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Synthesis and properties of new polyimides derived from 1,1-bis[4-(4-aminophenoxy)phenyl]cyclododecane

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Abstract

A new diamine, 1,1-bis[4-(4-aminophenoxy)phenyl]cyclododecane (**BAPCD**), bearing flexible ether and bulky pendant cyclododecylidene groups was prepared in three steps from cyclododecanone. The monomer was reacted with six conventional aromatic dianhydrides in *N*,*N*-dimethylacetamide (DMAc) to obtain the corresponding new polyimides via the poly(amic acid) precursors and thermal or chemical imidization. The poly(amic acid)s obtained had inherent viscosities ranging from 1.10–1.42 dl g^{-1}. All the poly(amic acid)s could be cast from DMAc solutions and thermally converted into transparent, flexible, and tough polyimide films, except for polymer $\mathbf{V_a}$ which was derived from pyromellitic dianhydride. X-ray diffractograms of polyimides indicated all polymers were amorphous except the polymer $\mathbf{V_a}$. Polyimides derived from 4,4′-sulfonyldiphthalic anhydride and 4,4′-hexafluoroisopropylidenediphathalic anhydride exhibited excellent solubility in various polar solvents. These polymers showed glass transition temperatures between 259°C and 276°C, and decomposition temperatures at initial and 10% mass loss ranging from 430°C to 483°C and from 504°C to 526°C in nitrogen, respectively. The polyimide films, except $\mathbf{V_a}$, had a tensile strength in the range 76–112 MPa and a tensile modulus in the range 2.1–3.5 GPa.

Keywords: 1,1-bis[4-(4-Aminophenoxy)phenyl]cyclododecane (**BAPCD**); Bulky pendent; Polyimides

1. Introduction

Polyimides have been of great interest in engineering and microelectronics for a number of applications as a result of their unique property combinations [1]. Although exceptional thermal stability is complemented by excellent mechanical and electrical performance, and dimensional stability over a wide range of temperatures, their insolubility in common organic solvents and their high glass and softening temperatures make these systems difficult to process and fabricate. Therefore, much effort has been made to ease their fabrication.

Among them, aromatic polyimides have been presented utilizing aromatic dianhydrides and/or aromatic diamines which were polyphenylated [2–4], or contained fluorine [5,6], chlorine [7], ether [8–13], etc. On the other hand, another successful approach to attain an enhanced solubility is introducing bulky pendent phenyl and alkyl groups into the polymer backbone [14–18].

In recent years, several studies have focussed on adopting polyalicyl dianhydrides [19–21] or diamines containing alicyclic structures [22,23] for the preparation of polyimides that were thermal stable and easier to fabricate. It is of note, that charge transfer usually occurs in polyimides composed of aromatic dianhydrides and aromatic diamines. The formation of intermolecular and intramolecular charge-transfer complexes would be the reason for coloration and poor fabricability. Therefore, incorporating alicyclic units into polyimides backbone eliminates the influence of the charge transfer, and results in a soluble polyimide [19–22]. Choi et al. [23] also reported that the intermolecular chain distance and chain rigidity of the polymer was increased as a result of the bulky pendant alicyclic structure, which restricted the dense packing and free rotation of the polymer chain. Hence, the obtained polymers showed good thermal stability as well as solubility.

According to these research trends, the present investigation deals with the synthesis of a new class of polyimides containing both ether and bulky pendant cyclododecylidene units in the polymer backbone. The polymers were prepared from the new diamine, 1,1-bis[4-(4-aminophenoxy)phenyl]-cyclododecane (**BAPCD**), and conventional aromatic

