Synthesis and Characterization of Diamide–Diimide–Diamines Based on *p*-Amino Benzoic Acid and Their Curing and Thermal Behavior with Epoxy Blends Containing Phosphorus/Silicon in the Main Chain

Geeta Durga, Anudeep Kumar Narula

School of Basic and Applied Sciences, Guru Gobind Singh Indraprastha University, Dwarka, New Delhi 110075, India

ABSTRACT: Diimide–diacid (**I**) having an imide group in its rigid structure was synthesized by the refluxing of 3,3′,4,4′-benzophenonetetracarboxylic dianhydride [4,4′-carbonyldiphthalic anhydride (BTDA)] and *p*-amino benzoic acid in a mixture of acetic acid and pyridine (3 : 2 v/v). The chloroderivative of the diacid (**I**) was synthesized by its reaction with thionyl chloride, this was followed by condensation with different diamines with phenyl, naphthyl, ether, sulfide, and cardo groups to generate a series of diamide–diimide–diamines (DADIDAs). The resultant DADIDAs were characterized by elemental analysis and spectroscopic techniques, namely, Fourier transform infrared spectroscopy and NMR spectroscopy, and were used as epoxy curing agents to impart flame retardancy to the epoxy system. Two epoxy blends (designated as ES and EP) were prepared by the homogeneous mixing of diglycidyl ether of bisphenol A (DGEBA) with 1,3-bis(3-glycidyloxypropyl)tetramethyl disiloxane and DGEBA with tris(glycidyloxy)phosphine oxide: each in a ratio of 3 : 2 respectively. The synergistic effect of phosphorus/silicon with nitrogen on the thermal properties of the modified epoxy system was studied. The curing behavior of the epoxy resins formulated by the reaction of stoichiometric amounts of ES/EP with the synthesized DADIDAs were determined by differential scanning calorimetry, and the thermal stabilities of the cured epoxies were evaluated by thermogravimetric analyses (TGAs) under nitrogen and air. TGA indicated that the residual weight percentage of polymers at 800°C was in the range 36.4–60.0 in nitrogen, and in air, it was up to 6.5. However, the major loss in weight in air occurred at elevated temperature; this demonstrated their potential use as flame-retardant epoxy systems for electronic/electrical encapsulants. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 124: 3685–3694, 2012

Key words: flame retardance; thermal properties; thermogravimetric analysis (TGA)

INTRODUCTION

Epoxy resins, having attractive properties and excellent processability; have gained wide commercial acceptance as encapsulating materials in the electronic/electrical industries and as advanced composite matrices for structural laminates over the past few decades. In these emerging areas of applications of epoxy resins, a high thermal stability or good flame retardancy has become crucial.[1–3] In general, polymers with relatively strong main-chain bonds and/or aromatic and heterocyclic structural units, such as polyquinoxalines and polyimides or imido compounds, are known to enhance the thermal stability of epoxy resins.[4–13] Imide structures can be incorporated into epoxy resin by a reactive polyimide,[4] poly(amic acid),[5] or imidomodified curing agents.[6] The thermal stability of the resultant imide–epoxy resins leads to a significant improvement, in terms of both the initial decomposition temperature (IDT) and the integral procedural decomposition temperature.[14] Furthermore, imide groups present in the epoxy resin provide an increase in char formation in the condensed phase, which improves the flame-retardant properties of the polymers. Epoxy resins modified with imide compounds through chemical reactions or physical blending have been reported to show good flame-retardant properties.[15]

Another method for improving the flame retardancy of epoxy formulations is the incorporation of phosphorus and/or silicon[16–20] into the epoxy matrix to replace conventional brominated compounds, the use of which has been banned because of environmental concerns and legislation. The incorporation of covalently bonded phosphorus/silicon into epoxy can be achieved with phosphorus/silicon-containing oxirane compounds or curing agents.[21] As a reactive-type flame retardant, an oxirane compound with a phosphorus/silicon in the backbone of an epoxy resin exhibited much better flame retardance and overcame several drawbacks associated with the physical blending of epoxy resins and flame retardants.[22,23] Several studies have revealed that a

synergistic combination of phosphorus/silicon with nitrogen could further lead to a significant improvement in the flame resistance of the cured epoxy resin.[24,25] Wang and Zhang[26] demonstrated the formation of crosslinked char for leveling up the flame retardance due to the synergetic effect between phosphorous and nitrogen. Wang et al.[27] studied an epoxy system having silicon in the main chain and cured with nitrogen-containing hardeners and observed an increase in the thermal stability in terms of a high char yield (CR).

Therefore, in view of these studies, we considered it of interest to use both the facts, that is, the incorporation of imide groups and phosphorous/silicon oxiranes for improving flame retardance and the generation of an epoxy system of combined compositions to exhibit a high efficiency in flame retardancy and thereby provide an approach for reducing the amounts of flame-retardant additives. Up to this point, very few studies have been reported that used diamide–diimide–diamines (DADIDAs) as curing agents or that used diglycidyl ether of bisphenol A (DGEBA) only.[28] However, in this study, epoxy blends containing either phosphorus or silicon were prepared by the mixture of the conventional epoxy resin DGEBA with epoxides having phosphorous/silicon, each in a ratio of 3 : 2 and cured with DADIDA hardeners. A series of these hardeners (containing preformed imide groups) were synthesized by the condensation of a diimide–diacid (I; DIDA) chloride with five different diamines, that is, 1,4-phenylene diamine (P), 1,5-diamino naphthalene (N), 4,4′-(9-fluorenyllidene) dianiline (F), 4,4′-diaminodiphenyl sulfide (DS), and 3,4′-oxydianiline (O); these were designated as BAP, BAO, BADS, BAN, and BAF, respectively. I (DIDA) was prepared by the reaction of 1 mol of 3,3′,4,4′-benzophenonetetracarboxylic dianhydride [4,4′-carbonyldiphthalic anhydride (BTDA)] and p-amino benzoic acid (PABA) in the presence of an acetic acid (AcOH)/pyridine (Py) mixture (3 : 2 v/v), which was converted into DIDA chloride on reaction with thionyl chloride (SOCl₂). Various epoxy resins were generated by the reaction of a stoichiometric amount of ES/EP with the DADIDAs, and their curing behavior and thermal stability were investigated.

