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Synthesis and Characterization of Soluble Polyimides Containing Cyclohexylidene Moiety with Various Alkyl-Substituents

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SYNTHESIS AND CHARACTERIZATION OF SOLUBLE POLYIMIDES CONTAINING CYCLOHEXYLIDENE MOIETY WITH VARIOUS ALKYL-SUBSTITUENTS

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Key Words: Soluble Polyimides, Alkyl-Substituents on the Cyclohexane Ring, a HCl-Catalyzed Condensation Reaction, Good Solubility, High Glass Transition Temperature, Thermal Stability, One-Step Polymerization

ABSTRACT

A series of novel aromatic diamines containing kinked cyclohexylidene moieties was synthesized by a HCl-catalyzed condensation reaction of excess aniline and corresponding alkyl-substituted cyclohexanone derivatives at 120–140°C for 24 hours. The structures of monomers were identified by ¹H-NMR, ¹³C-NMR, and FT-IR. The yields of monomer preparation were above 70%. The polymers were synthesized from the obtained diamines and various aromatic dianhydrides by one-step polymerization in *m*-cresol at 200°C for 6–8 hours. The inherent viscosities were in the

range of 0.74-1.66 dL/g and the prepared polyimides showed excellent thermal stabilities as well as good solubility. That is, all polymers were readily soluble in common organic solvents such as dimethylacetamide, dimethylformamide, tetrahydrofuran, chloroform, etc. and the glass transition temperatures were in the range of 261-348°C. The solubility and the glass transition temperature of the polymers increased as the bulkiness of the alkyl-substituents increased.

INTRODUCTION

Aromatic polyimides have been known for their high temperature stability, low dielectric properties, and chemical resistance. However, the processability of these polymers is poor due to their infusible and insoluble characters, which makes them difficult to process [1-2]. Therefore, various approaches have been devoted to improve the processability and solubility without sacrificing their excellent properties. The introduction of alicyclic monomers or monomers containing bulky substituents to a polymer backbone have been found to be an effective approach to improve the solubility of polyimides [3-12]. We have previously reported the synthesis of polyamideimides with good solubility as well as high thermal stability by the introduction of an alicyclic diamine containing the 3,3,5-trimethyl cyclohexane structure. And, we had found that the introduction of cyclohexane moiety with trimethylsubstituents along the polymer backbone decreased the intermolecular interactions, which resulted in the improvement of the solubility. At the same time, the rigidity of the polymer chain was also increased due to the incorporation of three methylsubstituents, which prevented the free rotation of the polymer chain [13-14]. As a continuation of this work, the present work was undertaken to synthesize and characterize new soluble polyimides from aromatic diamines containing a cyclohexane moiety with alkyl-substituents. The alkyl-substituents on the cyclohexane ring were varied in order to study the effects of the substituents on the solubility and thermal stability of the polymers.

EXPERIMENTAL

Starting Materials

m-Cresol (Aldrich Chemical Co., Inc., 99%, bp; 203°C) was freshly distilled under reduced pressure. Aniline (Aldrich Chemical Co., Inc., 99%, bp; 184°C), 4-ethylcyclohexanone (Tokyo Chemical Industry Co., Ltd., >98%), 4-*t*-butyl-

cyclohexanone (Tokyo Chemical Industry Co., Ltd., >98%, mp; 50°C) and 4-n-pentylcyclohexanone (Tokyo Chemical Industry Co., Ltd., >90%) were used as received. Pyromellitic dianhydride (PMDA, 99.2%, Daicel Chemical Industry, Ltd.), 3,3',4,4'-benzophenonetetracarboxylic dianhydride (BTDA, 98.6%, Daicel Chemical Industry, Ltd.), and 3,3',4,4'-biphenyltetracarboxylic dianhydride (BPDA, 99.6%, Mitsubishi Kasei) were used without further purification, because they were of a highly purified grade for polyimide synthesis. 2,2-Bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride (HFDA, Tokyo Chemical Industry Co., Ltd., >98%) and 3,3',4,4'-tetracarboxydiphenyl oxide dianhydride (ODPA, Tokyo Chemical Industry Co., Ltd., >98%) were used as received. 1,4-bis(3,4-dicarboxyphenoxy) benzene dianhydride (HQDPA) was prepared from 4-chlorophthalic anhydride.

