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## SOLUBLE AND COLORLESS POLYIMIDES FROM ALICYCLIC DIAMINES

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**Key Words:** Soluble Polyimides, Colorless, Alicyclic Diamine, Good Solubility, High Glass Transition Temperature, Crystallinity, Thermal Stability, One-Step Polymerization, Excellent Transparency

### ABSTRACT

A series of novel polyimides was synthesized from alicyclic diamines and various aromatic dianhydrides by one-step polymerization in *m*-cresol without a catalyst. The polymerization was conducted for 4 hours with refluxing, which was enough to obtain the polymers with high molecular weight. The inherent viscosities of the resulting polyimides were in the range of 0.30 ~ 1.29 dL/g. The prepared polyimides showed excellent thermal stabilities and good solubility. All the polymers were readily soluble in common organic solvents such as chloroform, tetrachloroethane (TCE), dimethylacetamide (DMAc), etc and the glass *transition* temperatures were observed at 199 to 311°C. UV-visible spectra were obtained to measure the *transparency* of polymer films. All the polymers showed high *transmission* above 90% in the wavelength of 400 ~ 700 nm.

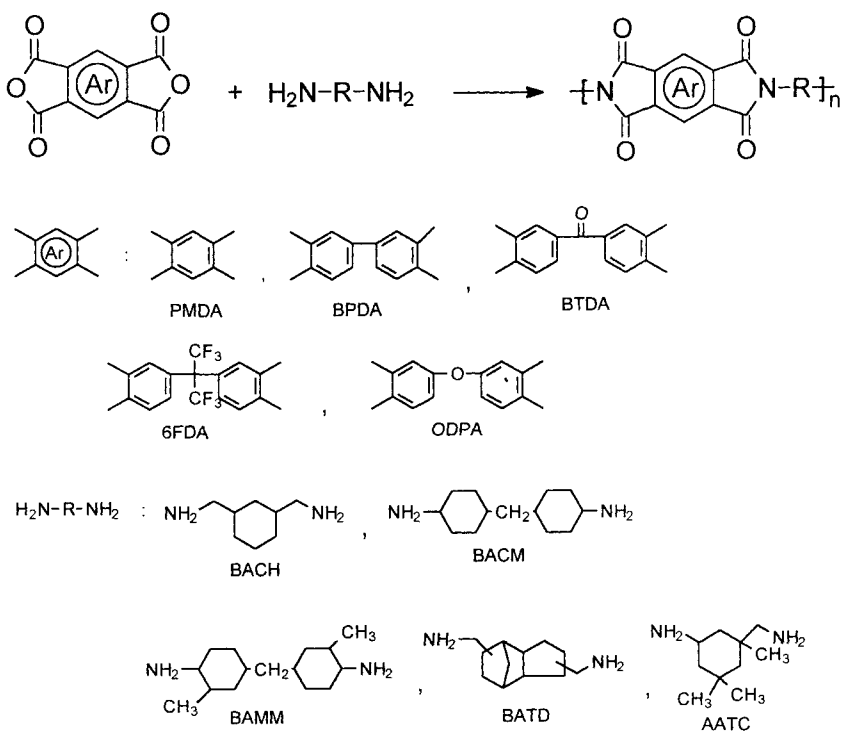
## INTRODUCTION

Aromatic polyimides have been noted for excellent characteristics such as thermo-oxidative stabilities, electrical property, as well as chemical resistance. So, these polymers have been widely used for a number of applications [1]. However, most of the aromatic polyimides have one major drawback, their intractability in fully imidized form. Furthermore, the formation of intermolecular charge transfer complex of aromatic polyimides was the reason for the coloration. Therefore, much effort has been focused on the synthesis of soluble and colorless polyimides without sacrificing their excellent properties. One approach was to introduce the flexible linkages like amides and ether groups, or the incorporation of bulky pendant groups [2]. Another approach was the introduction of alicyclic structure using alicyclic dianhydride reported by Kurosaki *et al.* [3-5]. Recently, W. Volkson *et al.* reported the preparation of alicyclic polyimides by two-step polycondensation. They showed that the aliphatic diamines, having higher basicity compared to aromatic ones, formed the salts with the carboxylic acid groups of polyamic acid during the polymerization, which prevented the increase of molecular weight of polymers [6-9]. Therefore, in this study, we would like to introduce the preparation of a class of soluble polyimides with high molecular weight from several alicyclic diamines by one step polymerization at high temperature as shown in Scheme 1. Furthermore, the properties of the resulting polymers such as solubility, thermal stability, and optical property were investigated in detail.