EXPERIMENTAL

Materials

DGEBA (grade LY556 having EEW 177) was procured from Hindustan Ciba Geigy, Ltd. (Mumbai, India) 1,3-Bis(3-glycidyloxypropyl)tetramethyl disiloxane (BGPTMSO; EEW180), BTDA, P, N, F, DS, and O were all purchased from Sigma Aldrich and were used as received. PABA (Merck), glacial acetic acid [Sisco Research Lab (SRL)], and sodium bicar-

bonate (NaHCO₃, SRL) were used without further purification. Py (Merck) was purified by distillation under reduced pressure over calcium hydride. Dimethylformamide (DMF; Merck) was dried under P₂O₅ and distilled *in vacuo*. Methanol (SRL) was dried over sodium and distilled.

The structural formulas of BGPTMSO (1, EEW 180) and synthesized[29] tris(glycidyloxy)phosphine oxide (2; EEW 120) were as follows:
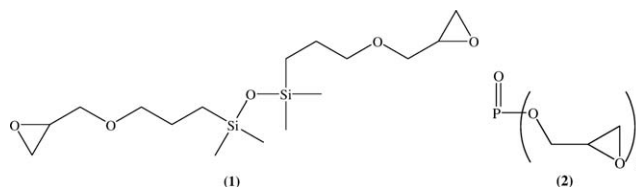
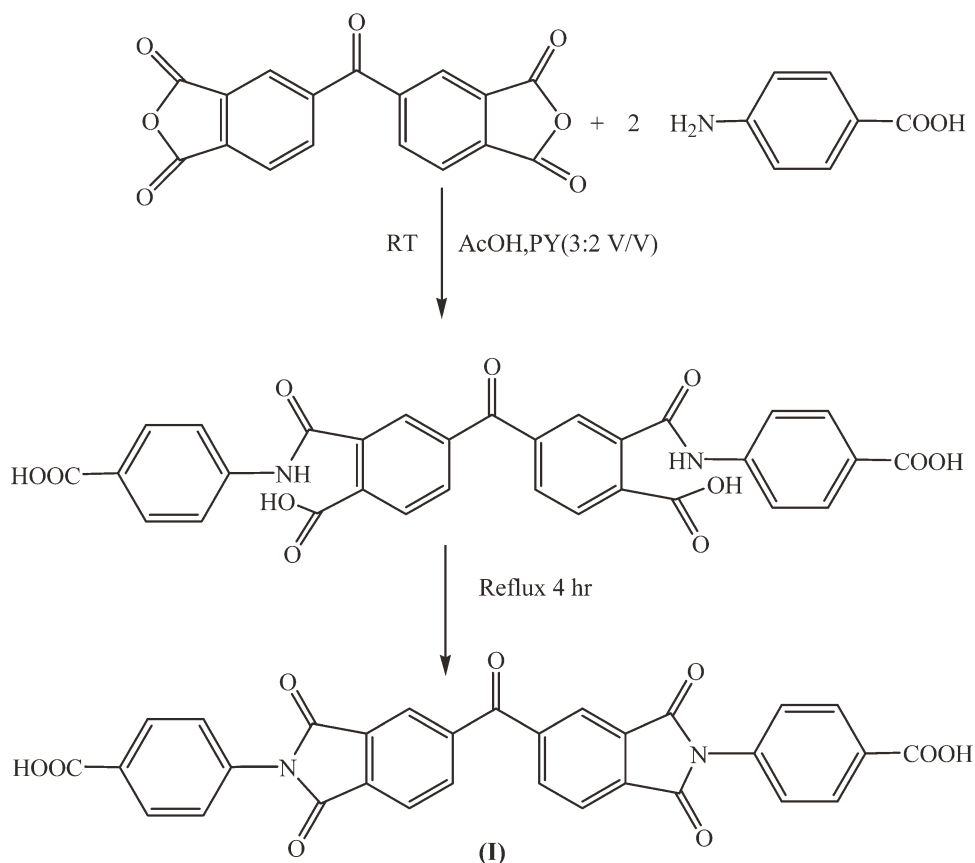### Synthesis

Step 1: Synthesis of I (DIDA)

Dianhydride (3.22 g, 10 mmol), BTDA, PABA (2.74 g, 20 mmol), acetic acid (40 mL), and Py (3 : 2 v/v) were placed in a 250-mL, round-bottom flask equipped with a stirring bar. The mixture was stirred at room temperature overnight and refluxed for 4 h[30] (Scheme 1). The solvent was removed under reduced pressure, and the dark brown viscous residue so obtained was dissolved in 100 mL of cold water. Then, the solution was decanted, and 5 mL of concentrated HCl was added. The solution was stirred until a light brown precipitate was formed. The precipitate was then filtered off and dried to give N,N′-(carbonyldiphthaloyl) bis-p-aminobenzoic diacid (I; yield 88%).

Step 2: Synthesis of DADIDA

I (3.00 g, 5.3 mmol) and thionyl chloride (10 mL, an excess amount) were placed into a 50-mL, round-bottom flask, and the mixture was refluxed for 5 h. Then, the reaction mixture was stirred at room temperature for 2 h. The excess thionyl chloride was removed first via distillation and then with dry benzene. To the well-stirred solution of N,N′-(carbonyldiphthaloyl) bis-p-aminobenzoic diacid chloride in DMF, F (11 mmol) in dry DMF was added dropwise, and the mixture was stirred further for 3–4 h at room temperature (Scheme 2). We precipitated the resulting DADIDA by pouring the mixture into ice-cold water with constant stirring. The product was washed with NaHCO₃ repeatedly to remove unreacted acid and then with water and was finally washed thoroughly with hot methanol and dried at 60°C for 10 h *in vacuo*. Other DADIDAs (Scheme 2) were prepared with the same procedure.

Characterization

The structures of the synthesized diamide–diacids (DIDA) and DADIDA were characterized with

Scheme 1 Synthesis of DIDA (RT = room temperature).

elemental analysis (EA), Fourier transform infrared (FTIR) spectroscopy, ^{1}H-NMR spectroscopy, and ^{13}C-NMR spectroscopy. EA was carried out with a Euro EA 3000 elemental analyzer (Milan, Italy). The FTIR spectra were recorded in the range 4400–600 cm[−1] by a Shimadzu-8700 FTIR spectrophotometer with KBr pellets. ^{1}H-NMR and ^{13}C-NMR spectra were recorded with a Bruker AVANCE II Fourier transform NMR spectrophotometer at frequency of 400 MHz with deuterated dimethyl sulfoxide (DMSO-d_6) as a solvent and tetramethylsilane as an internal reference. These DADIDAs were used as epoxy curing agents.