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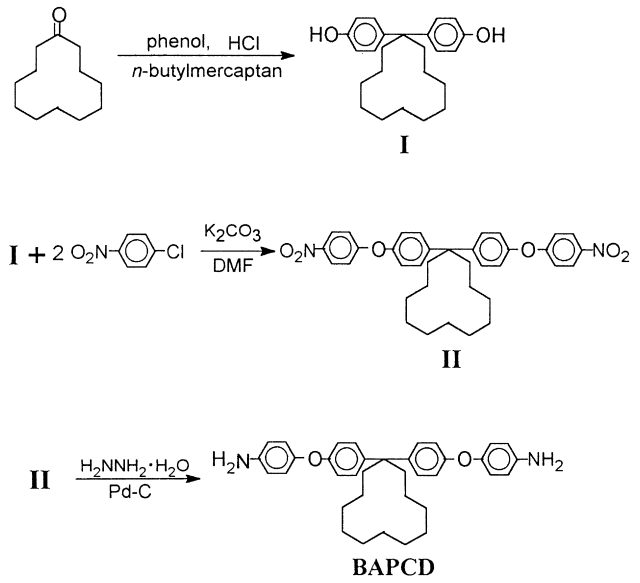


Scheme 1. Synthesis of BAPCD.

dianhydrides. The properties of these polymers will also be investigated herein.

2. Experimental

2.1. Materials

Cyclododecanone (from ACROS), *n*-butylmercaptan (from ACROS), *p*-chloronitrobenzene (from Merck), anhydrous potassium carbonate (from ACROS), hydrazine monohydrate (from ACROS), 10% palladium on activated carbon (Pd-C, from Merck) and acetic anhydride (from Merck) were used as received. Reagent grade aromatic dianhydrides such as pyromellitic dianhydride (**III$_a$**, from CHRISKEV), 3,3′4,4′-biphenyltetracarboxylic dianhydride (**III$_b$**, from CHRISKEV), 4,4′-oxydiphthalic anhydride (**III$_c$**, from TCI), 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (**III$_d$**, from CHRISKEV), 4,4′-sulfonyldiphthalic anhydride (**III$_e$**, from New Japan Chemical Co.), and 4,4′-hexafluoroisopropylidenediphathalic anhydride (**III$_f$**, from CHRISKEV) were recrystallized from acetic anhydride before use. *N*,*N*-Dimethylacetamide (DMAc) and pyridine were vacuum distilled over calcium hydride before use in the polymerization.

2.2. Monomers synthesis

Scheme 1 illustrates the synthesis of 4,4′-cyclododecylidenebisphenol (**I**), 1,1-bis[4-(4-nitrophenoxy)phenyl]-cyclododecane (**II**) and 1,1-bis[4-(4-aminophenoxy)phenyl]-cyclododecane (**BAPCD**).

2.2.1. 4,4′-Cyclododecylidenebisphenol (I)

A flask was charged with a mixture of cyclododecanone (4.7 g, 26 mmol), phenol (16.4 g, 0.17 mol), and *n*-butylmercaptan (0.15 ml). Heat was applied and when the reaction mixture became liquid at 58°C, anhydrous hydrogen chloride was introduced until the solution became saturated. Stirring was continued at 60°C for several hours, during which period white solids began to separate out from the reddish-orange reaction mixture. The solid obtained was filtered off, washed with dichloromethane and dried to afford **I** in 83% yield. It was recrystallized from toluene. m.p. 207°C–209°C. ([24]: 207°C–208.5°C). IR spectrum (KBr) exhibited absorptions at 3200–3300 cm^{-1} (O–H).

2.2.2. 1,1-bis[4-(4-Nitrophenoxy)phenyl]cyclododecane (II)

A mixture of **I** (3.52 g, 0.01 mol), *p*-chloronitrobenzene (13.8 g, 0.022 mol), potassium carbonate (3.31 g, 0.024 mol) and *N*,*N*-dimethylformamide (DMF, 10 ml) was refluxed for 8 h. The mixture was then cooled and poured into methanol. The crude product was recrystallized from DMF to provide yellow needles (m.p. 243°C–245°C) in 89% yield. The IR spectrum (KBr) exhibited absorptions at 1577 and 1331 cm^{-1} (NO_2), 1245 cm^{-1} (C–O–C). ^{1}H-NMR ($CDCl_3$): δ(ppm) = 8.19–8.16 (*d*, 4H, aromatic *ortho* to NO_2 groups); 7.24–7.21 (*d*, 4H, aromatic *ortho* to cyclododecane groups); 7.02–6.96 (*dd*, 8H, *meta* to NO_2 and cyclododecane groups); 2.13–1.00 (*m*, 22H, cyclododecane H). ^{13}C-NMR ($CDCl_3$): δ(ppm) = 163.39, 152.83, 146.64, 143.00, 129.46, 125.91, 119.74, 117.29, 48.34, 33.64, 26.72, 26.41, 22.61, 22.41, 20.27.