Measurements

All NMR spectra were taken on a Bruker AMX 500 using CDCl₃ or dimethyl sulfoxide-d₆ (DMSO-d₆) as a solvent. Infrared spectra were obtained with a Bio-Rad Digilab Division FTS-165 FT-IR spectrophotometer. Elemental analysis were measured with FISON'S Instruments EA-1108. The inherent viscosities of polymers were measured with a Ubbelohde viscometer at a concentration of 0.5g/dL in *m*-cresol at 30°C. The melting points of the prepared monomers were determined by DSC. Thermogravimetric analysis (TGA) was done with a Perkin Elmer TGA 7 at heating rate of 20°C/min under nitrogen atmosphere. Differential scanning calorimeter (DSC) analysis was performed on a Du Pont model 910.

Monomer Synthesis

Novel aromatic diamines were synthesized by a HCl-catalyzed condensation reaction of corresponding 4-alkyl-cyclohexanone derivatives with excess aniline in an autoclave at temperature of 120-140°C or 24 hours. A typical synthetic route is as follows.

1,1-Bis(4-aminophenyl)-4-ethylcyclohexane (BAET)

To a solution of 4-ethylcyclohexane (50.0g, 0.4 mol) in 140 ml of 35% aqueous HCl solution in a 1L autoclave equipped with a mechanical stirrer was added excess aniline (155.3g, 1.68 mol) and the mixture was stirred at 125°C for 20 hours. After cooling, the solution was made basic to pH 10 with aqueous NaOH solution and the oily layer was separated and distilled to remove the unreacted aniline. The residual crude product was recrystallized from ethyl alcohol to afford 95.4 g (83.1% yield) of white crystal: mp 134°C; ¹H-NMR δ (CDCl₃) 0.84 (3H, t,

CH₃), 1.06-1.89 (9H, m), 2.49, 2.54 (2H, bs), 3.50 (4H, s, amine protons), 6.50-7.24 (8H, m, phenyl ring protons) ppm; ¹³C-NMR δ (CDCl₃) 11.54, 29.18, 29.48, 36.87, 39.43, 44.75 (aliphatic carbons), 114.78, 115.15, 127.02, 128.71, 136.06, 142.54, 143.41, 143.50 (aromatic carbons) ppm; FT-IR (KBr pellet) 1226 (C-N stretching), 1618 (N-H deformation), 3362, 3446 (N-H stretching) cm⁻¹; Elemental Analysis Calcd. for C₂₀H₂₆N₂: C, 81.63; H, 8.85; N, 9.52, Found: C, 81.41; H, 9.10; N, 9.49%.

1,1-Bis(4-aminophenyl)-4-butylcyclohexane (BABU)

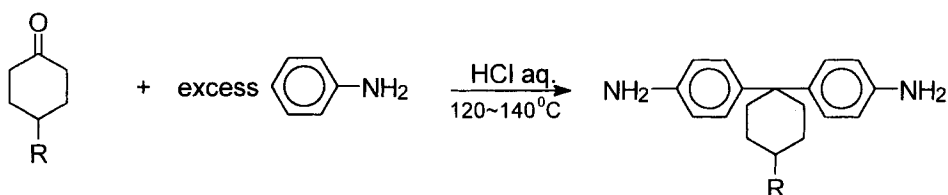
BABU was prepared in the similar manner to that of BAET with 4-butylcyclohexane instead of 4-ethylcyclohexanone to give a white crystal by recrystallization from isopropanol: yield, 81%; mp 141.0°C; ¹H-NMR δ (CDCl₃) 0.76 (9H, s, CH₃), 1.10-1.85 (8H, m), 2.56, 2.60 (2H, bs), 3.51 (4H, s, amine protons), 6.51-7.26 (8H, m, phenyl ring protons) ppm; ¹³C-NMR δ (CDCl₃) 23.80, 27.48, 32.30, 37.64, 44.43, 48.34 (aliphatic carbons), 114.76, 115.20, 126.95, 128.74, 135.87, 142.91, 143.31, 143.48 (aromatic carbons) ppm; FT-IR (KBr pellet) 1244 (C-N stretching), 1625 (N-H deformation), 3352, 3429 (N-H stretching) cm⁻¹; Elemental Analysis Calcd. for C₂₂H₃₀N₂: C, 81.99; H, 9.32; N, 8.69, Found: C, 81.76; H, 9.56; N, 8.68%.