Curing studies

Two blends of epoxy resins (ES and EP) were prepared by the homogeneous mixing of DGEBA with BGPTMSO and **2,** respectively, each at a ratio of 3 : 2. The polymer samples were prepared by the homogeneous mixing of a stoichiometric amount of synthesized DADIDAs with ES and EP with a mixture of ethyl methyl ketone and DMF as a solvent. Later, the solvent was stripped off *in vacuo*. The two series of polymers hence generated were designated as BAP/ES, BAO/ES, BADS/ES, BAN/ES, and BAF/ES and BAP/EP, BAO/EP, BADS/EP, BAN/EP, and BAF/EP, respectively. A TA 2100 thermal analyzer having

a DSC (New Castle, DE, USA) 910 module was used to record the differential scanning calorimetry (DSC) scans to evaluate the curing behavior of the epoxy resins under a nitrogen atmosphere (50 cm[3]/min) at a heating rate of 10°C/min from room temperature up to 300°C. Freshly prepared samples with weights of 4 ± 2 mg were used for each DSC experiment.

Thermal stability

The epoxy thermosets prepared as discussed previously were cured isothermally by heating at 180°C (2 h) followed by postcuring at 250 ± 10°C in an air oven. To observe the weight loss behavior and thermal stability of the cured resins, thermogravimetric analysis was done with a PerkinElmer Diamond thermogravimetry (TG)/dynamic thermal analysis instrument (California, USA). The instrument was programmed at a heating rate of 10°C/min under nitrogen/air flowing at a rate of 20 cm[3]/min, and powdered samples of the size 6 ± 2 mg were used in each experiment.

RESULTS AND DISCUSSION

Structural characterization of DADIDAs

Polyimides are well-known high-performance, heat-resistant polymers, but their low processability and
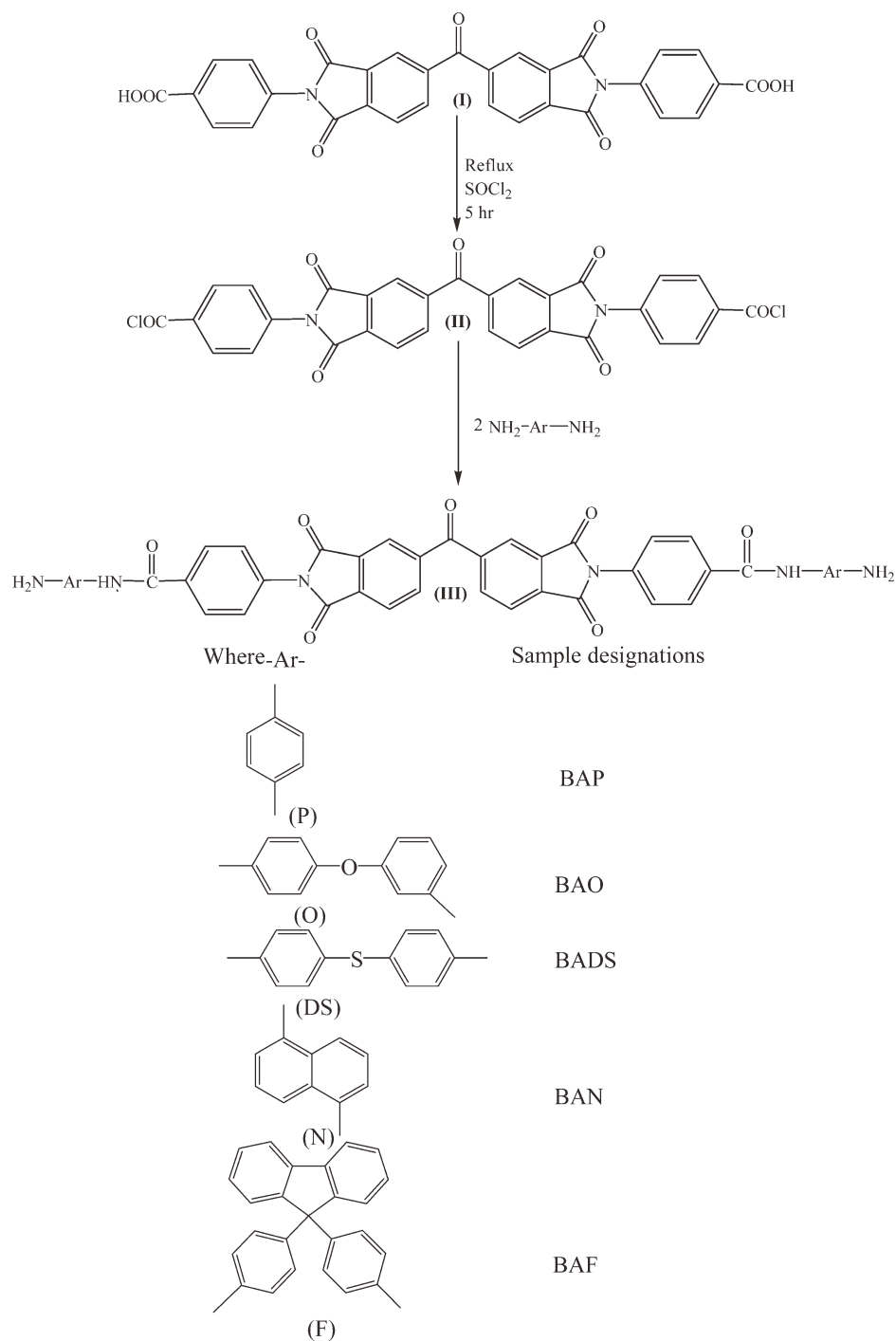
Scheme 2 Synthesis of DADIDA.

solubility have limited their applications.[11,30] Various attempts have been made to improve the solubility and processability of these materials, including the incorporation of flexible linkages and bulky groups such as amide and phenyl/naphthyl/4,4-oxo diphthalyl into the polymer backbones of aromatic polyimides and polyarylates without any significant reduction in the thermal stability.