ANAL. calcd for $C_{36}H_{38}O_6N_2$: C, 72.70%; H, 6.44%; N, 4.71%. Found: C, 72.16%; H, 6.40%; N, 4.72%.

2.2.3. 1,1-bis[4-(4-Aminophenoxy)phenyl]cyclododecane (BAPCD)

Hydrazine monohydrate (20 ml) was added dropwise to a mixture of **II** (1.19 g, 2 mmol), ethanol (60 ml), and a catalytic amount of Pd–C (0.05 g) at the boiling temperature. The reaction was refluxed for 24 h, and **BAPCD** was precipitated during this period. Enough ethanol was then added to the mixture to dissolve the **BAPCD** and the mixture was filtered to remove Pd–C. After cooling, the precipitated crystals were isolated by filtration and recrystallized from 1,2-dichlorobenzene. The yield was 73%; m.p. 222°C–224°C. The IR spectrum (KBr) exhibited absorptions at 3418, 3346 and 1610 cm^{-1} (N–H), 1230 cm^{-1} (C–O–C). ^{1}H- and ^{13}C-NMR (DMSO-*d*6) were shown in Fig. 1(A) and (B), respectively.

ANAL. calcd for $C_{36}H_{42}O_2N_2$: C, 80.86%; H, 7.92%; N, 5.24%. Found: C, 80.56%; H, 7.89%; N, 5.34%

2.3. Polymer synthesis

The general procedure for polymerization was as follows. To a stirred solution of **BAPCD** (0.588 g, 1.1 mmol) in DMAc (7 ml), was gradually added **III** (0.392 g, 1.1 mmol). The mixture was stirred at room temperature for 2–4 h under an argon atmosphere to form the poly(amic acid) with an inherent viscosity of 1.38 dl g^{-1}, measured at a concentration of

