

## SOLUBLE POLYIMIDES FROM A SEMI-ALIPHATIC DIAMINE CONTAINING ESTER, AMIDE AND ETHER GROUPS

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**Abstract** A diamine was synthesized by two successive reactions. Nucleophilic reaction of 4-hydroxybenzoic acid with terephthaloyl chloride yielded terephthaloyl bis(4-oxybenzoic) acid. Then reaction of this compound with 1,8-diamino-3,6-dioxaoctane *via* Yamazaki method resulted in preparation of diamine named terephthalic acid bis(4-{2-[2-(2-aminoethoxy)ethoxy]ethyl carbamoyl}phenyl) ester. After fully characterization it was used to prepare new polyimides through polycondensation with different dianhydrides using trimethylchlorosilane. Characterization of polymers was achieved by common methods and their physical properties including inherent viscosity, thermal behavior, thermal stability, crystallinity and solubility were studied. Prepared polyimides showed improved solubility and good thermal stability.

**Keywords:** Polyimide; Polycondensation; Synthesis; Thermal properties.

### INTRODUCTION

Aromatic polyimides are well accepted as high performance polymers and they are well as high temperature resistant materials<sup>[1]</sup>. They reveal favorable balance of physical and chemical properties and display excellent thermal, mechanical and electrical properties, and they are extensively used in microelectronic and aerospace engineering<sup>[2, 3]</sup>.

However, the technological and industrial applications of rigid polyimides are limited by processing difficulties due to their high melting or glass transition temperatures and lack of solubility in most organic solvents<sup>[4]</sup>. Strong interaction between polyimide polar chains and their rigid structures are the main reasons for these behaviors.

To overcome such problems, different methods have been introduced based on the modification of polymer structure. Much effort has been focused on designing and preparing new dianhydrides<sup>[5–8]</sup> and diamines<sup>[9–15]</sup>, and therefore producing a great variety of soluble and processable polyimides for various purposes and applications. Copolycondensation is one of the possible ways for modification of polymer properties. Incorporation of flexible units such as ether, ester and amide units is among the most important approaches to overcome these processing problems<sup>[14, 16]</sup>. In this way, poly(amide-imide)s have deserved particular interest, as they are a class of copolyimides that most closely resemble the thermal properties of aromatic polyimides. Furthermore the insertion of an amide group into the polyimide backbone increases its processability, solubility and moldability<sup>[17]</sup>. Poly(amide imide)s provide a favorable balance between processability and performance and are useful in numerous applications in electrical wire enamel, adhesives and injection-molding and extrusion products<sup>[18]</sup>.

This article presents synthesis of a semi-aliphatic diamine containing preformed ester, ether and amide groups and polycondensation reactions of the diamine with different aromatic dianhydrides to prepare new soluble, thermally stable polyimides.

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## EXPERIMENTAL

### Materials

All chemicals were purchased either from Merck or Aldrich Chemical Co. Dianhydrides including pyromellitic dianhydride (PMDA), benzophenone-3,3',4,4'-tetracarboxylic dianhydride (BTDA) and 4,4'-(hexafluoroisopropylidene)diphthalic anhydride (6FDA) were dried in a vacuum oven at 110°C for 5 h. Lithium chloride was dried in a vacuum oven at 120°C for 10 h. 1,8-Diamino-3,6-dioxaoctane (DADO) was distilled under reduced pressure. Terephthaloyl chloride (TPC) was purified by sublimation. Pyridine was vacuum distilled over KOH. *N*-methyl-2-pyrrolidone (NMP), *N,N*-dimethyl acetamide (DMAc), *N,N*-dimethyl formamide (DMF), dimethyl sulfoxide (DMSO) and toluene were vacuum distilled over calcium hydride.