## EXPERIMENTAL

### Materials

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 (BP.DA, 99.6%, Mitsubishi Kasei) were used without further purification, since they were of a highly purified grade for polyimide synthesis. 2,2-Bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride (6FDA, 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. 1,3-Bis(aminomethyl)cyclohexane (BACH, a *cis* and *trans* mixture, Tokyo Chemical Industry Co., Ltd., 98%, bp. 113°C), bis(4-amino cyclohexyl)methane (BACM, mixture of isomer, Tokyo Chemical Industry Co., Ltd.), bis(4-amino-2-methyl-



Scheme 1. Synthesis of Polyimides

cyclohexyl)methane (BAMB, a mixture of isomer, Aldrich Chemical Co., Inc., 99%, bp. 93-100°C), 1-amino-3-aminomethyl-3,5,5-trimethylcyclohexane (AATC, *cis* and *trans* mixture, Tokyo Chemical Industry Co., Ltd.) and 3(4), 8(9)-bis(amino-methyl)tricyclo(5,2,1,0<sup>2,6</sup>)decane (BATD, Tokyo Chemical Industry Co., Ltd., >97%, bp. 162°C) were used without further purification. *m*-Cresol was freshly distilled under reduced pressure.

### Measurements

All NMR spectra were taken on a Bruker AMX 500, using CDCl<sub>3</sub> or dimethyl-*d*<sub>6</sub> sulfoxide(DMSO-*d*<sub>6</sub>) as solvents. Infrared spectra were obtained with a Bio-Rad Digilab Division FTS-165 FT-IR spectrophotometer. UV-Visible spectra were recorded on a Shimadzu UV 2100 UV-visible spectrophotometer. The inherent viscosities of polymers were measured with a Ubbelohde viscometer at a concentration of 0.5 g/dL in *m*-cresol at 30°C. Thermogravimetric analysis (TGA) was done with a Du Pont model 915 coupled to a Du Pont Thermal analysis station 9900 at a heating rate of 10°C/min. under nitrogen

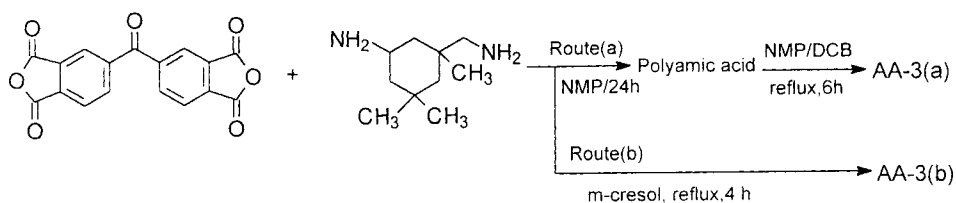
atmosphere. Differential scanning calorimeter (DSC) analysis was performed on a Du Pont model 910 at a heating rate of 10°C/min. under nitrogen atmosphere. The obtained polyimide powders were heated to the temperature of 350°C to remove the residual solvents or water and then  $T_g$ s were determined by the second heating trace. Wide-angle X-ray diffraction measurements for powdery samples were performed at room temperature on a Siemens Kristalloflex D 5000 X-ray diffractometer using Ni-filtered CuK  $\alpha$  radiation (40 kV, 15mA). The scanning rate was 2°C/min. over a range of  $2\theta = 5\sim 50$  deg. An Instron universal tester Model 1130, with a load cell 5 kg, was used to measure the tensile strength of films.