An aromatic diacid (**I**) having a rigid imide group was synthesized via the nucleophilic substitution of

PABA with dianhydride (BTDA) in the presence of a mixture of Py and glacial acetic acid. The resulting DIDA was reacted with SOCl$_2$ to activate the —COOH group and then condensed with various diamines having phenyl, naphthyl, ether, sulfide, and cardo groups to generate a series of DADIDAs. The DIDAs and DADIDAs were cream white to brown in color and soluble in ethyl methyl ketone, chloroform, DMF, and DMSO and were obtained in the range 73–87%.

TABLE I
CHN Analytical Data of the DADIDAs

Sample designation	Molecular formula	Found (calculated; %)		
		C	H	N
BAP	$C_{43}H_{28}N_6O_7$ (741)	69.45 (69.72)	3.90 (3.81)	11.21 (11.35)
BAO	$C_{55}H_{36}N_6O_9$ (925)	71.33 (71.42)	3.85 (3.92)	8.99 (9.09)
BADS	$C_{55}H_{36}N_6O_7S_2$ (957)	68.95 (69.02)	3.82 (3.79)	8.67 (8.78)
BAN	$C_{51}H_{32}N_6O_7$ (841)	72.68 (72.85)	3.77 (3.84)	9.74 (9.99)
BAF	$C_{81}H_{52}N_6O_7$ (1221)	79.42 (79.66)	4.40 (4.29)	6.76 (6.88)

The structures of DIDA and DADIDAs were confirmed by EA and spectroscopic methods. Table I shows the results of CHN analysis. The elemental analyses results were in good agreement with the calculated percentages of carbon, hydrogen, and nitrogen.

The FTIR spectrum of DIDA showed a broad band at 3500–2800 cm^{-1} (O—H stretching), a weak band at 2902 cm^{-1} (C—H stretching), and a band of medium intensity at 1388 cm^{-1} (C—N stretching). Two absorption bands characteristic of imide rings were observed around 1720 and 1784 cm^{-1}. A stretching band due to the C=O group of carboxylic acids was also observed at 1654 cm^{-1}. The FTIR spectra of all of the DADIDAs of varied structures exhibited characteristic bands at 3370 ± 20 cm^{-1} (v, N—H), 1778 ± 4 cm^{-1} (v_{asymm}, C=O), 1718 ±2 cm^{-1} (v_{symm}, C=O), 1660 ± 10 cm^{-1} (v, N—H) of amide, 1360± 20 cm^{-1} (v, C—N) of imide, and 713 ± 2 cm^{-1} (imide heterocycle). The disappearance of the broad spectrum in the range 2800–3500 cm^{-1} in the spectrum of the DADIDAs confirmed the occurrence of the reaction. The spectra of DIDA and BAF are shown in Figure 1(a,b), respectively.

In the ^{1}HNMR spectra of DADIDAs, a broad peak due to amine (—NH$_2$ group) was observed at δ = 3.4–4.2. A singlet due to the secondary amide (—CONH—) group was also observed at δ = 7.9–8.0, whereas a complex pattern of signals in the region of δ = 6.7–8.0 pertained to aromatic protons. The signals due to the aromatic protons of BTDA appeared downfield from the others. A peak at δ = 10.5 (due to —COOH protons) was absent in all of the spectra of DADIDAs; this indicated the occurrence of the reaction. The ^{1}H-NMR and ^{13}C-NMR spectra of the samples BADS and BAN, representative of the DADIDAs, are shown in Figures 2 and 3, respectively. In the ^{13}C-NMR spectra, the resonance lines in the spectrum fell into three regions, that is, δ = 113–142 for aromatic carbons, δ = 165–168 for carbonyl carbons of the amide and imide groups, and δ = 193 for the *C=O of the BTDA moiety.

Curing studies

The curing reaction of the DADIDAs with the epoxy blends were directly observed with DSC monitoring.

The DSC scans of ES/EP containing stoichiometric amounts of DADIDAs exhibited two exothermic transitions in the temperature range 70–300°C. The exotherm at lower temperature may have been due to the epoxy–amine reaction, and the high-temperature exotherm may have been due to etherification and the cocuring of resins. The resulting products were, therefore, expected to possess high crosslinking densities. Figure 4 shows the DSC thermograms of thermosettable BAO/ES and BADS/EP epoxy resins, respectively, at a heating rate of 10°C/min, and the characteristic DSC temperatures are presented in Table II.

The curing of epoxy resin proceeds by the nucleophilic attack of amine on the oxirane carbon. Generally, a curing system exhibiting a lower exothermic peak temperature under the same set of curing conditions is more reactive (higher nucleophilicity) for the epoxy resins. However, in this study, the peak temperature of curing of ES/EP not only depended on the nucleophilicity but also on the amine structure and epoxy blend. The temperatures of the exothermic peak of most of the samples were higher for the blend EP than for ES. It was found to be highest for BAP/EP for the first and second transitions. However, the curing of the epoxy blends with these amines was a complex process, and the parts of
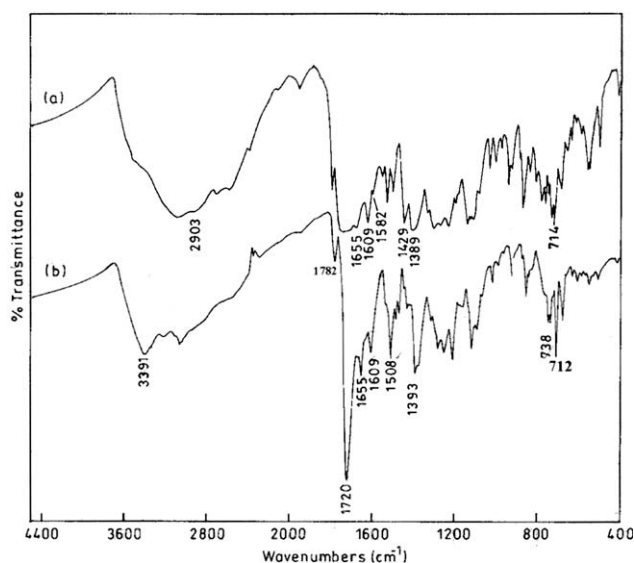