### Instruments

Infrared measurements were performed on a Bruker-IFS 48 FTIR spectrometer (Ettlingen, Germany). The <sup>1</sup>H-NMR spectra were recorded in dimethyl sulfoxide (DMSO-*d*<sub>6</sub>) solution using a Bruker Avance DPX 250 MHz instrument (GmbH, Germany). A CHN-O-Rapid Heraeus elemental analyzer performed elemental analyses (Wellesley, MA). Differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) were recorded on a Stanton Redcraft STA-780 (London, UK). Differential thermogravimetric (DTG) traces were recorded on a Polymer Lab TGA-1500 (London, UK). The dynamic mechanical measurements were performed on a Polymer Laboratories Dynamic Mechanical Thermal Analyzer (Model MK-II) over a temperature range of 25–200°C at 1 Hz and a heating rate of 5 K/min (Surrey, UK). The value of  $\tan\delta$  and the storage modulus versus temperature were recorded for each sample. Inherent viscosities were measured by using an Ubbelohde viscometer in a concentration of 0.5 g/dL in DMF at 30°C. Wide angle X-ray diffraction patterns were performed at room temperature on an X-ray diffractometer (Siemens model D 5000) using Ni-filtered Cu K $\alpha$  radiation (40 kV, 25 mA) with scanning rate of 3 (°)/min. The liquid crystalline texture of the polymers was observed with an optical microscope (Zeiss) fitted with crossed polarizers. The temperature was controlled to  $\pm 1^\circ\text{C}$  by means of a computerized Linkam TMS 94 heat controller. Mechanical tensile testing was performed with an ADAMEL testing instrument MTS 10/M equipped with a chamber. Stress-strain curves were obtained at a stretching speed of 25 mm/min for samples cut into a dumbbell shape [ASTM D638].

### Monomer Synthesis

#### *Synthesis of terephthaloyl bis(4-oxybenzoic) acid (TOBA)*

164 mmol (23.1 g) of 4-hydroxybenzoic acid was dissolved in 350 mL of 0.1 mol/L aqueous solution of sodium hydroxide. After cooling the solution to 10°C, 56 mmol (11.6 g) of terephthaloyl chloride dissolved in 115 mL of tetrachloroethane was added drop-wise to the mixture with stirring. The reaction mixture was stirred at room temperature for 6 h. The mixture was soaked in 430 mL of 3 mol/L HCl for 14 h. The product was obtained by filtration and washed with water, ether and ethanol successively. Then it (TOBA) was vacuum dried at 60°C overnight (yield 85%).

#### *Synthesis of terephthalic acid bis(4-{2-[2-(2-amino ethoxy)ethoxy]ethyl carbamoyl}phenyl) ester (TABE)*

A mixture of 11.5 mmol (1.70 g) of 1,8-diamino-3,6-dioxaoctane (DADO), 5.00 mmol (2.03 g) of TOBA, 1.80 g of calcium chloride, 4.00 mL of triphenylphosphite, 4.00 mL of pyridine and 20.0 mL of NMP was refluxed for 3 h. After cooling, the reaction mixture was poured into a large amount of methanol with stirring. The obtained precipitate was filtered and purified by solvent-non solvent technique using DMF-H<sub>2</sub>O solvents. Then it was filtered and washed with methanol and hot water. Finally it was dried at 60°C under vacuum overnight (yield 78%).

### Polyimide Synthesis

A typical procedure for the preparation of poly(amic acid) was as follows: A 100 mL, two-necked, round-bottomed flask equipped with a magnetic stirrer, nitrogen gas inlet tube was charged with 0.530 g of LiCl and 15.0 mL of dry NMP. The mixture was stirred at room temperature. Then 5.00 mmol of TABE was added. The

