

## Soluble Tetraphenylthiophene containing poly (amide – imide)s: Synthesis and characterization

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**ABSTRACT:** A monomer 2, 5 – bis (4-trimellitimidophenyl)-3, 4 – diphenyl thiophene (BTPDT) was synthesized, characterized by physical and spectroscopic methods and poly (amide-imide) and copoly (amide-imide)s were prepared from the (BTPDT) and different aromatic diamines using Yamazaki's direct phosphorylation reaction. Copoly (amide-imide)s from oxy-bisbenzoic acid and (BTPDT) with bis(4-aminophenyl) ether / bis(4-aminophenyl) sulfone were synthesized in quantitative yields. These poly(amide - imide) and copoly(amide - imide)s were characterized by IR spectroscopy, viscosity measurements and thermal analysis. Inherent viscosities of poly (amide-imide)s was in the range of 0.51 to 1.40 dL/g, whereas the copoly (amide-imide)s had the inherent viscosities in the range of 0.48 to 2.35 dL/g. Copoly(amide - imide)s had better solubility than corresponding poly(amide - imide)s in polar aprotic solvents such as N-methyl-2-pyrrolidone, N-N-dimethyl acetamide, dimethyl sulphoxide etc. These polymers were easily castable into transparent and flexible films. Polymers had glass-transition temperature of 252 - 360 °C under nitrogen; and thermogravimetric analysis showed no weight loss below 407 °C. The structure – property correlation among these polymers is discussed.

**Key words:** 2, 5 – Bis (4-trimellitimidophenyl)-3, 4 – diphenyl thiophene, poly (amide-imide)s, solubility, film forming, thermal properties.

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### I. INTRODUCTION

Aromatic polyimides, polyamides have excellent thermal stability and mechanical properties. The high temperature resistance and thermal stability of polymers could be enhanced by modifying structures of the side groups, but leaving the main carbon chain unchanged, or by introducing heat-resistant groups such as aromatic rings into the chain. However these polymers have the poor processability due to their insolubility in common organic solvents and extremely high glass transition or melt temperature. Much research efforts have been focused on the synthesis of soluble polyimides / polyamides without deteriorating their otherwise excellent properties. Aromatic poly(amide imide)s (PAIs) are an important class of high performance polymers having a favorable balance of physical, chemical and thermal properties [1,2]; and find use as an engineering thermoplastic material in electrical wire enamels, adhesives and various injection or extrusion molding products. They inherit desirable characteristics from both polyimides and polyamides, and possess good thermal properties compared to polyamides and better melt processability than polyimides. To further improve processability and solubility of PAIs, several approaches have been adapted which include copolymerization, introduction of bulky pendent / cardo groups, flexible linkages and kinked or unsymmetrical structures into the polymer backbone [3-10].

Soluble high-temperature polymers such as polyimides, poly (amide-imide)s, poly (ester-imide)s, poly (imide carbonate)s, poly (amide-ester-imide)s, polyhydrazides, and polyazomethines, which contain phenylated unit are reported [11-19]. However, there are no reports on the synthesis of copoly (amide-imide)s from (BPTDT); with bis(4-aminophenyl) ether / bis(4-aminophenyl) sulphone or poly (amide-imide) from 1,5-naphthalene diamine. Hence we thought that it will be interesting to use (BPTDT) for preparation of new high molecular weight poly (amide-imide) polymers and copoly (amide-imide)s having better solubility without sacrificing thermal stability; by inserting flexible ether / sulphone groups, pendant phenyl groups in the polymer backbone and a technique of copolymerization. Thus we report synthesis of these poly (amide-imide) and copoly (amide-imide)s and their characterization by measurement of inherent viscosity, solubility, film-forming property, IR-spectroscopy and thermal properties. The effect of copolymerization by varying mole ratio of (BPTDT) to oxy bis(benzoic acid) on properties of copoly (amide-imide)s is also reported.