#### Preparation of Polyimide [AA-3(a)] by Synthetic Route (a)

The polyimide, AA-3(a) was prepared by the polycondensation reaction of BTDA and AATC followed by thermal imidization in NMP. A typical procedure for polyimide, AA-3 (a) is as follows. To a solution of AATC (10 mmol, 1.70 g) in 44 mL of NMP, BTDA (10.0 mmol, 3.22 g) was added and the solution was stirred for 24 hours. To the reaction mixture, 10 mL of 1,2-dichlorobenzene was added to remove the water produced by thermal imidization and the temperature was raised to 170~180°C. The resulting viscous solution was poured into a large amount of methanol with vigorous stirring. The precipitated polymer was pulverized using methanol in a Waring blender® and washed with hot methanol using a Soxhlet extractor. The polymer was dried at 100°C for 12 hours under reduced pressure and the yield was quantitative.

#### Preparation of Polyimide [AA-3(b)] by Synthetic Route (b)

The polymer AA-3(b) was prepared by one-step polymerization at high temperature. A typical polymerization procedure is as follows. To a solution of AATC (10 mmol, 1.70 g) in 44 mL of freshly distilled *m*-cresol, BTDA (10 mmol, 3.22 g) was added at room temperature under nitrogen atmosphere. The reaction mixture was heated to 70~90°C over 2 hours and kept at that temperature for 2 hours, and then the solution temperature slowly raised to 200°C over 2 hours and refluxed for 4 hours. The work-up process was the same as that of synthetic route (a). The yield was quantitative.  $^1\text{H NMR } \delta$  ( $\text{CDCl}_3$ ) 8.18-8.13 (m, 4H,  $\text{H}_{a,b}$ ), 8.03-7.97 (m, 2H,  $\text{H}_c$ ), 5.00-4.24 (m, 1H,  $\text{H}_d$ ), 3.55 (s, 2H,  $\text{H}_f$ ), 1.05-0.98 ( $\delta$ , 9H,  $\text{H}_{g,i,j}$ ); FT-IR (KBr pellet) 1782 ( $\nu_{\text{C=O}}$ , asym.), 1710 ( $\nu_{\text{C=O}}$ , sym.), 1380-1350 ( $\nu_{\text{C-N}}$ ), 750-720 (imide ring deformation); Elemental Anal. Calcd. for  $\text{C}_{27}\text{H}_{24}\text{N}_2\text{O}_5$ : C, 71.1; H, 5.3 ; N, 6.1, Found : C, 70.1; H, 5.2 ; N, 6.0%.



Scheme 2. Preparation of Soluble Polyimides by Different Synthetic Methods

## RESULTS AND DISCUSSION

### Synthesis of Polymers

Generally, it is very difficult to obtain the polyimides with high molecular weight from aliphatic diamines because of the higher basicity of aliphatic diamines compared to that of aromatic ones, which resulted in the formation of the salt with the pendant carboxylic acid group of polyamic acid during polymerization. Therefore, in this study, we have varied the polymerization condition to obtain the polyimides with high molecular weight from alicyclic diamines. As shown in Scheme 2, the polymerization of AATC with BTDA were carried out by conventional two-step polyimidization reaction via polyamic acid precursor. It was found that the considerable gelation occurred during the addition of BTDA to a solution of AATC and then the solution became clear on stirring. After the solution was stirred for 24 hours, the resulting polyamic acid solution was thermally imidized using 1,2-dichlorobenzene (DCB) as the azeotropic solvent as shown in route (a). But the obtained polyimide [AA-3(a)] had low molecular weight with the inherent viscosity of 0.20 dL/g. It was probably due to the steric hindrance of the three methyl substituents of AATC and the formation of salt complex between highly basic alicyclic diamine and carboxylic acid group of polyamic acid, which prevented the chain growth of polymer.