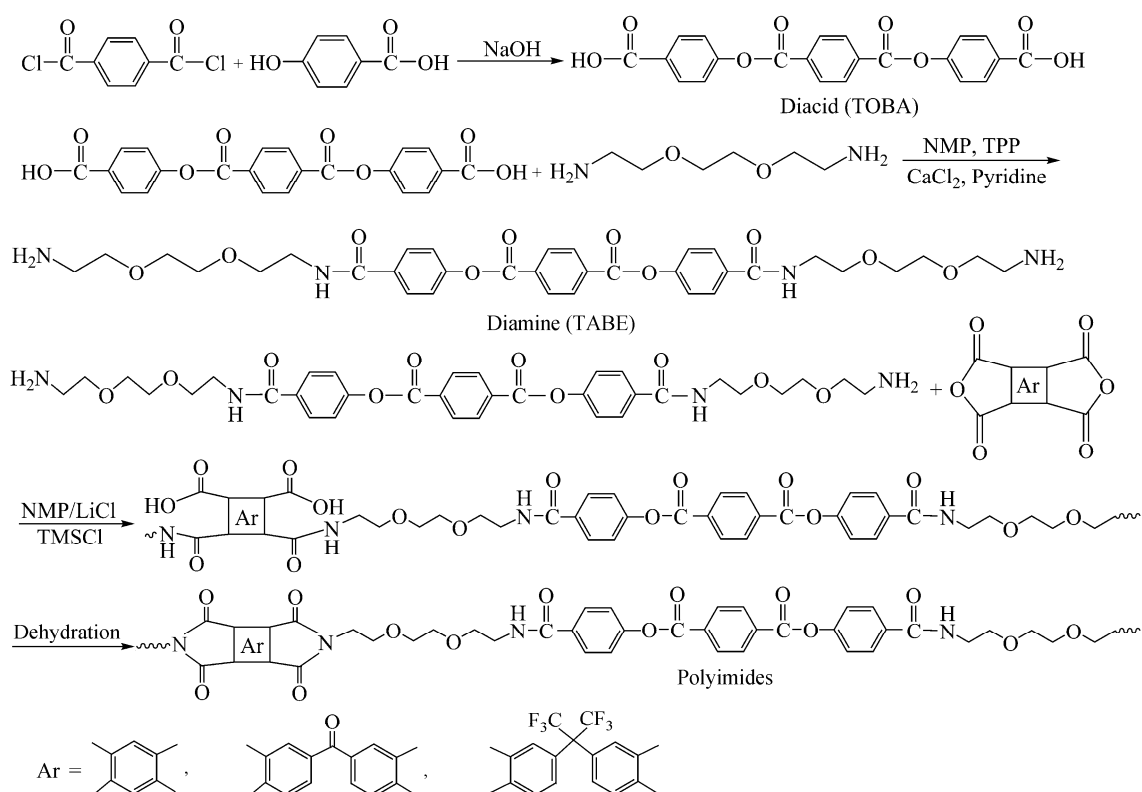
mixture was cooled to  $-5^{\circ}\text{C}$  and 2.00 mmol of trimethylchlorosilane (TMSCl) was added to it. The temperature was raised to room temperature and the solution was stirred for 15 min. Then the solution was once more cooled to  $-5^{\circ}\text{C}$ , and 5.0 mmol of dianhydride (PMDA, BTDA or 6FDA) was rapidly added. The reaction mixture was stirred for 0.5 h at this temperature, after that it was heated to room temperature and stirred for 24 h. Poly(amic acid) was precipitated by pouring the flask contents into water; it was then filtered, washed under vacuum at  $30^{\circ}\text{C}$ . Yields over 80% were obtained.

Chemical cyclization was applied for conversion of poly(amic acid) to polyimide: Into a 100 mL, two-necked, round-bottomed flask equipped with a magnetic stirrer, nitrogen gas inlet tube, and a reflux condenser were placed 1.66 g of poly(amic acid) and 8.20 mL of dry NMP. The mixture was stirred until all solids were dissolved. Then 8.20 mL of acetic anhydride and 4.25 mL of pyridine were added. The mixture was stirred for 0.5 h and then slowly heated to  $140^{\circ}\text{C}$  and held for 6 h at the same temperature. After that, the mixture was cooled and poured into water, it was then filtered, washed with hot water and methanol, and dried overnight under vacuum at  $120^{\circ}\text{C}$ . Yields over 75% were obtained.

## RESULTS AND DISCUSSION

The major purpose of this research was to prepare novel polyimides with improved solubility without too much sacrificing their thermal stability. In this way, synthesis of a diamine with built-in ester, ether and amide units as a building monomer for the preparation of final polymers was considered.

Nucleophilic reaction of 4-hydroxybenzoic acid with terephthaloyl chloride in the presence of NaOH yielded terephthaloyl bis(4-oxybenzoic) acid (TOBA) (Scheme 1). TOBA was converted to TABE by reaction of TOBA with DADO *via* Yamazaki method in the presence of NMP, triphenylphosphite,  $\text{CaCl}_2$  and pyridine (Scheme 1).