## II. EXPERIMENTAL

### Materials :

Commercially available sulfur (sd. fine) was purified by refluxing the sulfur with calcium oxide in water then filtrate was acidified with conc. HCl and precipitated sulfur powder was filtered, washed with water and dried under vacuum at 80 °C for 5h. Benzyl chloride (sd. fine) was fractionally distilled. N-methylpyrrolidone (sd. fine) was dried by azeotropic removal of water with benzene for 6h, distilled under reduced pressure and stored over Linde type 4A molecular sieves. N-N Dimethylacetamide (sd. fine) was refluxed over barium oxide for 4h, the liquid was decanted in a separate round bottom flask and distilled at reduced pressure over calcium hydride and stored over Linde type 4A molecular sieves. Lithium chloride (sd. fine) was dried under vacuum at 150 °C for 6h. Pyridine (sd. fine) was refluxed with potassium hydroxide pellets, fractionally distilled and stored over Linde type 4A molecular sieves.

All the aromatic diamines utilized in the present investigation were purified prior to use. p-Phenylenediamine (p-PDA) (sd. fine) was purified by sublimation under reduced pressure; m-phenylene diamine (m-PDA) (sd. fine) was purified by vacuum distillation. Bis(4-aminophenyl) ether (ODA) (Himedia), and bis(4-aminophenyl) sulfone (SDA) (Himedia) were recrystallized from alcohol. 1, 5-Diamino naphthalene (NDA) (Aldrich) was purified by sublimation under vacuum (at 180 °C at <1mm Hg). 2, 5-Bis (4-aminophenyl) 3, 4-diphenyl thiophene (TPTPDA) was synthesized as reported by Basutkar et. al. [18] and it was recrystallized from toluene. Trimellitic anhydride (Benzene-1,2,4-tricarboxylic 1,2-anhydride) was obtained by sublimation of trimellitic acid under reduced pressure, and the sublimate was washed with dry hexane under inert atmosphere. Oxy – bis(benzoic acid), o-BBA (Aldrich) was used as received.

### Measurements:

Melting points were recorded on Rigaku TG-DSC at heating rate of 10 °C /min under nitrogen atmosphere. IR spectra were taken on Perkin-Elmer infrared spectrophotometer - 883 using KBr pellet. NMR spectra were recorded on Varian 80 spectrometer using tetramethyl silane as an internal reference in deuterated solvents such as deuteriochloroform or hexadeutero dimethyl sulphoxide. High-resolution mass spectra were obtained on IIMS-30 double beam mass spectrometer. Inherent viscosities were measured in N-N dimethylacetamide containing 6 % lithium chloride at 30°C using an Ubbelohde suspended level viscometer, at 0.5% (W/V) concentration of polymers. Wide-angle X-ray diffractograms were obtained on Philips X-ray unit (Philips generator, PW - 1730) and a nickel filtered CuK $\alpha$  radiations. Solubilities were determined at 3 % (W/W) concentration in various solvents at room temperature or on warming if needed. Thermal stability and glass transition temperature were determined on a Rigaku-Thermoflex – 8800 TG – DSC at a heating rate 10 °C/min under nitrogen.

### Synthesis of 2, 5 – bis (4-trimellitimidophenyl)-3, 4 – diphenyl thiophene (IV) (BTPDT):

The dicarboxylic acid, 2, 5-bis (4-trimellitimidophenyl)-3, 4-diphenyl thiophene (IV) was synthesized in several steps.

Tetraphenyl thiophene (I) was prepared in good yields by refluxing benzyl chloride with sulfur. The dinitro derivative (II) was synthesized by nitration of (I) using conc. HNO<sub>3</sub> in glacial acetic acid. The IR spectrum (KBr) of (II) showed absorption at 1510 and 1340 cm<sup>-1</sup> (-NO<sub>2</sub>). The 2, 5-bis (4-aminophenyl)-3, 4-diphenyl thiophene (III) was prepared by reduction of (II) with stannous chloride and conc. HCl. The infrared spectrum (III) showed absorption at 3420 and 3340 cm<sup>-1</sup> (-NH Stretching).