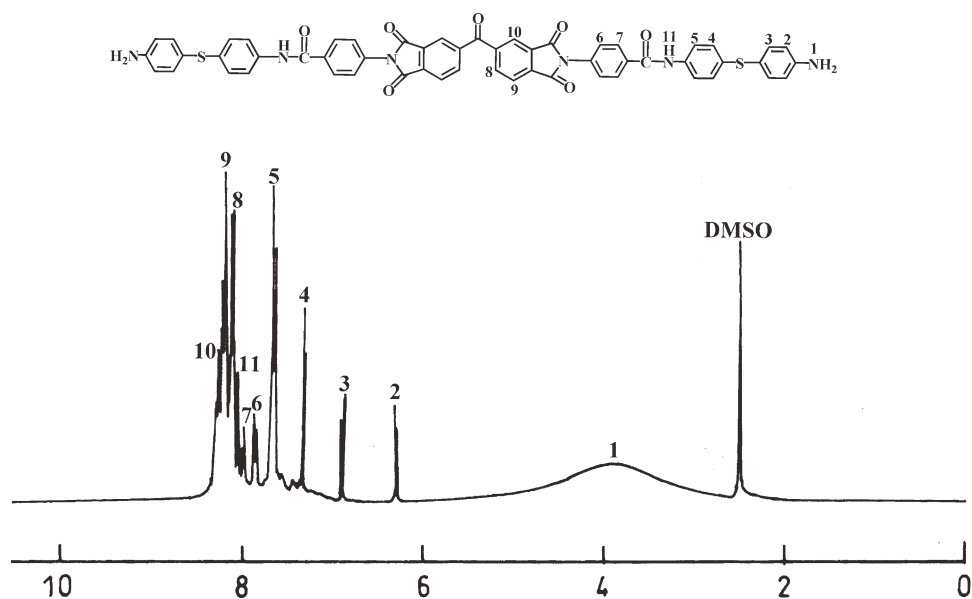**Figure 1** IR spectra of (a) DIDA and (b) BAF.

Figure 2 ^{1}H-NMR spectrum of the DADIDA BADS.

amines per hundred parts of resin used were significantly different from each other because of variation in the molecular weights. This basically reduced the epoxy content per gram of resin with increasing molecular weight of the amine. Therefore, the factors affecting the curing process were not only the nucleophilicity of the amine but also the diffusion processes and the reduced oxirane content

Thermal stability

We determined the thermal stability of the isothermally cured epoxy blend with a stoichiometric amount of DADIDAs by recording TG/differential thermogravimetry (DTG) traces in a nitrogen/air atmosphere (Table III). The TG/DTG traces of some of the isothermally cured samples in nitrogen and air are shown in Figures 5 and 6, respectively. We
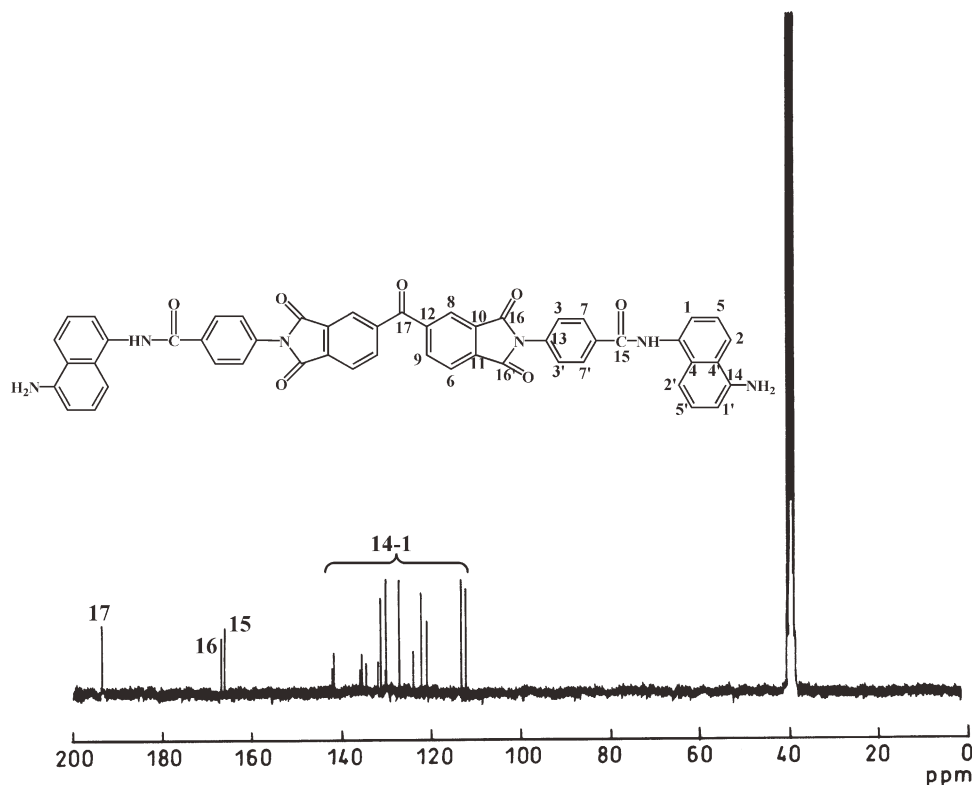

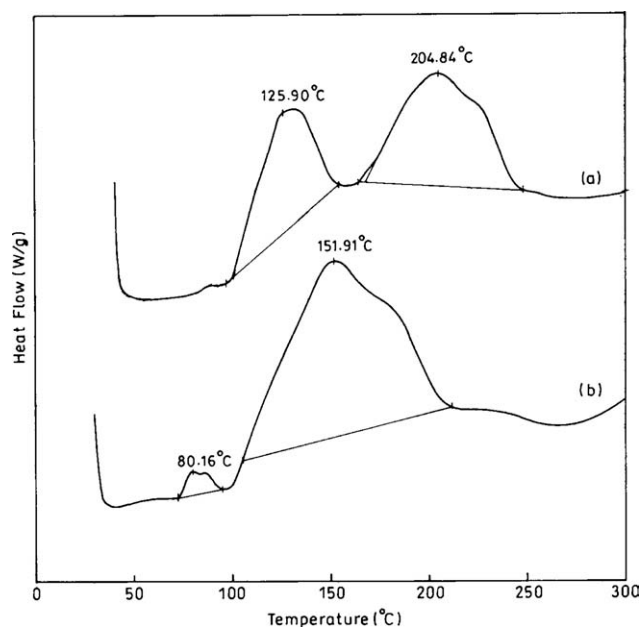**Figure 3** ^{13}CNMR spectrum of the DADIDA BAN.