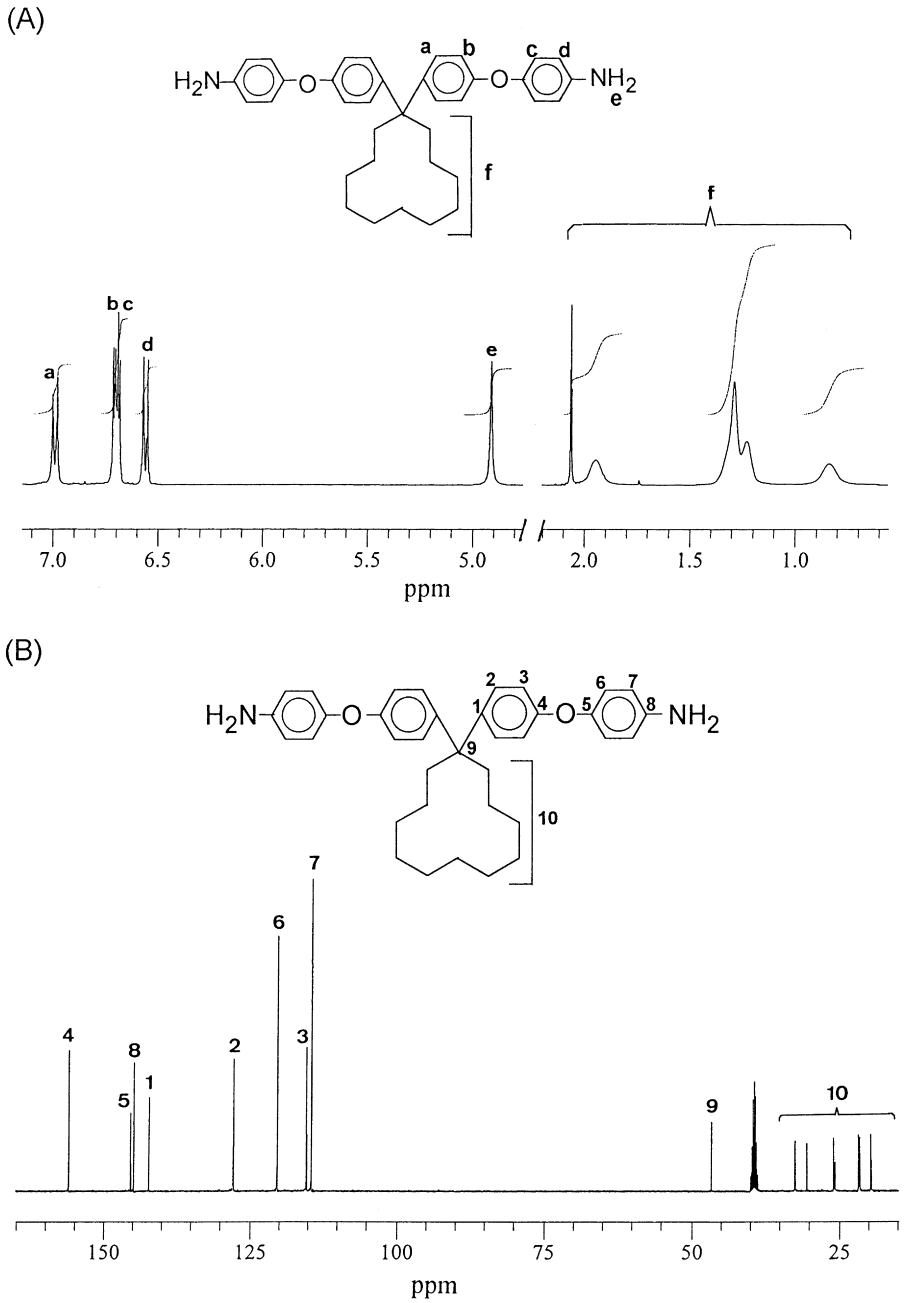
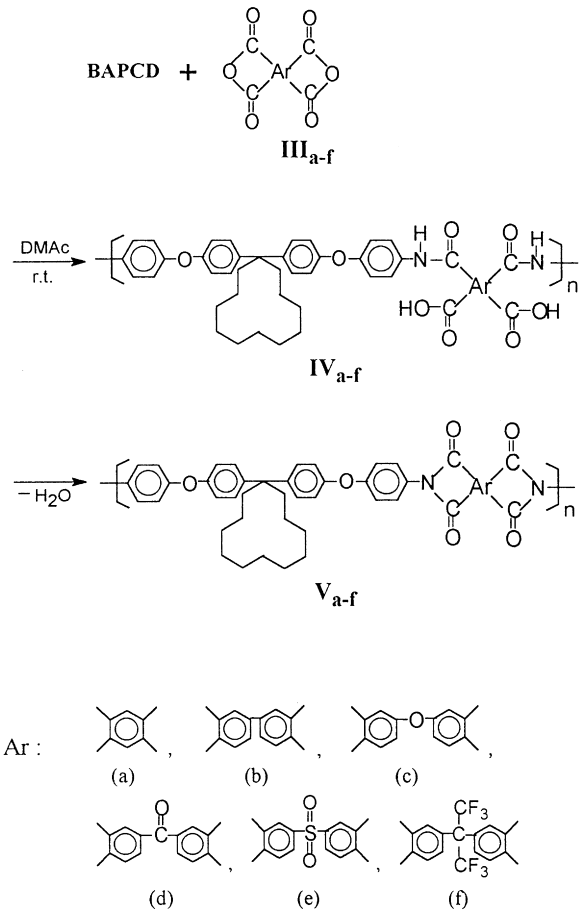


Fig. 1. NMR spectra of 1,1-bis[4-(4-aminophenoxy)phenyl]-cyclododecane, using DMSO-d_6 as solvent: (A) ^{1}H-NMR; and (B) ^{13}C-NMR.