2, 5-Bis (4-trimellitimidophenyl)-3, 4-diphenyl thiophene (IV) was synthesized through the reaction of (III) with 2 moles of trimellitic anhydride as detailed below.

In to a 250 mL three neck round bottom flask equipped with Dean stark assembly, a magnetic stirrer, a calcium chloride guard tube and a nitrogen gas inlet were placed 4.18g (0.01mol) (III) in 80 mL N-N-dimethyl acetamide. It was cooled to 0°C with stirring and trimellitic anhydride 3.84 g (0.02 mol) was added in small lots over 10 min. to the reaction solution. Solid dissolved as the reaction proceeded. The reaction mixture was stirred at 0°C for 1 h, ice bath was removed and temperature of reaction solution was allowed to rise to 25 °C, where it was stirred for 5h. Then 50 mL toluene was added and mixture was heated with stirring to reflux using Dean – stark assembly until calculated amount of water was distilled off azeotropically, over a period of 6 hrs. The solution was allowed to cool to 25°C and solid (IV) was isolated by filtration, washed with methanol and dried.

Yield: 6.20 g (80.93%), Mp. 345°C. (Lit. mp. 346-347 °C)

IR spectrum of (IV) showed absorptions at 3436cm<sup>-1</sup> (-COOH), 1778 cm<sup>-1</sup> asymmetrical stretching of imide carbonyl, 1713, 1721, 1727cm<sup>-1</sup> acid carbonyl, and 729cm<sup>-1</sup> imide ring deformation.

### III. POLYMERIZATION

Polymerization of (IV) and various aromatic diamines was performed by direct solution polymerization by Yamazaki's phosphorylation method [20] to yield high molecular weight poly (amide-imide)s. A typical procedure for synthesis of poly (amide-imide)s is given below.

#### Synthesis of poly(amide-imide)s:

In a 100 mL three necked round bottom flask equipped with a reflux condenser, a magnetic stirrer, a calcium chloride guard tube and a nitrogen gas inlet were placed 0.766 g (1mmol) (IV), 0.200 g (1mmol) 4,4'-diaminodiphenyl ether (ODA), 0.200 g lithium chloride [8 wt % based on solvent N-methyl pyrrolidone (NMP) and pyridine mixture] and 0.744 g (0.63 mL, 2.4 mmol) triphenyl phosphite (TPP), 0.5 mL pyridine and 2 mL NMP. The mixture was stirred well and temperature was slowly raised to 100°C over a period of 30 min. and the mixture maintained at 100°C for 3h. After cooling to 25°C the resultant viscous solution was poured into rapidly stirred 200 mL of methanol. The precipitated polymer (P-1) was filtered, washed with methanol and dried under vacuum at 60 °C for 8 hrs. The polymer was purified by dissolving in N, N-dimethylacetamide (DMAc) and reprecipitating in methanol. It was filtered, washed with methanol and dried under vacuum at 100°C for 8 hrs. The yield was 99% and the viscosity of polymer in DMAc was 1.40 dL/g.

All other poly (amide-imide)s [P-2 to P-5] were prepared similarly.