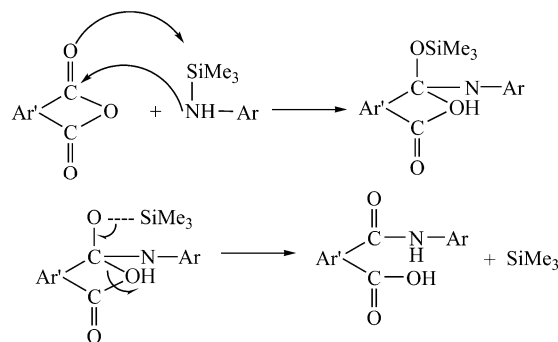
**Scheme 1** Preparation of TOBA, TABE, and polyimides

TOBA and TABE were characterized using FTIR,  $^1\text{H-NMR}$  and elemental analysis techniques, and the results are tabulated in Table 1.

**Table 1.** Characterization of TOBA and TABE

| Name | IR (KBr, cm <sup>−1</sup> )                                                                                                  | <sup>1</sup> H-NMR (DMSO-d <sub>6</sub> , δ)                                                                                                                                                                                                                                                                                       | Elemental analysis |      |      |       |      |      | Yield |
|------|------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------|------|------|-------|------|------|-------|
|      |                                                                                                                              |                                                                                                                                                                                                                                                                                                                                    | Calc.              |      |      | Found |      |      |       |
|      |                                                                                                                              |                                                                                                                                                                                                                                                                                                                                    | C                  | H    | N    | C     | H    | N    |       |
| TOBA | 3250 (O—H);<br>1748 (COOR);<br>1688 (COOH);<br>1425(C=C);<br>1285 (C—O)                                                      | 13.11 s (2H, COOH); 8.28 s (4H, aromatic) 8.14 d (4H, aromatic); 7.79 d (4H, aromatic)                                                                                                                                                                                                                                             | 65.02              | 3.45 | —    | 65.27 | 3.19 | —    | 85%   |
| TABE | 3298 (N—H);<br>3076 (Ar—H);<br>2921, 2870 (CH <sub>2</sub> );<br>1740 (COO);<br>1630 (CONH);<br>1547 (NH, CN);<br>1281 (C—O) | 8.63 s (2H, NH); 8.19 d (4H, aromatic); 7.90 d (4H, aromatic); 7.40 d (4H, aromatic); 3.61 t (4H, —O—CH <sub>2</sub> —); 3.52 t (4H, —O—CH <sub>2</sub> —); 3.42 t (8H, O—CH <sub>2</sub> —CH <sub>2</sub> —); 3.36 t (4H, —CH <sub>2</sub> —NH—); 2.54 t (4H, —CH <sub>2</sub> —NH <sub>2</sub> ); 1.23 s (4H, —NH <sub>2</sub> ) | 61.26              | 6.30 | 8.40 | 60.85 | 6.16 | 8.28 | 78%   |

Therefore a flexible diamine (TABE) with built-in ester, amide, and ether groups and methylene linkages was synthesized and used to prepare related polyimides. In the polycondensation reaction of the diamine with dianhydrides, *in situ* silylation of TABE was considered in order to increase the reactivity of the diamine toward dianhydride and therefore improving polymer formation with higher molecular weight<sup>[19–24]</sup>. The following two steps nucleophilic addition-elimination mechanism has been proposed for the acyl substitution of an anhydride with an *N*-silylated amine<sup>[25]</sup>:



In the first step, due to the strong affinity of Si for oxygen, the carbonyl oxygen of the anhydride is attracted to the Si atom in the amine derivatives, which in turn facilitates the nucleophilic attack of the N atom of the *N*-silylated amine on the carbonyl carbon. In the second step, the ring opening tetrahedral intermediate is enhanced by the presence of the  $\beta$ -silicon through the  $\sigma$ - $\pi$  effect, affording rapidly the amide product along with TMSiOH. Additionally, use of TMSiCl ensures that small amount of water in the reaction solution will not destroy the moisture sensitive anhydride. This important advantage is attributed to the high tendency of TMSiCl in reaction with water.

Thus, polycondensation reactions of the TABE with different aromatic dianhydrides including pyromellitic dianhydride (PMDA), benzophenone-3,3',4,4'-tetracarboxylic dianhydride (BTDA) and 4,4'-(hexafluoroisopropylidene)diphthalic anhydride (6FDA) resulted in preparation of three semi-aromatic polyimides. It is worth to mention that these polyimides were prepared *via* two-step methods. First, corresponding polyamic acids were prepared and then they were converted to polyimides by chemical cyclization using acetic anhydride and pyridine reagent (Scheme 1).

Structures of the polymers were characterized and confirmed using FTIR,  $^1\text{H-NMR}$  and elemental analysis. The results are collected in Table 2.