1,1-Bis(4-aminophenyl)-4-amylicyclohexane (BAAM)

BAAM was prepared in a similar manner to that of BAET with 4-amylicyclohexane instead of 4-ethylcyclohexane to give a white crystal by recrystallization from isopropanol: yield, 81%; mp 98.0°C; ¹H-NMR δ (CDCl₃) 0.70 (9H, s, CH₃), 0.77 (2H, t), 1.15-1.86 (9H, m), 2.56, 2.59 (2H, bs), 3.50 (4H, s, amine protons), 6.51-7.24 (8H, m, phenyl ring protons) ppm; ¹³C-NMR δ (CDCl₃) 8.33, 23.54, 23.61, 24.48, 32.74, 34.82, 37.99, 44.78, 45.84 (aliphatic carbons), 114.97, 115.27, 127.18, 129.00, 136.15, 143.20, 143.55, 143.72 (aromatic carbons) ppm; FT-IR (KBr pellet) 1270 (C-N stretching), 1623 (N-H deformation), 3329, 3401 (N-H stretching) cm⁻¹; Elemental Analysis Calcd. for C₂₃H₃₂N₂: C, 82.14; H, 9.52; N, 8.34, Found: C, 81.89; H, 9.77; N, 8.34%.

Polymer Synthesis

To a solution of BABU (2.66 mmol, 0.86g) in 14 mL of freshly distilled *m*-cresol, HQDPA (2.66 mmol, 1.07 g) and isoquinoline (0.95mL) as a catalyst were added at room temperature under nitrogen atmosphere. The reaction mixture was heated to 70-80°C over 2 hours and kept at that temperature for 2 hours. The solution temperature was then slowly raised to 200°C over 2 hours and refluxed for



Where R is CH_3CH_2- , $(\text{CH}_3)_3\text{C}-$, $(\text{CH}_3)_2\text{CHCH}_2\text{C}-$

Scheme 1. Synthesis of Monomers

6 hours. The polymerization was performed under a gentle nitrogen stream to remove the water produced during imidization. The resulting reaction mixture was cooled to room temperature and then poured into excess methyl alcohol. The precipitated polymer was extracted by methyl alcohol in soxhlet extraction apparatus and dried at 100°C for 12 hours *in vacuo*. FT-IR (KBr pellet) 3061, 2949, 1780 and 1726 ($\nu_{\text{C=O}}$), 1379 ($\nu_{\text{C=N}}$), 727 (imide ring deformation) cm^{-1} .