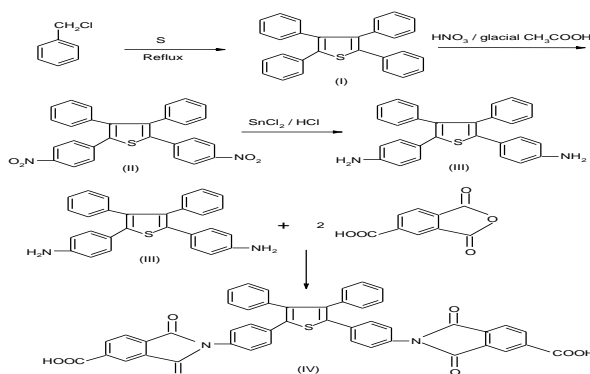
#### Copoly(amide-imide)s:

Further, two series of copoly (amide-imide)s were synthesized utilizing above procedure, where in a mixture of different mol % proportion of (IV) and oxybisbenzoic acid were polycondensed with bis(4-aminophenyl) ether [CPO-1 to CPO-4] or bis (4-aminophenyl) sulfone [CPS-1 to CPS-4].

Poly(amide-imide) and copoly(amide-imide)s from ODA / SDA synthesized are given in Table-1.

#### Results and Discussion:

2, 5-Bis (4-trimellitimidophenyl)-3, 4-diphenyl thiophene,(IV) was synthesized in several steps (Scheme-I); in good yield.



**Scheme – I: Synthesis of 2, 5-bis (4-trimellitimidophenyl)-3, 4-diphenyl thiophene (IV) (BTPDT).**

The dicarboxylic acid (IV) was characterized by acid value, IR, NMR and mass spectra. Acid value calculated theoretically for (IV) is 145.5 mg KOH/g; and found was 145.4 mg KOH/g. DSC analysis of (IV) suggest that it did not melt upto 345°C and underwent degradation / decomposition above this temperature. In the IR spectrum of (IV), characteristic imide absorption bands were observed at 1778, 1709, 1385 and 729cm<sup>-1</sup>. The broad band around 3436 cm<sup>-1</sup> was due to –OH group of diacid (COOH). The <sup>1</sup>H NMR spectrum of (IV) showed multiplet in the region 8.7 to 10.2 ppm due to aromatic protons, and signal due to – COOH group at 13.5 ppm. The mass spectrum of (IV) was consistent with proposed structure, the m/e 766 corresponded to the molecular ion peak, and fragmentation peaks at m/e 576 and 386 indicated loss of carboxyl imidophenyl unit.

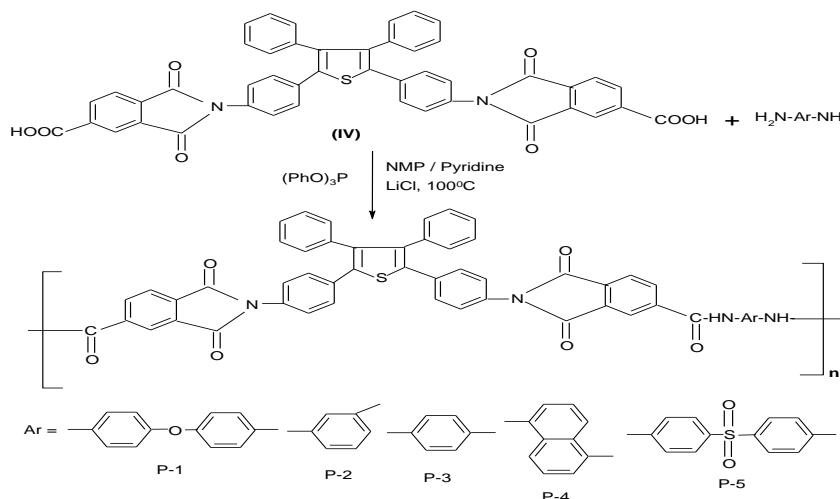
A series of poly (amide-imide)s were synthesized from (IV) by direct polycondensation with aromatic diamines using Yamazaki's phosphorylation method. (Scheme - II) Similar results on molecular weight of polymers ( $\eta_{inh}$ ) were obtained in the present study, as reported earlier [17] for polymers derived from ODA, m-PDA, p-PDA and SDA. The inherent viscosity of new polyamide based on (IV) and 1,5 – naphthalene diamine (ie. Polymer P-4) was 0.98 dL/g in DMAc indicating high molecular weight built-up. Also two series of copoly (amide-imide)s were synthesized from (IV) with oxy bisbenzoic acid and aromatic diamines (ODA / SDA) with triphenyl phosphite as a condensing agent in the mixture of N-methyl pyrrolidone and pyridine (4:1 by volume) containing 8 wt % anhydrous lithium chloride. The polymerizations were carried out at 100°C for 3h under nitrogen (Scheme – III). The polymerization proceeded smoothly giving highly viscous solution. The use of anhydrous lithium chloride avoided the precipitation of polymers during polymerization. The yields and inherent viscosities of poly (amide-imide)s and co-poly(amide-imide)s are given in Table – 1.