Therefore, in an attempt to prepare the polyimides with high molecular-weight from alicyclic diamines, we performed the polymerization by a one-step method. As a result, the polymerization in *m*-cresol at high temperature by route (b) gave the polyimides with high molecular weights. All the polymers remained fully soluble during polymerization. The results of polymerization are summarized in Table 1. The inherent viscosities of obtained polymers were in the range

TABLE 1. Synthesis of Polyimides from Alicyclic Diamines

Polymers	Dianhydrides <sup>a</sup>	Diamines <sup>b</sup>	$\eta_{inh}$ (dL/g) <sup>c</sup>
CH-1	PMDA	BACH	0.30
CH-2	BPDA	BACH	0.84
CH-3	BTDA	BACH	1.02
CH-4	6FDA	BACH	0.39
CH-5	ODPA	BACH	1.06
CM-1	PMDA	BACM	- <sup>d</sup>
CM-2	BPDA	BACM	0.94
CM-3	BTDA	BACM	0.91
CM-4	6FDA	BACM	0.93
CM-5	ODPA	BACM	0.80
MM-1	PMDA	BAMM	1.29
MM-2	BPDA	BAMM	0.72
MM-3	BTDA	BAMM	0.38
MM-4	6FDA	BAMM	0.49
MM-5	ODPA	BAMM	0.83
TD-1	PMDA	BATD	0.83
TD-2	BPDA	BATD	0.71
TD-3	BTDA	BATD	0.51
TD-4	6FDA	BATD	0.62
TD-5	ODPA	BATD	0.80
AA-1	PMDA	AATC	0.79
AA-2	BPDA	AATC	0.81
AA-3	BTDA	AATC	0.83
AA-4	6FDA	AATC	0.32
AA-5	ODPA	AATC	0.73

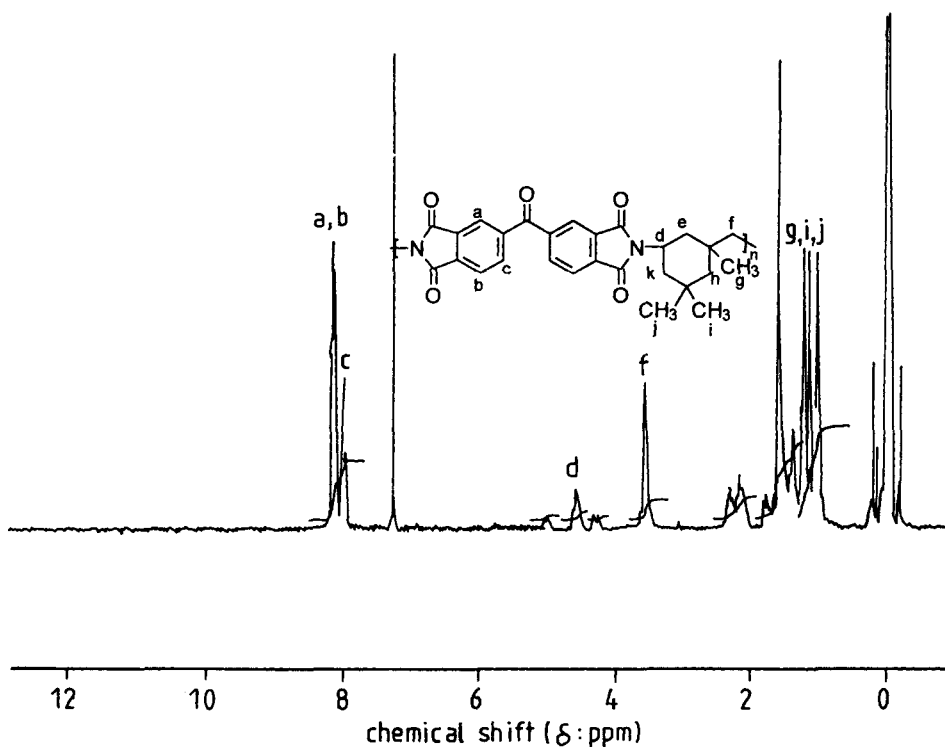
<sup>a</sup> Abbreviations: BACH; 1,3-bis(aminomethyl)cyclohexane, BACM; bis(4-aminocyclohexyl)methane, BAMM; bis(4-amino-2-methylcyclohexyl)methane, BATD; 3(4), 8(9)-bis(aminomethyl)tricyclo (5,2,1,0<sup>2,6</sup>)decane, AATC; 1-amino-3-aminomethyl-3,5,5-trimethyl- cyclohexane.