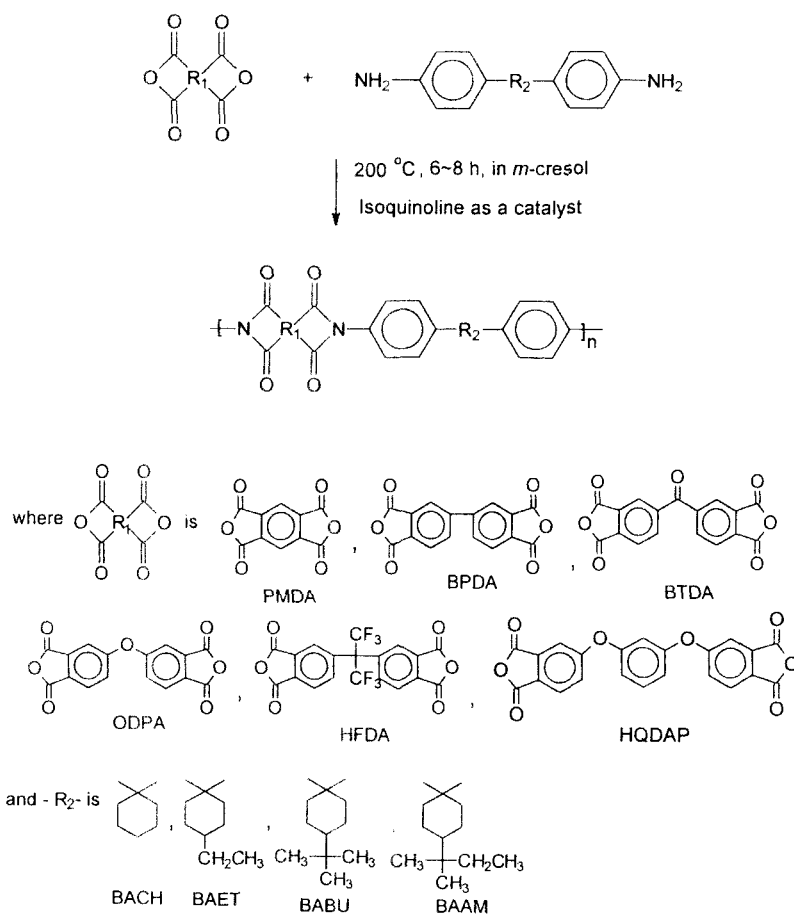
RESULTS AND DISCUSSION

Monomer Synthesis

The synthetic route of the monomers is shown in Scheme 1. Three aromatic diamines containing the cyclohexylidene moiety with different alkyl-substituents were synthesized by a HCl-catalyzed condensation reaction of the corresponding cyclohexanone derivatives with excess aniline in an autoclave at a temperature of 120-140°C for 24 hours. The yields of the monomer preparation were always above 70%. The structures of prepared monomers were characterized by FT-IR, ¹H- and ¹³C-NMR spectroscopy, and elemental analysis.

Polymer Synthesis

From our previous results [15], it was shown that the incorporation of methyl-substituted cyclohexylidene groups improved the solubility of the resulting polyimides. Thus, in this study we have performed the one-step polyimidization in *m*-cresol at 200°C as shown in Scheme 2. Depending on the structure of dianhydride, the solution concentration were varied from 10% to 20%. That is, the concentration had been kept low for the polymerization of PMDA with rigid structure to prevent the gel formation. The results of the polymerization are summarized in



Scheme 2. Synthesis of Soluble Polyimides

Table 1. The inherent viscosities measured in *m*-cresol at 30°C ranged from 0.74 to 1.66 dL/g. The structures of the polyimides were characterized by FT-IR and ¹H- and ¹³C-NMR spectroscopy. The FT-IR spectra of all polyimides exhibit the characteristic imide absorptions band in the regions of 1782, 1720 (ν C=O asym. and sym. stretching, respectively), 1380-1350 (C-N stretching), and 750-720 (imide ring deformation) cm⁻¹. A typical ¹H-NMR spectrum of P-3(d) is shown in Figure 1.

Polymer Characterization

Solubility

The polyimides synthesized were expected to exhibit an enhanced solubility in common organic solvents due to the presence of the cyclohexylidene moiety with

TABLE 1. Preparation of Soluble Polyimides by One-Step Polymerization

Polymers	Monomers		Inherent Viscosity (dL/g) ^a
	Diamines ^b	Dianhydrides ^c	
P-1(a)	BAET	PMDA	1.27
P-1(b)	BAET	BPDA	1.26
P-1(c)	BAET	BTDA	1.23
P-1(d)	BAET	HFDA	0.92
P-1(e)	BAET	ODPA	1.25
P-1(f)	BAET	HQDAP	1.57
P-2(a)	BABU	PMDA	0.93
P-2(b)	BABU	BPDA	1.01
P-2(c)	BABU	BTDA	0.95
P-2(d)	BABU	HFDA	0.91
P-2(e)	BABU	ODPA	0.81
P-2(f)	BABU	HQDAP	1.51
P-3(a)	BAAM	PMDA	1.24
P-3(b)	BAAM	BPDA	1.66
P-3(c)	BAAM	BTDA	1.14
P-3(d)	BAAM	HFDA	1.00
P-3(e)	BAAM	ODPA	1.01
P-3(f)	BAAM	HQDAP	0.74
R-1(a) ^d	BACH	PMDA	- ^e

^a Measured in *m*-cresol solution of 0.5 g/dL at 30 °C.