Figure 4 DSC scans at a heating rate of 10°C/min of (a) ES in the presence of a stoichiometric amount of BAO and (b) EP in the presence of a stoichiometric amount of BADS.

compared the relative thermal stability of the cured resins by noting IDT, final decomposition temperature (FDT), temperature of maximum weight loss (T_{max}), and percentage CR at 800°C. All of the samples in the nitrogen environment were stable in the range 250–336°C, depending on the structure;

TABLE II
Results of the DSC Scans of ES/EP in the Presence of a Stoichiometric Amount of DADIDA at a Heating Rate of 10°C/min

Sample designation	T_i (°C)	T_{onset} (°C)	T_{max} (°C)	T_f (°C)
BAP/ES	105.4	105.4	130.3	141.8
	148.91	151.8	213.4	266.2
BAO/ES	96.3	100.8	125.9	164.2
	167.9	154.2	204.8	248.1
BADS/ES	71.4	72.4	79.3	92.7
	105.7	105.7	161.7	223.6
BAN/ES	71.1	71.2	85.6	101.6
	129.4	134.9	169.3	209.3
BAF/ES	129.0	131.6	145.4	167.0
	172.0	180.0	216.1	252.0
BAP/EP	138.6	141.5	163.4	197.3
	202.9	210.4	239.8	252.4
BAO/EP	142.2	142.3	158.9	184.5
	185.5	186.0	213.7	231.0
BADS/EP	71.4	73.2	80.2	94.9
	104.8	105.4	151.9	211.8
BAN/EP	94.2	97.0	107.1	117.6
	130.4	130.5	147.7	177.7
BAF/EP	68.8	71.1	83.1	97.4
	164.6	178.0	237.2	296.1

T_i, Kick off temperature, where curing starts; T_{onset}, temperature where the first detectable heat was released this was obtained by extrapolation of the steepest portion of the curve; T_p, temperature on the peak of the exotherm; T_f, temperature at the end of the curing exotherm obtained by extrapolation of the curve.

TABLE III
Results of the TG/DTG Traces of Isothermally Cured Epoxy Thermosets Scanned at a Heating Rate of 10°C/min under Nitrogen and Air

| Sample designation	Nitrogen					Air				
	IDT (°C)	T_{max} (°C)	FDT (°C)	Age CR (%; 800°C)	Calculated LOI (%)	IDT (°C)	T_{max} (°C)	FDT (°C)	Age CR (%; 800°C)	Calculated LOI (%)
BAP/ES	336.8	363.4	426.3	39.3	33.2	340.5	594.7	615.7	5.4	19.7
	478.9	505.3	594.7							
BAO/ES	301.1	374.4	418.9	36.4	32.1	397.8	512.6	632.9	4.1	19.1
	503.2	528.2	565.2							
BADS/ES	321.0	357.0	407.7	44.5	35.3	366.7	485.9	558.0	6.3	20.0
	489.4	543.4	615.8							
BAN/ES	285.2	294.7	350.5	53.5	38.5	255.3	304.7	357.9	5.8	19.8
	493.1	525.1	557.4			539.1	606.6	611.7		
BAF/ES	289.1	299.2	341.1	46.1	35.9	388.4	571.8	593.5	4.9	19.5
	490.7	522.1	570.0							
BAP/EP	302.1	370.3	425.7	43.0	34.7	312.6	580.1	605.4	5.2	19.6
	468.1	525.7	619.0							
BAO/EP	290.1	345.4	415.2	47.9	36.7	386.7	494.5	625.0	4.3	19.2
	505.0	530.6	590.0							
BADS/EP	280.6	312.7	419.2	55.8	40.0	323.8	473.2	530.7	5.9	19.9
	446.0	510.6	659.3							
BAN/EP	*253.5*	*290.6*	*410.8*	*60.0*	*41.5*	*283.6*	*287.5*	*313.4*	*6.5*	*20.1*
	513.7	*546.4*	*685.4*			*540.2*	*571.6*	*580.3*		
BAF/EP	277.1	296.4	368.1	50.0	37.5	376.9	572.4	581.2	5.1	19.5
	527.3	565.1	647.3							
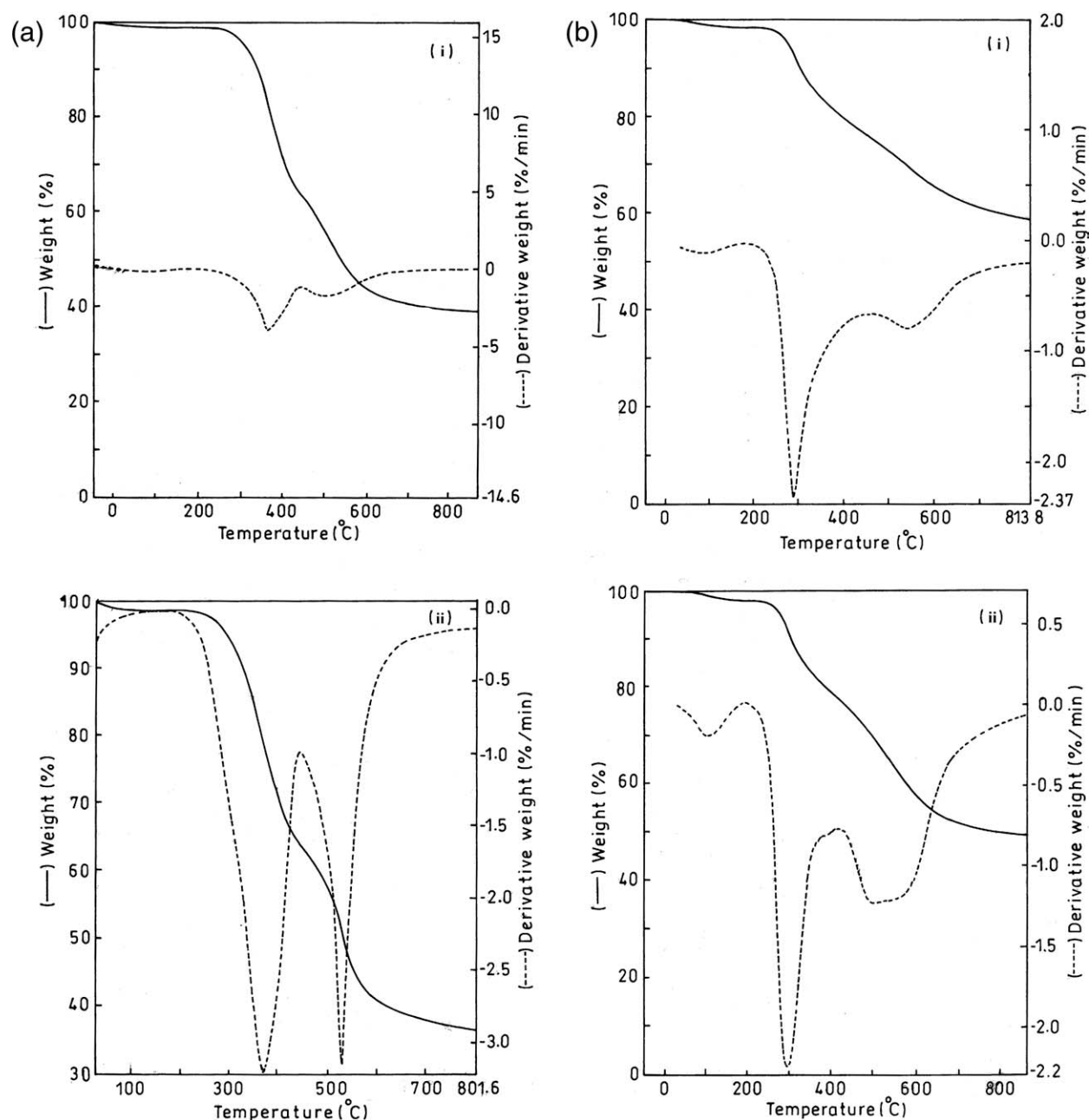
Figure 5 TG/DTG traces at a heating rate of 10°C/min under nitrogen of (a) ES cured isothermally with BAP and BAO and (b) EP cured isothermally with BAN and BAF.