**Table 1: Yield, viscosity of poly (amide-imide)s from (IV) with aromatic diamines<sup>(a)</sup> and copoly(amide-imide)s<sup>(b)</sup> from ODA / SDA with (IV + o-BBA<sup>(c)</sup>).**

Polymer Code	Diamine	Diacid (mol %)		Yield (%)	Viscosity ( $\eta_{inh}$ ) dL/g <sup>(d)</sup>
		o-BBA	IV		
P-1	ODA	--	100	99	1.40
P-2	m-PDA	--	100	96	0.97
P-3	p-PDA	--	100	99	0.75
P-4	NDA	--	100	84	0.98
P-5	SDA	--	100	93	0.51
CPO – 1	ODA	100	00	100	1.25
CPO – 2	ODA	75	25	99	1.63
CPO – 3	ODA	50	50	99	1.99
CPO – 4	ODA	25	75	100	2.35
P -- 1	ODA	00	100	99	1.40
CPS – 1	SDA	100	00	100	0.95
CPS – 2	SDA	75	25	99	0.48
CPS – 3	SDA	50	50	99	0.52
CPS – 4	SDA	25	75	99	0.70
P -- 5	SDA	00	100	93	0.51

- a) Polymerization was carried out with 1mmol each of diamine and IV.  
 b) Copolymerization was carried out with 1mmol each of diamine (ODA / SDA) and (IV + o-BBA) in different mole %.  
 c) o-BBA, 4, 4'-oxybis (benzoic acid).  
 d) Measured at concentration of 0.5 g/dL in DMAc containing 6 % LiCl at 30<sup>o</sup>C.

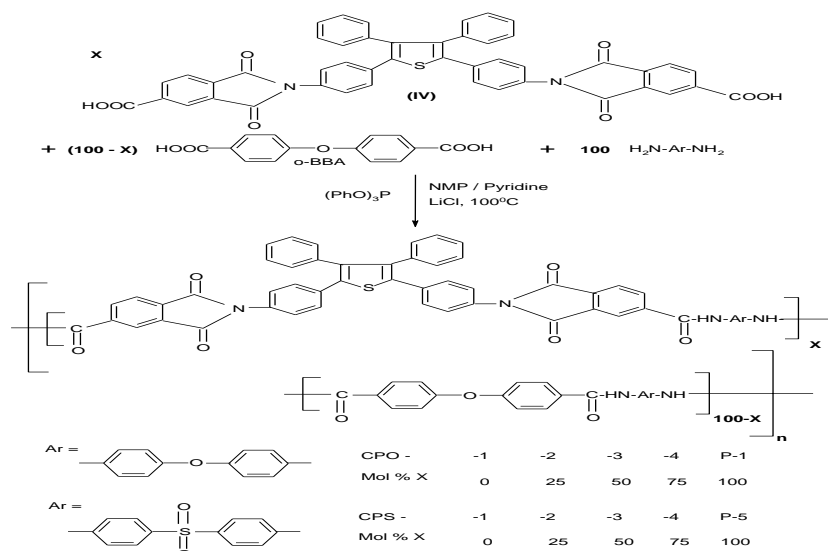
All these copolymers were obtained in almost quantitative yield (99 to 100 %) as a white to pale yellow fibrous material. As a mole % of (IV) increased the copolymers become more fibrous in nature. The inherent viscosities of these polymers were in the range of 0.48 to 2.35 dL/g. As the mol % of (IV) increased the viscosity of the corresponding co-polymer increased. These copolymers gave the transparent and flexible films when cast from the solution in DMAc implying the formation of high molecular weight copolymers in confirmation of high values of inherent viscosities.