^b Abbreviations: BACH; 1,1-Bis(4-aminophenyl)cyclohexane, BAET; 1,1-Bis(4-aminophenyl)-4-ethylcyclohexane, BABU; 1,1-Bis(4-aminophenyl)-4-n-butylcyclohexane, BAAM; 1,1-Bis(4-aminophenyl)-4-amylicyclohexane.

^c Abbreviations : PMDA; pyromellitic dianhydride, BTDA ; 3,3',4,4'-benzophenonetetracarboxylic dianhydride, BPDA ; 3,3',4,4'-biphenyltetracarboxylic dianhydride, ODPA ; 3,3',4,4'-tetracarboxy-diphenyl oxide dianhydride, HFDA; 2,2-bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride, HQDAP; 1,4-bis(3,4-dicarboxyphenoxy) benzene dianhydride.

^d Prepared in our previous study¹⁵

^e Insoluble

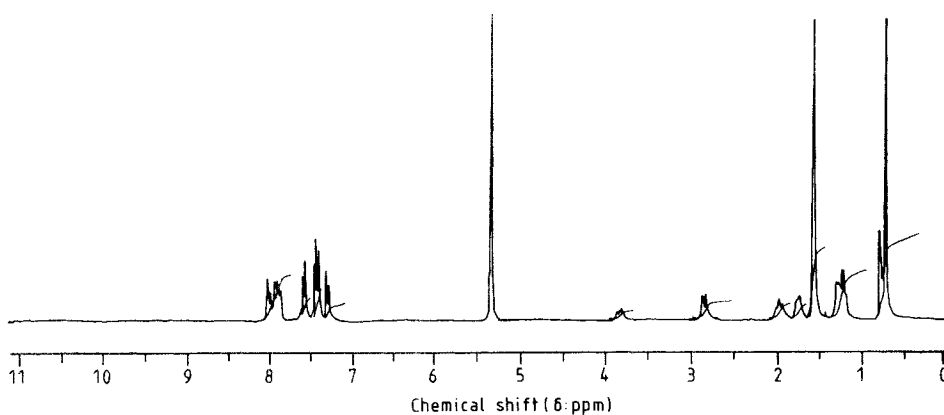


Figure 1. 300M Hz ^1H -NMR spectrum of P-3(d) based on BAAM and HFDA

a bulky-substituent. Table 2 shows the solubility of polyimides [P-1(a)-P-3(f)] synthesized in this study and that of polyimide [R-1(a)] from our previous result [15]. As shown in Table 2, the solubility of polymers was improved by the incorporation of alkyl groups and increased with the increase of the bulkiness of the substituent group. All of these polymers were soluble in *m*-cresol, N-methyl-2-pyrrolidone, dimethylacetamide, and tetrachloroethane at room temperatures.

Thermal Property

The thermal properties of polyimides were investigated by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) under a nitrogen atmosphere. The results are summarized in Table 3. The glass transition temperatures (T_g 's) of polyimides ranged from 261 to 348°C, which was higher than those of polyimides, R-1(a)-(f) prepared from the diamine lacking a substituted cyclohexylidene moiety. That is, the T_g 's were increased by the introduction of alkyl-substituents onto cyclohexane ring. However, it was difficult to determine the relationship between T_g 's and the size of the alkyl substituents. The typical DSC thermograms and TGA curves are shown in Figures 2 and 3, respectively. As shown in Figure 3, The initial decomposition temperature of polyimides were more than 400°C and the residual weight at 800°C were above 20%.

CONCLUSION

A series of polyimides was synthesized from the aromatic diamines containing kinked cyclohexylidene moiety, which was prepared by a HCl-catalyzed

TABLE 2. Solubility of Polyimides Based on PMDA

Polymers	Monomers		Solvents ^a								
	Diamines	Dianhydrides	NMP	DMSO	TCE	Dioxane	THF	CHCl ₃	CH ₂ Cl ₂	Acetone	
P-1(a)	BAET ^b	PMDA ^c	+	+	+	+	+	+	+	—	
P-2(a)	BABU	PMDA	+	+	+	+	+	+	+	—	
P-3(a)	BAAM	PMDA	+	+	+	+	+	+	+	—	
R-1(a) ^d	BACH	PMDA	—	—	+	—	—	—	—	—	

^a Abbreviations : NMP, N-methyl-2-pyrrolidone, DMF, dimethylformamide, TCE; 1,1,2,2-tetra chloro ethane
^b Abbreviations: BACH; 1,1-Bis(4-aminophenyl)cyclohexane, BAET; 1,1-Bis(4-aminophenyl)-4-ethyl- cyclohexane, BABU; 1,1-Bis(4-aminophenyl)-4-n-butylcyclohexane, BABU; 1,1-Bis(4-aminophenyl)-4- amylcyclohexane.