<sup>b</sup> Abbreviations : PMDA; pyromellitic dianhydride, BTDA ; 3,3',4,4'-benzophenonetetracarboxylic dianhydride, BPDA ; 3,3',4,4'-biphenyltetracarboxylic dianhydride, 6FDA; 2,2-bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride ODPA ; 3,3',4,4'-tetracarboxydiphenyl oxide dianhydride.

<sup>c</sup> Measured at a concentration of 0.5 g/dL in *m*-cresol at 30 °C.

<sup>d</sup> Insoluble.

of 0.30-1.29 dL/g, which is much higher than those of polyimides prepared by conventional two-step methods. It can be seen that the water produced during imidization could be easily removed in water-immiscible solvent, *m*-cresol. Furthermore, the solubility of the salt complex might be increased at high temperature and the imidization reaction readily occurred in acidic solvent [10],



**Figure 1.** <sup>1</sup>H NMR (300-MHz) spectrum of polyimide, AA-3 in DMSO-*d*<sub>6</sub>.

which can result in the high molecular weight of polymer. The structures of polyimides were characterized by FT-IR and <sup>1</sup>H-NMR spectroscopy. A typical <sup>1</sup>H-NMR spectra of polyimide, AA-3 is shown in Figure 1, which reveals that the polycondensation and imidization reaction proceeded successfully.

### Polymer Characterization Solubility

The obtained polymers are expected to exhibit an enhanced solubility in common organic solvents due to aliphatic character. Table 2 shows the solubility behavior of polymers. Except for the polymer, CM-1, all the other polymers were soluble *m*-cresol and 1,1,2,2-tetrachloroethane (TCE). The lower solubility of polymer, CM-1 was due to the symmetric structure of BACM. Furthermore, polyimides based on BACH, BAMM, and BATD showed higher solubility in organic solvents. All of them were readily soluble not only in typical polar aprotic solvents such as *N*-methyl-2-pyrrolidone (NMP), dimethyl sulfoxide



TABLE 2. Solubility of Polyimides

Polymers	Solvents <sup>a</sup>							
	<i>m</i> -cresol	NMP	DMAc	DMSO	TCE	CHCl <sub>3</sub>	THF	Acetone
CH-1	+ + <sup>b</sup>	+ +	+ +	+ +	+ +	+ +	+ +	---
CH-2	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
CH-3	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
CH-4	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
CH-5	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
CM-1	+ --- <sup>b</sup>	---	---	---	+ ---	---	---	---
CM-2	+ +	+ ---	---	---	+ +	+ ---	---	---
CM-3	+ +	+ ---	---	---	+ +	+ ---	---	---
CM-4	+ +	+ ---	---	---	+ +	+ ---	---	---
CM-5	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
MM-1	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
MM-2	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
MM-3	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
MM-4	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
MM-5	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
TD-1	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
TD-2	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
TD-3	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
TD-4	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
TD-5	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
AA-1	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
AA-2	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
AA-3	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
AA-4	+ +	+ +	+ +	+ +	+ +	+ +	+ +	---
AA-5	+ +	+ +	+ +	+ +	+ +	+ +	+ +	+

<sup>a</sup> Abbreviations : NMP, *N*-methyl-2-pyrrolidone, DMAc, dimethylacetamide, TCE; 1,1,2,2-tetrachloro ethane.

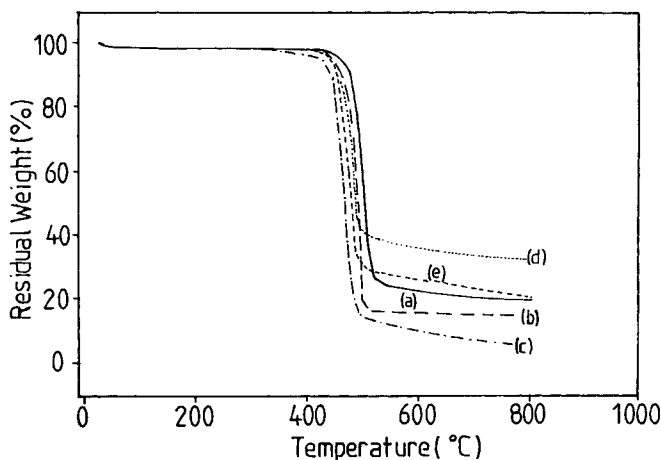
<sup>b</sup> + + ; soluble, + ; soluble on heating, + --- ; partially soluble, --- ; insoluble.