20–30% of weight loss occurred in first step, and the remaining weight loss occurred in the second step. Phosphorus/silicon-containing epoxy resins did not show a significant change in IDT even because of having rigid imide group but exhibited a multistep degradation with a higher final CR compared to the system where only DGEBA was taken as a base resin.[28,29] During the first degradation step, a thermally stable char was formed and served as an outer layer to prevent the further degradation of the bulk; this, thus, improved the flame retardancy. However, in an air atmosphere, all of the samples except BAN showed single-step degradation, but their major weight loss occurred at a much ele-

vated temperature compared to that under the nitrogen, and they exhibited a CR up to 6.5.

The ability to form char is related to the flammability of a polymer. The higher the amount of residual char is after combustion, the lower the amount of combustible material will be available to perpetuate the flame and the greater is the degree of flame retardance of the material.[31] Therefore, one of the ways to achieve high degrees of flame retardancy or noncombustability of a polymeric material is to increase the amount of char produced on combustion. Van Krevelen[32] proposed that the char residue on pyrolysis is linearly proportional to
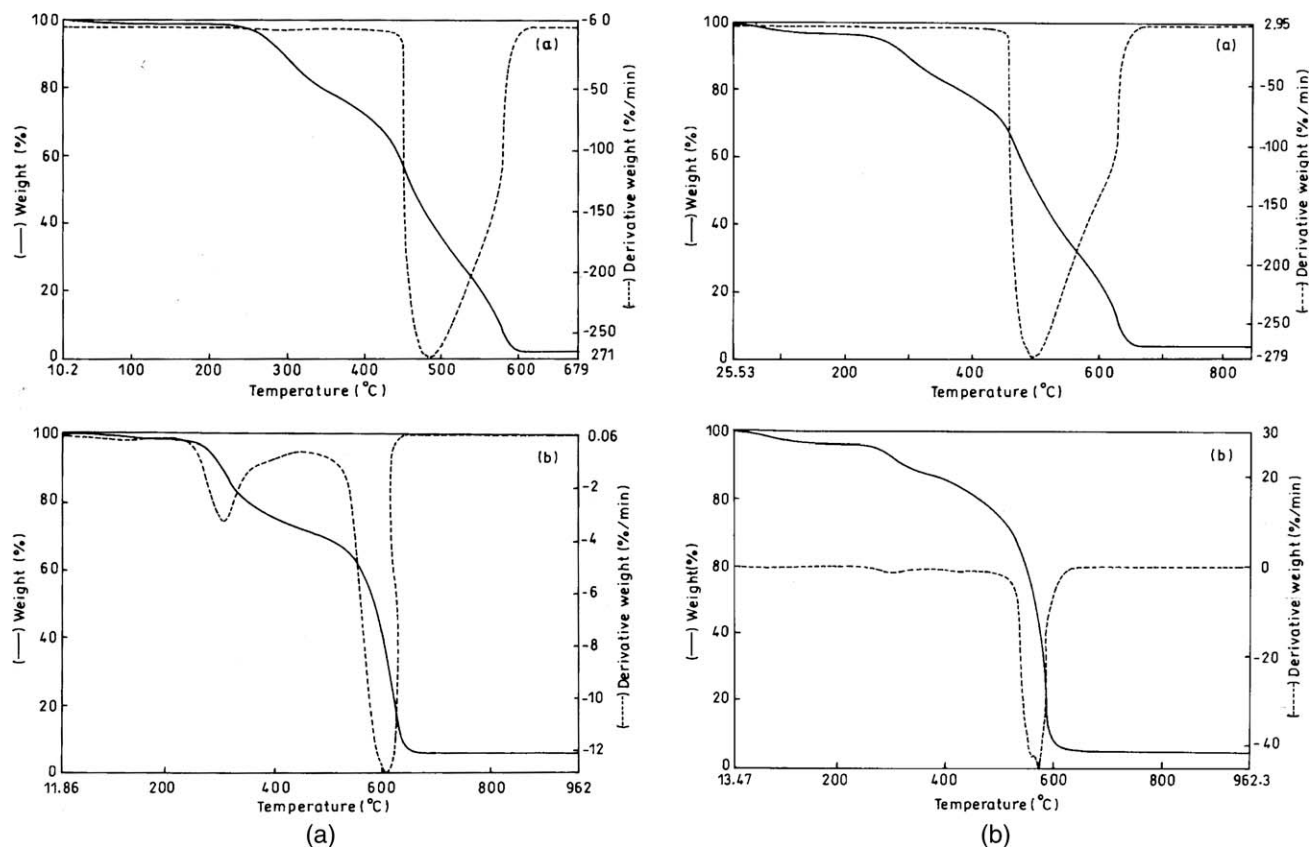
Figure 6 TG/DTG traces at a heating rate of 10°C/min in air of (a) ES cured isothermally with BAS and BAN and (b) EP cured isothermally with BAO and BAF.