**Scheme -II: Synthesis of poly (amide – imide)s from aromatic diamines with (IV)**

IR spectra of copolymers showed the characteristic absorption bands at 1779, 1720 and 700cm<sup>-1</sup> corresponding to imide carbonyl, 3332 cm<sup>-1</sup> (-NH stretch) and 1660 cm<sup>-1</sup> amide carbonyl.

The solubility characteristics of these poly (amide-imide)s and co-poly (amide-imide)s were determined in different common organic solvents. Polymer P-1 to P-3 and P-5 had similar trend of solubility as reported earlier [17], whereas P-4; prepared in the present study exhibited partial solubility only; likely due to rigid aromatic fused naphthalene ring units. This is supported by XRD data for P-4 given below. It was

observed that copolymers exhibited very good solubility (Table-2) in various solvents such as DMAc, DMSO, NMP, DMF, pyridine, m-cresol and conc. H<sub>2</sub>SO<sub>4</sub> but were insoluble in chlorinated solvents like CHCl<sub>3</sub>, DCM, TCE, etc. Overall copolymers CPS- 1 to -4, P -5 showed better solubility than copolymers CPO -1 to -4, and P -1. Copolymers based on SDA were soluble even in pyridine and m-cresol. Enhanced solubility may be attributed to the sulphone linkage in polymer backbone and comparatively lower viscosities of SDA based copolymers (0.48 to 0.95 dL/g) than those of ODA based copolymers which had higher viscosities (1.25 to 2.35 dL/g). Further it is evident that as the mole proportions of (IV) increased the solubility also increased accordingly.



**Scheme -III: Synthesis of Copoly(amide – imide)s from ODA / SDA with (IV + o- BBA).**

**Table 2: Solubility of poly (amide-imide)s from (IV) with aromatic diamines and copoly(amide-imide)s from ODA / SDA with (IV + o-BBA).**

Polymer ↓ Solvent →	DMAc	DMSO	NMP	DMF	Pyridine	m- Cresol	Conc. H <sub>2</sub> SO <sub>4</sub>	Nitro benzene	THF
P-1	++	++	++	+-	++	+-	++	-	-
P-2	++	++	++	+-	+	++	++	-	-
P-3	+	+	+	+-	+-	+-	++	-	-
P-4	+-	+-	+-	-	+-	+-	++	-	-
P-5	++	++	++	++	++	++	++	+-	+-
CPO-1	-	-	+-	-	-	-	+-	-	-
CPO-2	++	++	++	+	+-	+-	++	-	-
CPO-3	++	++	++	++	+	+	+	-	-
CPO-4	++	++	++	+-	+	+	+	-	-
P-1	++	++	++	+-	++	+-	++	-	-
CPS-1	++	++	++	++	++	++	++	+-	+-
CPS-2	++	++	++	++	++	++	++	+-	+-
CPS-3	++	++	++	++	++	++	++	+-	+-
CPS-4	++	++	++	++	++	++	++	+-	+-
P-5	++	++	++	++	++	++	++	+-	+-

++ : Soluble at room temperature

+ : Soluble on heating

+- : Partly soluble

- : Insoluble

Thermal behavior of all the poly (amide-imide)s was evaluated by means of thermogravimetry and differential scanning calorimeter. TG curves are shown in Fig. 1 and Fig 2; whereas DSC curves are shown in Fig 3 and Fig 4.