(DMSO), and dimethylacetamide (DMAc), but also in low-boiling solvents like tetrahydrofuran and chloroform at room temperature. Polymer AA-5 was even soluble in acetone. The good solubility of this polymer might be apparently due to the introduction of the flexible structure of ODPA, as well as three methyl substituents of AATC. Most of the polyimides could be cast into flexible, tough, and colorless films from TCE solutions. Some of the obtained films were subjected to measurement of tensile strength and the results are summarized in Table 3. As shown in Table 3, the tensile strength measured in film state at room

TABLE 3. Tensile Strength of Polyimides

Polymers	T.S. (kg/cm <sup>2</sup> ) <sup>a</sup>
AA-1	913
AA-2	949
AA-3	898
AA-5	873
MM-1	1,127
MM-2	991
MM-5	965
TD-1	1,021
TD-2	943
TD-5	864

<sup>a</sup> T.S. ; tensile strength measured at film state.



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

(a) AA-1 (b) AA-2 (c) AA-3 (d) AA-4 (e) AA-5

temperature ranged from 864~1,127 kg/cm<sup>2</sup>, which is comparable to those of other alicyclic polyimides.

### Thermal Property

The thermal stabilities of the obtained polyimides were evaluated by thermal gravimetric analysis (TGA). The representative TGA curves of polyimides are shown in Figure 2. All of them are stable up to 400°C, and the residual weights at 800°C were in the range of 26.0-46.3% under nitrogen

TABLE 4. Thermal Properties of Polyimides

Polymers	T <sub>g</sub> (°C) <sup>a</sup>	T <sub>m</sub> (°C) <sup>c</sup>	T <sub>hc</sub> (°C) <sup>d</sup>	IDT(°C) <sup>e</sup>
CH-1	263	-	-	417
CH-2	234	-	-	417
CH-3	213	-	-	425
CH-4	222	-	-	439
CH-5	199	-	-	-
CM-1	- <sup>b</sup>	-	-	392
CM-2	242	-	485	435
CM-3	254	-	496	389
CM-4	265	-	-	429
CM-5	240	-	-	428
MM-1	-	489	-	407
MM-2	260	-	-	421
MM-3	240	-	-	396
MM-4	225	-	-	405
MM-5	250	-	477	403
TD-1	265	-	477	431
TD-2	230	-	-	430
TD-3	217	-	475	399
TD-4	207	-	481	410
TD-5	201	-	489	406
AA-1	-	-	-	424
AA-2	311	-	-	423
AA-3	275	-	-	430
AA-4	278	-	-	427
AA-5	266	-	-	434

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

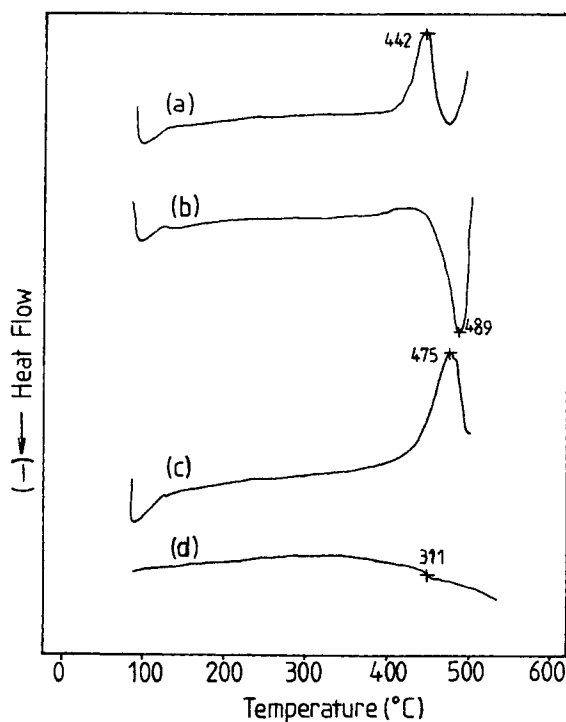
<sup>b</sup> T<sub>g</sub>s were not detected.