**Table 2.** Characterization of polymers

| Product   | IR (KBr, $\text{cm}^{-1}$ )                                       | $^1\text{H-NMR}$ ( $\text{DMSO-d}_6$ , $\delta$ )                                                          | Elemental analysis           |              |              |
|-----------|-------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------|------------------------------|--------------|--------------|
|           |                                                                   |                                                                                                            | C                            | H            | N            |
| TABE-PMDA | 3305, 3097, 2985, 2868, 17921713, 1656, 1544, 1496, 13701220, 729 | 8.82/2H, 8.73/2H, 8.29/4H, 8.06/4H, 7.50/4H, 3.81/4H, 3.71/4H, 3.69/4H, 3.56/8H, 3.38/4H                   | Calcd: 62.22<br>Found: 61.98 | 4.71<br>4.94 | 6.59<br>6.38 |
| TABE-BTDA | 3305, 3104, 2989, 2870, 17791716, 1690, 1572, 1507, 13711218, 729 | 8.71/2H, 8.68/2H, 8.31/4H, 8.23/2H, 8.13/2H, 8.04/4H, 7.43/4H, 3.79/4H, 3.70/4H, 3.68/4H, 3.55/8H, 3.36/4H | Calcd: 64.24<br>Found: 64.02 | 4.60<br>4.32 | 5.88<br>6.01 |
| TABE-6FDA | 3312, 3097, 2985, 2868, 17921713, 1656, 1544, 1496, 13701220, 728 | 8.68/2H, 8.25/4H, 8.04/2H, 8.02/4H, 7.96/2H, 7.53/2H, 7.31/4H, 3.76/4H, 3.68/4H, 3.64/4H, 3.53/8H, 3.35/8H | Calcd: 59.90<br>Found: 59.69 | 4.09<br>3.91 | 5.20<br>5.01 |

Since the inherent viscosity is a suitable criterion for estimation of molecular weight, the inherent viscosity of the polyimides in DMF at a concentration of 0.5 g/dL at 30°C was measured (Table 3). According to the obtained viscosities (0.536–0.551 dL/g), the polyimides prepared *via* activation with TMSCl showed reasonable molecular weights whereas in the absence of TMSCl these values were much lower (0.310–382 dL/g).

**Table 3.** Reactants, structures, yields and inherent viscosities of the polymers

| Polymer   | Structures | Inherent viscosity (dL/g) | Yield (%) |
|-----------|------------|---------------------------|-----------|
| TABE-PMDA |            | 0.551                     | 87        |
| TABE-BTDA |            | 0.549                     | 85        |
| TABE-6FDA |            | 0.536                     | 82        |

The polyimides showed improved solubility in dipolar aprotic solvents (Table 4) including DMF, DMAC, NMP, DMSO and also in less efficient solvent such as *m*-cresol. The improved solubility (about 2.4–3.3 g/dL) was attributed to the introduction of flexible ether and methylene groups into the polymer backbone. Presence of carbonyl and hexafluoroisopropylidene groups in BTDA- and 6FDA-derived polyimides increased solubility in comparison with rigid PMDA-derived polyimide.

**Table 4.** Solubility behavior of polymers

| Polymer   | NMP | DMAC | DMF | DMSO | <i>m</i> -Cresol | Pyridine | THF | $\text{CHCl}_3$ |
|-----------|-----|------|-----|------|------------------|----------|-----|-----------------|
| TABE-PMDA | +   | +    | +   | +    | +                | +-       | -   | -               |
| TABE-BTDA | ++  | ++   | ++  | ++   | ++               | +-       | -   | -               |
| TABE-6FDA | ++  | ++   | ++  | ++   | ++               | +-       | -   | -               |

Solubility: ++ soluble at room temperature; + soluble on heating; +- partially soluble; - insoluble

Films for mechanical tests were prepared by dissolution of the polymer (50 g/L) in DMAc followed by casting solution on a glass substrate by doctor blading. The removal of solvent was accomplished by heating the polymer with a temperature programming procedure. According to the obtained stress-strain curves, the maximum stress was in the range of 13–30 MPa, and the maximum strain was about 19%–23%.