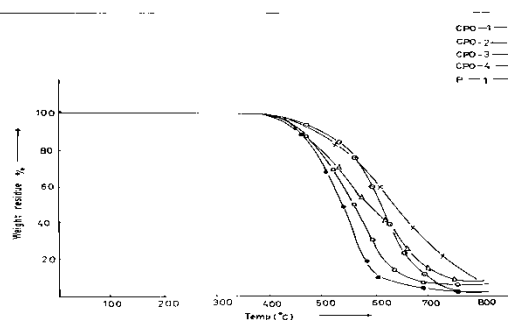


Fig. 1: TG curves of CPO -1 to CPO -4, P-1.

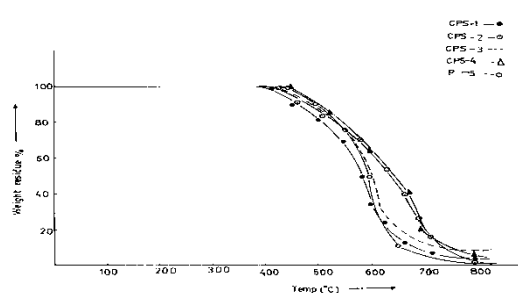


Fig. 2: TG curves of CPS -1 to CPS -4, P-5.

Table -3 incorporates the thermal data such as glass transition temperature ( $T_g$ ), initial decomposition temperature ( $T_i$ ), temperature at 10% wt. loss ( $T_{10}$ ) and temperature at 50% wt. loss ( $T_{50}$ ) for CPO - 1 to CPO-4, P-1 and for CPS - 1 to CPS- 4, P - 5. Copoly (amide-imide)s CPO - 1 to CPO- 4 and P - 1 have  $T_g$  in the range of 252 to 325  $^{\circ}\text{C}$  and CPS - 1 to - 4 and P - 5 showed  $T_g$  in range of 312 to 340  $^{\circ}\text{C}$ .  $T_g$  values for P-1 and P-5; based on ODA and SDA respectively are in the same range as reported earlier [17]. Among copoly (amide-imide)s;  $T_g$  follow the trend CPO - 4 > CPO - 3 > CPO - 2 > CPO - 1 and CPS - 3  $\approx$  CPS - 4 > CPS - 1  $\approx$  CPS - 2. Aromatic and imide group content may be responsible for the considerable changes in  $T_g$  with change in mol % of (IV).

All the copoly (amide-imide)s showed a similar pattern of thermal decomposition with no weight loss below 407  $^{\circ}\text{C}$  in nitrogen, and  $T_{50}$  in range of 560 to 655  $^{\circ}\text{C}$ . It is interesting to note that for these copoly (amide-imide)s derived from ODA and SDA have almost same thermal stability probably because the linkages of ether and sulphone have similar thermal stability. However along the series of copolymers as the mol % of (IV) increased, the thermal stability of copolymers increased in each series; as evidenced by  $T_i$  and  $T_{10}$  values.

**Table 3: Thermal analysis <sup>a</sup> of poly (amide-imide)s from (IV) with various aromatic diamines and copoly (amide-imide)s from ODA / SDA with (IV + o-BBA).**

Polymer Code	Temperature for various % Decomposition			$T_g$ (C) <sup>e</sup>
	$T_i$ <sup>b</sup>	$T_{10}$ <sup>c</sup>	$T_{50}$ <sup>d</sup>	
P-4	430	520	620	320
CPO - 1	420	455	560	252
CPO - 2	407	460	590	275
CPO - 3	410	505	610	280
CPO - 4	425	495	655	320
P -- 1	415	455	535	320
CPS - 1	407	450	585	315
CPS - 2	420	470	640	312
CPS - 3	420	480	600	340
CPS - 4	440	490	650	340
P -- 5	440	490	590	330

- Thermogravimetric analysis at heating rate of 10 $^{\circ}\text{C}/\text{min}$  under nitrogen.
- Temperature at which initial wt. loss observed.
- Temperature at which 10% wt. loss observed.
- Temperature at which 50% wt. loss observed.
- Determined by DSC.