<sup>c</sup> The melting temperature was measured by DSC.

<sup>d</sup> Crystallization temperatures by heating were measured by DSC.

<sup>e</sup> IDT: initial decomposition temperature.

atmosphere. The relatively lower char residue for these polymers may be associated with the lower aromatic ring density of these polymers. The thermal properties of polymers were also determined by differential scanning calorimetry (DSC) under nitrogen atmosphere. The results are summarized in Table 4. The glass transition temperature of polymers were in the range of 199-311°C. The T<sub>g</sub> of polyimide (CM-3) derived from BACM was higher than that of aromatic polyimides having phenyl ring instead of the cyclohexane structures [7]. Typical



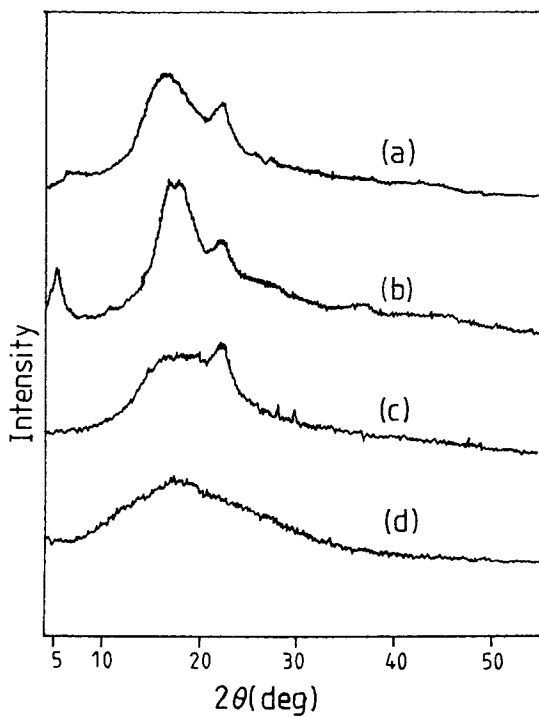
**Figure 3.** Typical DSC curves of polyimides under nitrogen atmosphere with a heating rate of 10°C/min.

(a) CM-5    (b) MM-1    (c) TD-3    (d) AA-2

DSC thermograms are shown in Figure 3. The exothermic peaks corresponding to crystallization were observed for polyimides, CM-5 and TD-3. Some of the polyimides showed the melting endothermic peaks near the temperature of 500°C. The partial crystallization of these polymers was also confirmed by wide angle X-ray diffraction patterns. Typical X-ray diffractograms of polyimides are shown in Figure 4, which reveals that the polyimides (CM-5, MM-1) prepared from the diamines, having symmetric structures, exhibited higher crystallinity compared to that of polyimides (AA-2) containing non-symmetric structure-like trimethyl-cyclohexyl moiety. On the other hand, partial crystallinity of polyimide, TD-3 was unexpected because of the non-symmetric polyalicyclic structure of BATD.

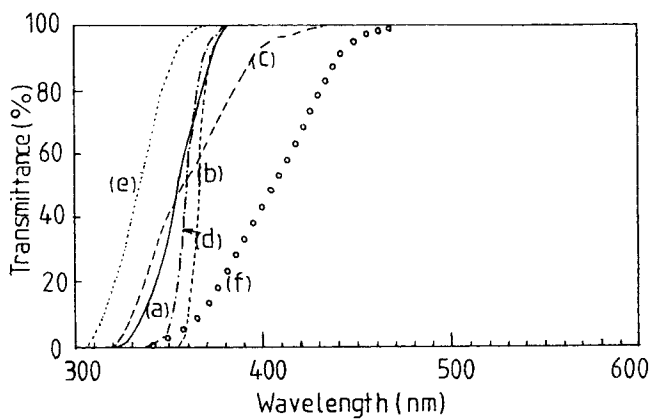
### Optical Property

Polyimides containing alicyclic structures are expected to have better *transparency* than aromatic polyimide due to the prohibition of electron