To evaluate the crystallinity of the polyimides, wide-angle X-ray diffraction patterns of the polyimides in the region of  $2\theta = 5^\circ$ – $70^\circ$  at room temperature were studied and semi-crystalline nature of the polymers was observed. The wide-angle X-ray diffractograms of the polymers are shown in Fig. 1.

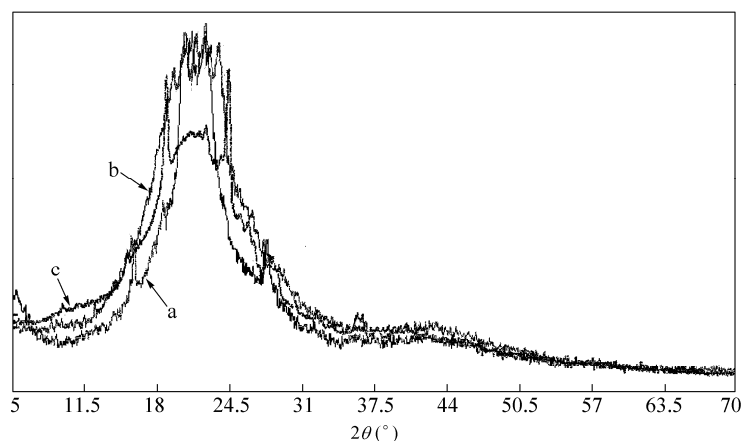


**Fig. 1** X-ray diffractograms of (a) TABE-PMDA, (b) TABE-BTDA and (c) TABE-6FDA

DSC and DMTA techniques were applied to study the thermal behavior of the polyimides. The glass transition temperature ( $T_g$ ) of the polymers was about  $143$ – $172^\circ\text{C}$  (Table 5).  $T_g$  was taken as the midpoint of the change in slope of the baseline in DSC curves, and on the other hand DMTA confirmed the results as  $\tan\delta$  went through a maximum. The DSC curves of the polymers are shown in Fig. 2. Thermogravimetric analysis (TGA) in air at a heating rate of 10 K/min was applied for evaluation of thermal stability. The initial decomposition temperatures ( $T_0$ ) of the polyimides were about  $269$ – $283^\circ\text{C}$ , and the temperatures for 10% gravimetric loss ( $T_{10}$ ) that are important criterion for evaluation of thermal stability were in the range of  $373$ – $394^\circ\text{C}$ , and also the char yields of polymers at  $600^\circ\text{C}$  were about 32%–35% (Fig. 3 and Table 5). Accordingly, the polymers showed reasonable thermal stability that could be attributed to the incorporation of imide units, preformed amide and ester groups, symmetry of structures and suitable increase of molecular weights. Rigid PMDA-derived polyimide showed higher thermal stability in comparison with other polyimides.