0.5 g dl^{-1} in DMAc at 30°C. From the poly(amic acid) solution, a film was cast onto a glass plate and heated (8 h at 80°C, 2 h at 150°C, 2 h at 200°C, 3 h at 250°C, 2 h at 300°C) under vacuum to convert the poly(amic acid) into a polyimide film. Chemical imidization was also carried out by adding DMAc, and an equimolar mixture of acetic anhydride and pyridine, into the aforementioned poly(amic acid) solution, stirring at room temperature for 1 h and then heating at 100°C for 2 h. It was subsequently poured into methanol and the brown solid precipitate was filtered off, washed with methanol and hot water, and dried to afford **V$_e$**. ^{1}H-NMR (DMSO-d_6): δ(ppm) = 8.61 (*m*, 4H, aromatic *ortho* and *meta* to SO_2 group); 8.17 (*d*, 2H, aromatic *ortho* to SO_2 group); 7.37 (*d*, 4H *ortho* to cyclododecane group), 7.18 (*d*, 4H, *meta* to cyclododecane group); 7.08 (*d*, 4H, *meta* to imide group); 6.96 (*d*, 4H, *ortho* to imide group); 2.13–1.00 (*m* 22H, cyclododecane H). Other polymers were synthesized by an analogous procedure.

2.4. Measurement

Melting points were measured in capillaries on a Büchi apparatus (Model BUCHI 535). IR spectra were recorded in the range 4000–400 cm^{-1} for the synthesized monomers and polymers on a JASCO IR-700 spectrometer. The inherent viscosities of all polyimides were measured using an



Scheme 2. Preparation of the polyimides.

Ubbelohde viscometer. Elemental analysis was made (Perkin-Elmer 2400 instrument). Wide-angle X-ray diffraction patterns were performed at room temperature with film specimens on an X-ray diffractometer (Philips model PW 1710) using Ni filtered Cu-Kα radiation (35 kV, 25 mA). The scanning rate was 3° min^{-1}. Thermogravimetric data were obtained on a Du Pont 2100 in flowing nitrogen (60 cm^3 min^{-1}) at a heating rate of 20°C min^{-1}. Differential scanning calorimetry (DSC) analysis was performed on a DuPont 2100 differential scanning calorimeter. Tensile properties were determined from stress–strain curves obtained with a Orientec Tensilon with a load cell of 10 kgf. A gauge of 3 cm and a strain rate of 2 cm min^{-1} were used for this study. Measurements were performed at room temperature with film specimens (0.5 cm wide, 6 cm long, and ca. 0.1 mm thick).

3. Results and discussion

3.1. Monomer synthesis

The diamine 1,1-bis[4-(4-aminophenoxy)phenyl]cyclododecane (**BAPCD**) was synthesized in three steps from cyclododecanone. The diphenol compound **I** was synthesized from the reaction of cyclododecanone with excess phenol in the presence of hydrogen chloride as catalyst and *n*-butylmercaptan as cocatalyst. The dinitro compound **II** was prepared by nucleophilic substitution reaction from **I** with *p*-chloronitrobenzene in the presence of potassium carbonate. The catalytic hydrogenation of compound **II** to the diamine **BAPCD** was accomplished by means of hydrazine monohydrate as well as a catalytic amount of Pd–C. Recrystallization from 1,2-dichlorobenzene gave high purity. The IR spectrum of **II** showed nitro group characteristic absorptions at 1577 and 1331 cm^{-1}. After hydrogenation, the characteristic absorptions, as a result of the nitro group, were absent and new absorptions at 3418 and 3346 (N–H stretching) and 1610 cm^{-1} (N–H deformation) appeared. Moreover, the structure of diamine **BAPCD** was further identified by NMR spectroscopy as shown in Fig. 1. These results clearly confirm that the diamine prepared herein is consistent with the proposed structure.