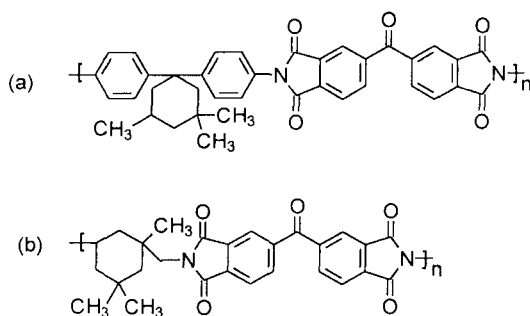
**Figure 4.** Wide angle X-ray diffraction curves of polyimides.

(a) CM-5 (b) MM-1 (c) TD-3 (d) AA-2



**Figure 5.** UV-visible spectra of soluble polyimides.

(a) AA-1 (b) AA-2 (c) AA-3 (d) AA-4 (e) AA-5 (f) Ref-PI11



Scheme 3. Structure of Polyimides  
(a) Ref-PI (b) AA-3

conjugation by the introduction of alicyclic moiety. *Transmission* UV-visible spectra of polyimides (0.2 % chloroform solution) are shown in Figure 5. The polyimides (AA-1~5) exhibited cutoffs at wavelengths shorter than 350 nm and were entirely colorless. As shown in Figure 5, the *transparency* depended on the structure of dianhydrides used. That is, the polyimides (AA-4, AA-5) prepared from dianhydrides with non-conjugated structure like 6FDA and ODPA showed better *transparency* compared to those of other polyimides. Furthermore, the *transparency* of polyimide (AA-3) with 3,3,5-trimethylcyclohexylidene moiety in polymer backbone was significantly improved as compared to that of polyimide (Ref-PI) prepared from aromatic diamines containing similar moiety between two phenyl rings reported by our previous work [11]. The chemical structures of Ref-PI and AA-3 were shown in Scheme 3. It can be seen that the introduction of alicyclic moiety into polymer backbone resulted in better *transparency*.

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## REFERENCES

- [1] D. Wilson, H. D. Stenzenberger, and P. M. Hergenrother, *Polyimide*, Blackie, Glasgow, 1990.

- [2] S. J. Huang and A. E. Hoyt, *TRIP*, 3, 262 (1995).
- [3] T. Matsumoto, and T. Kurosaki, *Macromolecules*, 30, 993 (1997).
- [4] Y. Tsuda, Y. Tanaka, K. Kamata, N. Hiyoshi, S. Mataka, Y. Matsuki, M. Nishikawa, S. Kawamura, and N. Bessho, *Polymer Journal*, 29, 574 (1997).
- [5] M. Kusama, T. Matsumoto, and T. Kurosaki, *Macromolecules*, 27, 1117 (1994).
- [6] W. Volkson, H. J. Cha, M. I. Sanchez, D. Y. Yoon, *Reactive & Functional Polymers*, 30, 61 (1996).
- [7] Q. Jin, T. Yamashita, K. Horie, R. Yokota, and I. Mita, *J. Polym. Sci., Part A: Polym. Chem.*, 31, 2345 (1993).
- [8] Q. Jin, T. Yamashita, and K. Horie, *J. Polym. Sci., Part A: Polym. Chem.*, 32, 503 (1994).
- [9] M. H. Yi, M. Y. Jin, and K.-Y. Choi, *Angew. Makromol. Chem.*, 233, 89 (1995).
- [10] S. V. Lavrov, A. Ya. Ardashnikov, I. Ye. Kardash, and A. N. Pravednikov, *Vysokomol. soyed, A19*, 1052 (1977).
- [11] M. H. Yi, W. Huang, M. Y. Jin, and K.-Y. Choi, *Macromolecules*, 30, 19 (1997).

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