^c Abbreviations : PMDA; pyromellitic dianhydride, BTDA ; 3,3',4,4'-benzophenonetetracarboxylic dianhydride, BPDA ; 3,3',4,4'-biphenyltetracarboxylic dianhydride, ODPA ; 3,3',4,4'-tetracarboxy- diphenyl oxide dianhydride, HFDA; 2,2-bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride, HQDAP; 1,4-bis(3,4-dicarboxyphenoxy) benzene dianhydride.

^d Polyimide prepared in our previous study.¹⁵

TABLE 3. Thermal Properties of the Resulting Polyimides

Polymers	Monomers		T _g (°C) ^c
	Diamines	Dianhydrides	
P-1(a)	BAET ^a	PMDA ^b	-
P-1(b)	BAET	BPDA	336
P-1(c)	BAET	BTDA	-
P-1(d)	BAET	HFDA	-
P-1(e)	BAET	ODPA	-
P-1(f)	BAET	HQDAP	-
P-2(a)	BABU	PMDA	-
P-2(b)	BABU	BPDA	331
P-2(c)	BABU	BTDA	318
P-2(d)	BABU	HFDA	319
P-2(e)	BABU	ODPA	310
P-2(f)	BABU	HQDAP	276
P-3(a)	BAAM	PMDA	348
P-3(b)	BAAM	BPDA	342
P-3(c)	BAAM	BTDA	318
P-3(d)	BAAM	HFDA	310
P-3(e)	BAAM	ODPA	298
P-3(f)	BAAM	HQDAP	261
R-1(a)	BACH	PMDA	-
R-1(b)	BACH	BPDA	-
R-1(c)	BACH	BTDA	305
R-1(d)	BACH	HFDA	293
R-1(e)	BACH	ODPA	290
R-1(f)	BACH	HQDAP	261

^a Abbreviations: BACH; 1,1-Bis(4-aminophenyl)cyclohexane, BAET; 1,1-Bis(4-aminophenyl)-4-ethylcyclohexane, BABU; 1,1-Bis(4-aminophenyl)-4-n-butylcyclohexane, BAAM; 1,1-Bis(4-aminophenyl)-4-amylicyclohexane.

^b Abbreviations : PMDA; pyromellitic dianhydride, BTDA : 3,3',4,4'-benzophenonetetracarboxylic dianhydride, BPDA ; 3,3',4,4'-biphenyltetracarboxylic dianhydride, ODPA ; 3,3',4,4'-tetracarboxy-diphenyl oxide dianhydride, HFDA; 2,2-bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride, HQDAP; 1,4-bis(3,4-dicarboxyphenoxy) benzene dianhydride.

^c From the second heating trace of DSC measurement conducted with a heating rate 10 °C/min. in nitrogen atmosphere