3.2. Preparation of polyimides

Polyimides were prepared by the conventional two-step polymerization method, as shown in Scheme 2, involving ring-opening polyaddition forming poly(amic acid) and subsequent thermal or chemical cyclodehydration. The poly(amic acid)s **IV$_{a–f}$** were prepared by adding the dianhydride to the diamine solution. The inherent viscosities of all the poly(amic acid)s are summarized in Table 1. The

Table 1
Preparation of polyimides

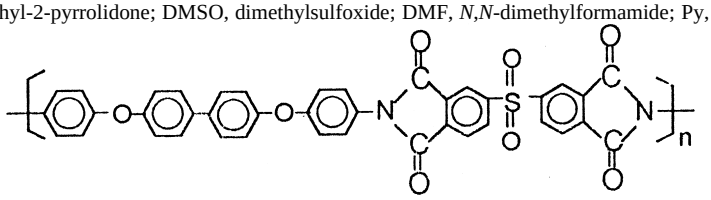
Dianhydride code	Poly(amic acid) Code	η^{a}_{inh} (dl g^{-1})	Polyimide Code	η^{b}_{inh} (dl g^{-1})	Film quality[c]
III$_a$	**IV$_a$**	1.42	**V$_a$**	0.79	Brittle
III$_b$	**IV$_b$**	1.21	**V$_b$**	0.69	Tough
III$_c$	**IV$_c$**	1.10	**V$_c$**	0.66	Tough
III$_d$	**IV$_d$**	1.24	**V$_d$**	0.68	Tough
III$_e$	**IV$_e$**	1.38	**V$_e$**	0.77[a]	Tough
III$_f$	**IV$_f$**	1.39	**V$_f$**	0.74[a]	Tough

[a]Measured in DMAc at a concentration of 0.5 g dl^{-1} at 30°C.
[b]Inherent viscosity of polyimide obtained by chemical imidization was measured in concentrated sulfuric acid at a concentration of 0.5 g dl^{-1} at 30°C.
[c]Polymer films was obtained by thermal imidization from the corresponding poly(amic acid)s.

Table 2
Solubility of polyimides[a]

Solvent[b]								
Polymer code	NMP	DMAc	DMF	DMSO	*r*-butyrolactone	Py	THF	Conc. H_2SO_4
V_a	−	−	−	−	−	−	−	+
V_b	+ −	−	−	−	−	−	−	+
V_c	+ −	−	−	−	−	−	−	+
V_d	−	−	−	−	−	−	−	+
V_e	+	+	+	+	+	+	+	+
V_f	+	+	+	+	+	+	+	+
Ref[c]	−	−	−	−	−	−	−	+

[a]Solubility: +, soluble at room temperature; +−, partial soluble; −, insoluble.
[b]Abbreviations: NMP, *N*-methyl-2-pyrrolidone; DMSO, dimethylsulfoxide; DMF, *N*,*N*-dimethylformamide; Py, pyridine.
[c]Analogous polyimide, **Ref** .



inherent viscosities of the poly(amic acid)s were in the range of 1.10–1.42 dl g^{-1}. The molar masses were high enough to cast flexible, tough, and transparent poly(amic acid) films. These films were further heated up to 300°C to produce flexible and tough polyimide films, except for polymer **V_a**. Alternatively, chemical imidization of poly(amic acid)s by adding dehydrating agents, acetic anhydride and pyridine, was also effective in obtaining polyimides. The resulting polyimides had inherent viscosities of 0.66–0.79 dl g^{-1} (Table 1).

The identification of imidization of poly(amic acid)s to polyimides was made by IR spectra. Fig. 2 shows the typical IR spectra of poly(amic acid) film **IV_c** and the corresponding polyimide film **V_c** obtained by thermal cyclodehydration. The poly(amic acid) **IV_c** showed major absorption peaks at 1536 and 1714 cm^{-1} as a result of the amide and acid carbonyl groups, respectively, and characteristic absorptions around 3266 cm^{-1} for N–H and O–H groups. In the spectrum of the polyimide **V_c**, the characteristic absorptions of amic acid at 1536 and 3266 cm^{-1} disappeared. Further, the imide group showed characteristic bands at 1769 and 1715 cm^{-1} (asymmetric and symmetric

