Synthesis and some physical properties of polyamides **5a-g** are summarized in Table 1. These polymers have inherent viscosities in a range from 0.22-0.42 dL/g. Polyamides derived from monomer **3** may range in colours from bright red to dark red. The structures of these polymers were confirmed as polyamides by

**Table 1.** Synthesis and some physical properties of polyamides **5a-g**.

Heterocyclic diamine	Polymer	Yield (%)	$\eta_{inh}$ (dL/g) <sup>a</sup>	Time <sup>b</sup> (min)
4a	5a	82	0.40	12
4b	5b	84	0.35	8
4c	5c	82	0.38	10
4d	5d	78	0.42	8
4e	5e	82	0.35	9
4f	5f	80	0.22	9.5
4g	5g	71	0.30	11

(a) Measured at a concentration of 0.5g /dL in DMF at 25°C. (b) Time of microwave irradiation.



**Figure 2.** TGA & DTG Thermogram of PAIs (**5a**).

means of FTIR spectroscopy and elemental analyses (Table 2).

The representative FTIR spectrum of polyamide **5a** is shown in Figure 1. The polymer shows absorption bands around 1740-1700  $\text{cm}^{-1}$  due to amide and ester carbonyl groups. Absorption bands around 1650-1600  $\text{cm}^{-1}$  and 1350-1460 are due to aromatic rings vibra-

**Table 2.** Elemental analysis of polyamides **5a-g**.

Polymer	Formula		C (%)	H (%)	N (%)
5a	$\text{C}_{28}\text{H}_{22}\text{N}_4\text{O}_5$ (494.30) <sub>n</sub>	calcd.	68.03	4.45	11.32
		found.	66.8	4.9	10.3
5b	$\text{C}_{28}\text{H}_{22}\text{N}_4\text{O}_4\text{S}$ (510.37) <sub>n</sub>	calcd.	65.89	4.31	10.97
		found.	64.6	5.0	10.0
5c	$\text{C}_{28}\text{H}_{21}\text{N}_4\text{O}_5\text{Cl}$ (528.80) <sub>n</sub>	calcd.	63.59	3.97	10.59
		found.	62.8	4.2	9.9
5d	$\text{C}_{28}\text{H}_{21}\text{N}_4\text{O}_4\text{SCl}$ (544.87) <sub>n</sub>	calcd.	61.72	3.85	10.27
		found.	60.5	3.9	9.8
5e	$\text{C}_{29}\text{H}_{24}\text{N}_4\text{O}_5$ (508.32) <sub>n</sub>	calcd.	68.52	4.72	11.01
		found.	67.1	5.1	10.2
5f	$\text{C}_{30}\text{H}_{26}\text{N}_4\text{O}_7$ (554.33) <sub>n</sub>	calcd.	65.00	4.69	10.10
		found.	63.9	4.6	10.1
5g	$\text{C}_{30}\text{H}_{26}\text{N}_4\text{O}_6\text{S}$ (570.39) <sub>n</sub>	calcd.	63.17	4.55	9.8
		found.	61.8	4.2	9.5

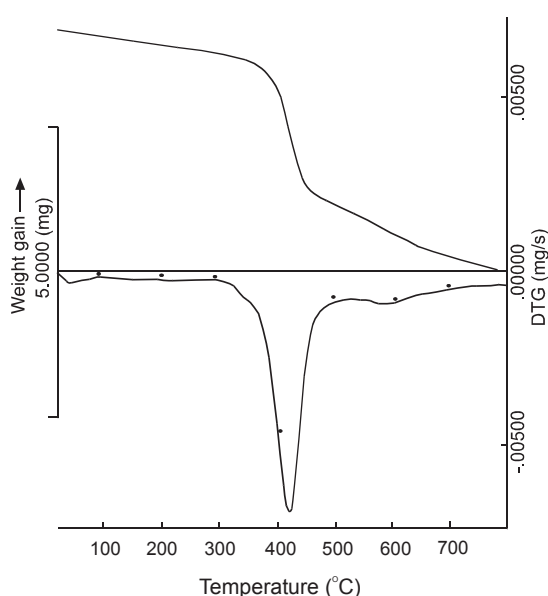
**Table 3.** Thermal behaviour of polyamides.

Polymer	Decomposition temperature(°C) $T_5^a$	Char yield(%) <sup>b</sup>
5a	275-280	15.0
5c	280-285	18.0

(a) Temperature at which 5% weight loss was recorded by TEA at a heating rate of 10°C/min.

tions, methyl and methylene stretching, respectively. Absorption bands around 1200-1100  $\text{cm}^{-1}$  correspond to C-O stretching of ester. The other spectra show a similar pattern. The elemental analysis values of the resulting polymers are in good agreement with the calculated values for the proposed structures (Table 2).

Comparing inherent viscosities of this method with the previous method [36,37], show that the internal heat generation of both the solvent and the monomers under the microwave irradiation was much more effective for the progress of polycondensation, producing polyamides having higher viscosity value in a shorter polymerization time, than the conventional external heating. The solubility of polyamides **5a-g** was investigated as 0.01 g of polymeric sample in 2 mL of solvent. All of the polymers are soluble in organic solvents such as DMF, DMAc, DMSO and chloroform at room temperature and insoluble in solvents such as methylene chloride, methanol, ethanol and water.

**Figure 3.** TGA and DTG thermogram of PAIs (5c).

### Thermal Properties

The thermal properties of polyamides **5a** and **5c** were evaluated by means of TGA/DTA in nitrogen atmosphere (Table 3). All of the polymers show similar decomposition behaviour (Figures 2-3). For **5a** which started to decompose at 275°C, lost 5 % of its weight due to decomposition by 280°C and by 600°C it had only 15.0% of its original weight. For **5c** it started to decompose at 285°C, and lost 5 % of its weight due to decomposition by 285°C and by 600°C it had only 18.0 % of its original weight.

### CONCLUSION

A new series of polyamides with azobenzene and tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine moieties in the main chain are synthesized and characterized by FTIR spectroscopy, elemental analysis, viscometry, and solubility test. These new polyamides (**5a-g**) are able to be synthesized by microwave assisted polycondensation reaction using the monomer **3** with seven different derivatives of tetrahydropyrimidinone and tetrahydro-2-thioxopyrimidine compounds (**4a-g**). These new polyamides are soluble in various organic solvents and have good thermal stability.

The above results demonstrate that microwave heating is an efficient method (shorter reaction time and high efficiency of energy) for the polycondensation reactions. We are currently using this method for the synthesis of novel monomers, polymers and modification of polymers.

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