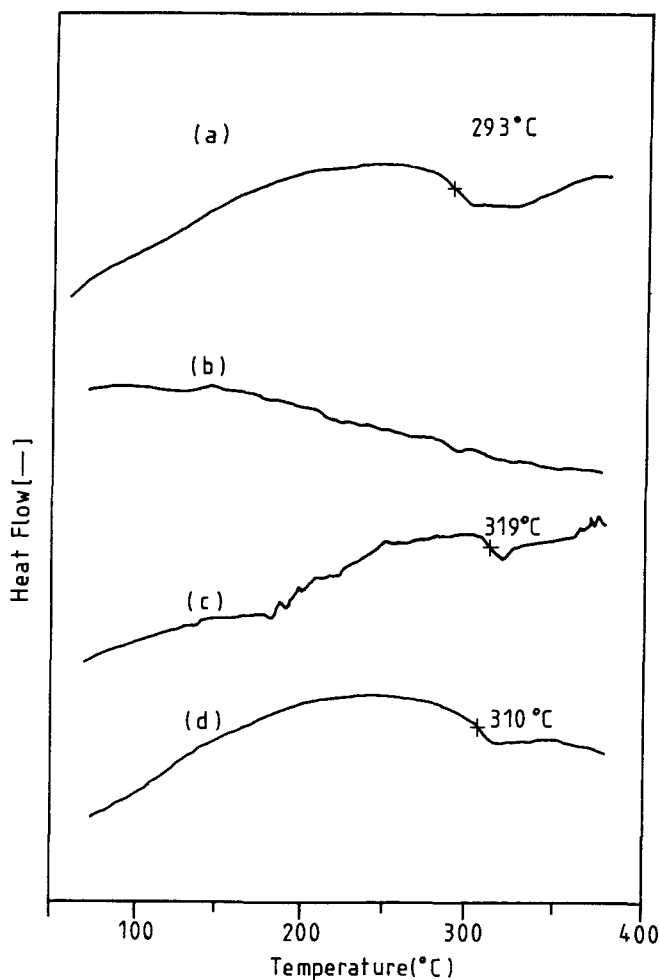


Figure 2. Typical DSC curves of polyimides under nitrogen atmosphere with a heating rate of 10°C/min.

(a)R-1(d) (b) P-1(d) (c) P-2(d) (d) P-3(d)

condensation reaction of excess aniline and corresponding alkyl-substituted cyclohexanone derivatives. The resulting polyimides were readily soluble in common organic solvents such as dimethylacetamide, dimethylformamide, tetrahydrofuran, etc. The glass transition temperatures were in the range of 261-348°C. The solubility and the glass transition temperature of the polymers increased as the bulkiness of the alkyl-substituents increased. Thus, we have concluded that the

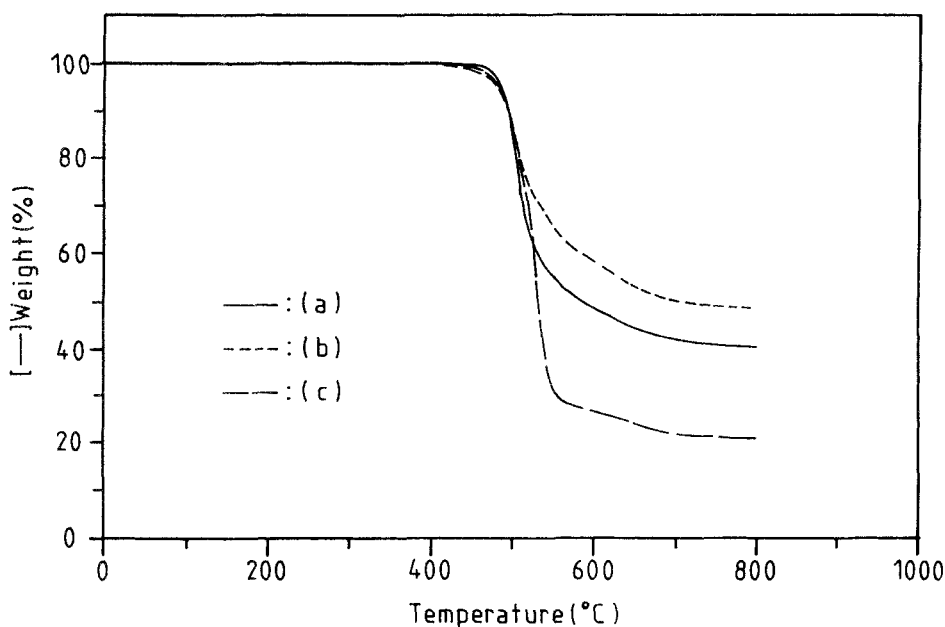


Figure 3. Typical TGA curves of polyimides based on ODPA under nitrogen atmosphere with a heating rate of 10°C/min.

(a) P-1(e) (b) P-2(e) (c) P-3(e)

introduction of a cyclohexylidene moiety with an alkyl-substituent along with the polyimide backbone improved the solubility of these polyimides with retention of its high thermal stability.

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