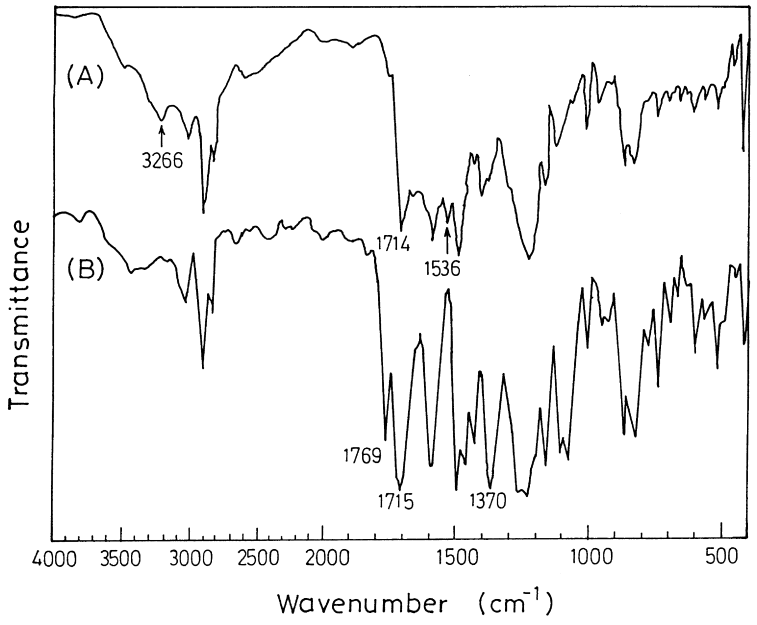


Fig. 2. Fig. 2IR spectra of (A) poly(amic acid) **IV_c**, and (B) corresponding polyimide **V_c**.

Table 3
Thermal properties of polyimides

Polymer code	T_g[a] (°C)	IDT[c] (°C)	PDT[c] (°C)	Yc[d] (%)
$\mathbf{V_a}$	—[b]	467	525	61
$\mathbf{V_b}$	273	483	526	58
$\mathbf{V_c}$	261	450	518	57
$\mathbf{V_d}$	259	442	519	54
$\mathbf{V_e}$	274	430	504	50
$\mathbf{V_f}$	276	450	515	54

[a]From DSC measurements conducted at a heating rate of 20°C min^{-1}.
[b]No T_g was observed.
[c]Initial decomposition temperature (IDT) and temperature at 10% mass loss (PDT) were determined by TG in nitrogen at a heating rate of 20°C min^{-1}.
[d]Char yield at 800°C in nitrogen.

stretches of carbonyl group of imide, respectively.) The C–N band at 1370 cm^{-1} also confirmed the formation of the imide structure. On the other hand, the structure of polyimide were also confirmed by elemental analysis. It was observed that the elemental composition of the polymer were almost in agreement with the calculated values for the proposed structures. Among these polymers, the structure of polymer $\mathbf{V_e}$ was further confirmed by ^{1}H-NMR (see Section 2).

3.3. Polymer properties

The solubility of these polyimides obtained by chemical cyclodehydration was tested qualitatively in various solvents. The results are shown in Table 2. Polyimides $\mathbf{V_a}$–$\mathbf{V_d}$ were almost insoluble in organic solvents, but dissolved in concentrated sulfuric acid. Polyimides $\mathbf{V_e}$ and $\mathbf{V_f}$, containing sulfone and hexafluoroisopropylidene linkages, exhibit excellent solubility toward test solvents. They dissolved in a variety of solvents such as *N*-methyl-2-pyrrolidone (NMP), DMAc, *N*,*N*-dimethylformamide (DMF), dimethylsulfoxide (DMSO), *r*-butyrolactone, pyridine and even in tetrahydrofuran at room temperature. For comparison, polyimide **Ref** containing biphenylene without pendant cyclododecylidene was further prepared from 4,4′-bis(4-aminophenoxy)biphenyl with dianhydride $\mathbf{III_e}$. It had been observed that polyimide **Ref** was nearly insoluble in the test solvents, and showed lower solubility than its analogous polymer $\mathbf{V_e}$. It may be as a result of the presence of the bulky pendant group which decreases the inter-chain interaction of rigid aromatic repeating units, resulting in improved solubility [25].