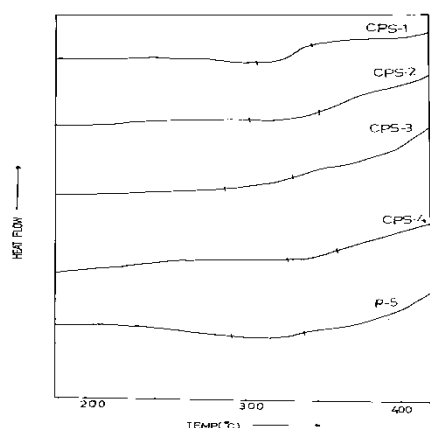


Fig. 3: DSC curves of CPS-1 to CPS-4 and P-5.

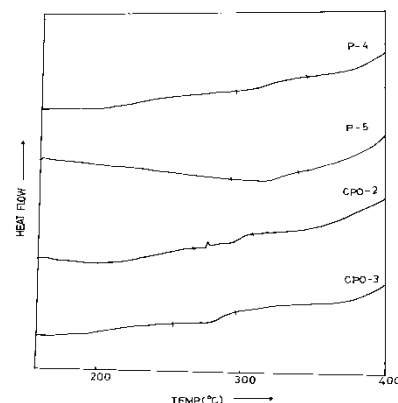


Fig. 4: DSC curves of P-4, P-5 and CPO-2 and CPO-3.

The crystallinity of poly (amide-imide) and copoly (amide-imide)s was examined by means of wide-angle X-ray diffraction. The polymer P-5, containing sulphone linkages and bulky tetraphenyl pendant groups, showed an amorphous nature, whereas the wide angle X-ray diffractograms of the polymer P-4 indicated that the polymer P-4 was semicrystalline (Fig. 5). This observation is reasonable, because the presence of the bulky pendant group and sulphone moiety caused decrease in crystallinity in P-5, whereas in P-4 probably rigid naphthalene ring in polymer chain caused crystallinity. The copolymers CPS- 1 to CPS-4 and CPO- 1 to CPO-4 as expected showed more amorphous nature XRD due to random disordered sequence of co-monomers and phenylated moiety in these copolymers.

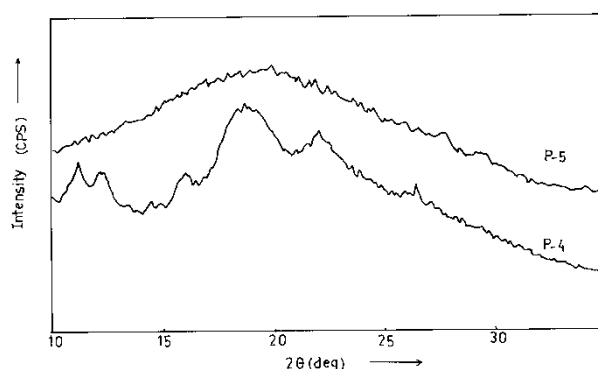


Fig. 5: XRD pattern of P-4 and P-5.

#### IV. CONCLUSION

New, poly (amide-imide) and copoly (amide-imide)s were synthesized from (IV) using the direct polycondensation method from 1,5-naphthalene diamine; and from ODA / SDA by employing various mol % of (IV) with oxybis (benzoic acid) and bis(4-aminophenyl) ether / bis(4-aminophenyl) sulfone. Polymers and copolymers were characterized by IR, viscosity, solubility, thermal analysis and XRD. Copolymers had better solubility because of random, symmetry-disturbing arrangement of bulky moieties and amorphous nature of copolymers. Thus the approach of incorporation of phenylated monomer (IV), sulphone or ether linkages and copolymerization was synergistically more effective for improving the solubility without sacrificing the thermal stability of poly (amide-imide)s polymers.

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