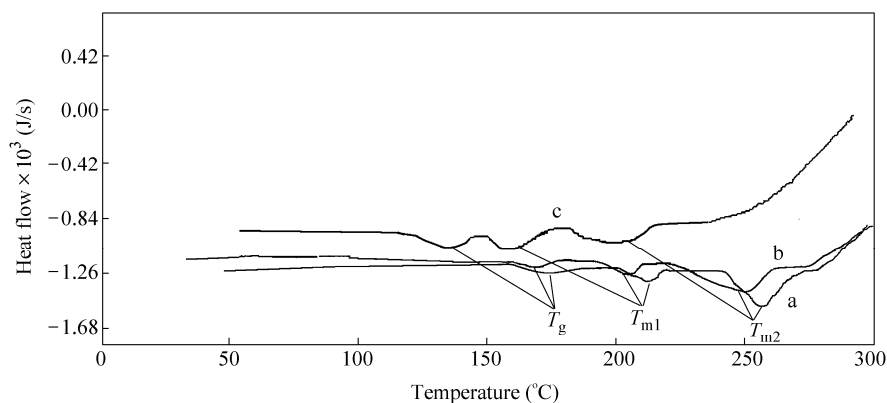


**Fig. 2** DSC curves of (a) TABE-PMDA, (b) TABE-BTDA and (c) TABE-6FDA

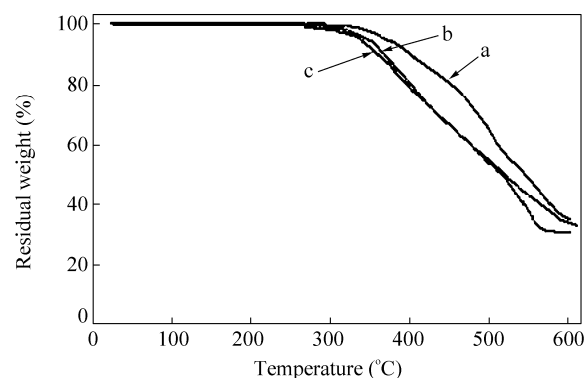
**Table 5.** Thermal analysis data

| Polymer   | $T_g$ (°C) | $T_{m1}$ (°C) | $T_{m2}$ (°C) | $T_0$ (°C) | $T_{10}$ (°C) | $T_{max}$ (°C) | Char yield at 600°C (%) |
|-----------|------------|---------------|---------------|------------|---------------|----------------|-------------------------|
| TABE-PMDA | 172        | 210           | 265           | 283        | 394           | 435            | 35                      |
| TABE-BTDA | 163        | 200           | 255           | 272        | 380           | 418            | 34                      |
| TABE-6FDA | 143        | 165           | 205           | 269        | 373           | 410            | 32                      |

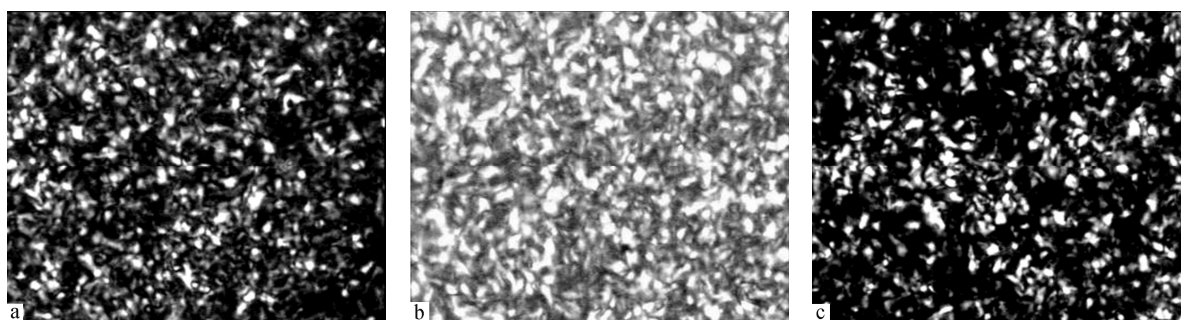
$T_g$ : glass transition temperature;  $T_0$ : initial decomposition temperature;

$T_{m1}$ : first phase transition;  $T_{m2}$ : second phase transition;

$T_{max}$ : maximum decomposition temperature;  $T_{10}$ : temperature for 10% weight loss

**Fig. 3** TGA curves of (a) TABE-PMDA, (b) TABE-BTDA and (c) TABE-6FDA

Crystallization morphology of thermotropic liquid crystalline polyimides was studied by DSC and polarized light microscopy (PLM) with a temperature-controlled hot stage. The polymers were heated to melting at 10 K/min and isothermally kept for 3 min (dark appearance) and the melt was subsequently cooled under control at 10 K/min. Presence of two broad endothermic peaks ( $T_{m1}$  and  $T_{m2}$ ) in DSC curves and their confirmation by PLM was assigned to the liquid crystalline nature of the polymers with the smectic phase (Fig. 4). Similar behaviors were observed for the TABE monomer by studying its crystalline structure using the mentioned methods. Accordingly, it can be concluded that the property of thermotropic liquid crystalline polyimide must result from TABE monomers.

**Fig. 4** Polarized microscope photographs of (a) TABE-PMDA in melt at 230°C, (b) TABE-BTDA in melt at 220°C and (c) TABE-6FDA in melt at 179°C

## CONCLUSIONS

A novel diamine with built-in ester, amide and ether groups (TABE) was prepared: TOBA was synthesized by nucleophilic reaction of 4-hydroxy benzoic acid with TPC and then reaction of TOBA with DADO *via* Yamazaki method yielded TABE. The structures of the intermediate and the diamine were confirmed by FTIR and  $^1\text{H}$ -NMR spectra, and also elemental analyses. Polycondensation reaction of TABE with dianhydrides by *in*

*situ* silylation using TMSCl resulted in preparation of different liquid-crystalline polyimides with good thermal stability, improved solubility and reduced melting and glass transition temperatures.

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