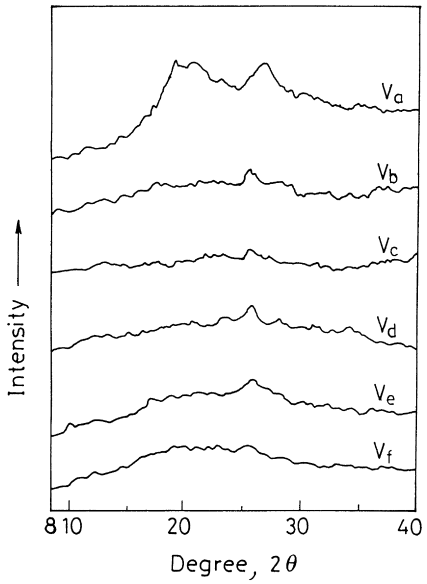


Fig. 3. Wide-angle X-ray diffractograms of polyimides.

An attempt was made to estimate the crystallinity of the polyimides by means of X-ray diffractograms. Fig. 3 presents the wide-angle X-ray diffractograms of the polyimide films obtained by thermal cyclodehydration. It was observed that polymer $\mathbf{V_a}$ containing pyromellitimide displayed two medium to strong reflection peaks around $2\theta = 20°$ and 27°, indicating higher crystallinity. The other polymers exhibited an amorphous nature even for polymers derived from rigid dianhydrides $\mathbf{III_b}$ and $\mathbf{III_d}$. These results could be explained by the presence of the bulky-pendant group inhibited close-packing of the polymer chains, leading to a decrease in crystallinity.

The thermal properties of the polyimides are tabulated in Table 3. Glass transition temperatures (T_gs) of polymers, determined by means of differential scanning calorimeter (DSC), were found to be in the range of 259°C–276°C. No glass transition was observed from DSC curves for polymer $\mathbf{V_a}$ up to temperatures around 350°C. The highest T_g value was observed for the hexafluoroisopropylidene-containing polymer $\mathbf{V_f}$. The initial decomposition temperature (IDT) and temperature at 10% mass loss (PDT), examined by thermogravimetric (TG) analysis, showed values reaching 430°C–483°C and 504°C–526°C, respectively, in nitrogen atmosphere. It implied that these polyimides showed good thermal stability irrespective of introducing aliphatic group. The residual masses in nitrogen were also determined by means of TGA. The char yeild ranged from 50%–61% at 800°C.

The tensile properties of the polyimide films prepared by thermal imidization are summarized in Table 4. The polymer films, except $\mathbf{V_a}$, had tensile strength of 76–112 MPa,

Table 4
Tensile properties of polyimides

Polymer code	Tensile strength (MPa)	Elongation at break (%)	Tensile modulus (GPa)
$\mathbf{V_a}$	-[a]	-[a]	-[a]
$\mathbf{V_b}$	100	5	2.6
$\mathbf{V_c}$	98	4	2.7
$\mathbf{V_d}$	84	3	3.4
$\mathbf{V_e}$	76	3	3.5
$\mathbf{V_f}$	112	5	2.1

[a]Polymer film was too brittle to measured.

elongation at break of 3%–5%, and tensile modulus of 2.1–3.5 GPa. Most polymer films ehibited high tensile strength, high modulus and low elongation; thus they could be considered as hard and strong materials.

4. Conclusions

In this study the new diamine **BAPCD** containing ether and bulky pendant cyclododecylidene units was successfully synthesized with high yield and purity. A series of polyimides derived from **BAPCD** were prepared via two-step method. The investigation of the properties of the polymers revealed that the introduction of bulky pendant cyclododecylidene groups into the polymer backbone resulted in polyamides with enhanced solubility, reduced crystallinity, as well as satisfactory thermal stability.

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