the limiting oxygen index (LOI) for a halogen-free polymer:

$$LOI = 17.5 + .4CR$$

The LOI was calculated for all of the samples with this equation and was found to be in the range 32.1–41.5 under the nitrogen atmosphere; in air, it was calculated in the range 19.1–20.1. Generally, epoxy resins having LOIs greater than 28 are considered to be flame retardant. The blended epoxy resin having phosphorus in the main chain, that is, EP cured with DADIDAs, showed better CRs than those having silicon, that is, ES cured with DADIDAs. However, in both series, the epoxy blends cured with BAN showed the highest CR in nitrogen; this may have been due to the compact, more rigid aromatic ring in the naphthalene moiety.

CONCLUSIONS

In this study, the synergistic effect of phosphorous/silicon with nitrogen on the flame retardancy of epoxy resin was studied. For this purpose, nitrogen-containing DADIDA curing agents were synthesized and characterized by EA and spectroscopic techniques. In the newly formulated epoxy resins, the DADIDA curing agents were reacted with DGEBA blended with a phosphorus/silicon-containing reactive comonomer. The curing behavior of the epoxy resins were evaluated by DSC, and the thermal stability and flame retardancy of the isothermally cured resins were determined by thermogravimetric analysis. The incorporation of phosphorus/silicon along with nitrogen into the polymer structures produced adequate CRs (36.4–60.0) on pyrolysis in the nitrogen atmosphere. The values of LOI calculated were found to be in the ranges 32.1–41.5 in nitrogen and 19.1–20.1 in air. The blends having phosphorus in the main chain, that is, EP cured with DADIDAs, showed better CRs in nitrogen than those having silicon, that is, ES cured with DADIDAs. The highest value of CR (60.0) was obtained in the case of BAN/EP in nitrogen and air.

References

1. Lee, H.; Neville, K. Handbook of Epoxy Resins; McGraw-Hill: New York, 1967.
2. Penn, L. S.; Chiao, T. T. In Handbook of Composites; Lubin, G., Ed.; Van Nostrand Reinhold: New York, 1982; p 57.
3. Bianchini, G.; Dyer, R.; Fream, A.; Heffer, P.; McOwan, S. P.; Oldring, P. Waterborne and Solvent Based Epoxies and Their End User Application; Wiley: New York, 1997.

4. Hay, J. N.; Woodfine, B.; Davies, M. High Perform Polym 1996, 8, 35.

5. Gaw, K.; Kikei, M.; Kakimoto, M.; Imai, Y. Polymer 1997, 38, 4413.

6. Agag, T.; Takeichi, T. Polymer 1999, 40, 6557.

7. Kim, W. G.; Nam, T. Y. J Polym Sci Part A: Polym Chem 1996, 34, 957.

8. Jeng, R.-J.; Lo, G.-S.; Chen, C.-P.; Liu, Y.-L.; Hsiue, G.-H.; Su, W.-C. Polym Adv Technol 2003, 14, 147.

9. Li, C.; Qiu, W. L.; Hua, W. S.; Lu, L.; Wang, X. Thermochim Acta 1995, 255, 355.

10. Liu, Y. L.; Chen., Y. J. Polymer 2004, 45, 1797.

11. Isfahani, H. N.; Faghihi, K.; Hajibeygi, M.; Bokaei, M. Polym Bull 2010, 64, 633.

12. Kumar, A. A.; Alagar, M.; Rao, R. M. V. G. K. J Appl Polym Sci 2001, 81, 2330.

13. Jain, P.; Choudhary, V.; Varma, I. K. Eur Polym J 2003, 39, 181.

14. White, L. A.; Weber, W. J.; Mathias, L. J. Polym Bull 2001, 46, 463.

15. Varma, I. K.; Gupta, A. K.; Sangita; Varma, D. S. J Appl Polym Sci 1983, 28, 191.

16. Wang, X. D.; Lin, J. J. Colloid Polym Sci 2005, 283, 593.

17. Liu, Y. L.; Hsiue, G. H.; Lee, R. H. J Appl Polym Sci 1997, 63, 895.

18. Chen, Y. W.; Lee, H. F.; Yuan, C. Y. J Polym Sci Part A: Polym Chem 2000, 38, 972.

19. Ai, H.; Xu, K.; Liu, H.; Chen, M.; Zhang, X. J Appl Polym Sci 2009, 113, 541.

20. Levchik, S. V.; Weil, E. D. Polym Int 2004, 53, 1901.

21. Hsiue, G.-H.; Liu, Y.-L.; Tsiao, J. J Appl Polym Sci 2000, 78, 1.

22. Cheng, X.; Shi, W. J Therm Anal Calorim 2011, 103, 303.

23. Li, H.-T.; Chuang, H.-R.; Wang, M.-W.; Lin, M.-S. Polym Int 2005, 54, 1416.

24. Liu, Y.-L.; Chang, G.-P.; Wu, C.-S. J Appl Polym Sci 2006, 102, 1071.

25. Liu, Y. L.; Chiu, Y. C.; Wu, C. S. J Appl Polym Sci 2003, 87, 404.

26. Wang, X.; Zhang, Q. Eur Polym J 2004, 40, 385.

27. Wang, W. J.; Perng, L. H.; Hsiuea, G. H.; Chang, F. C. Polymer 2000, 41, 6113.

28. Darshan, M. P.; Narula, A. K. Ind J Chem 2009, 48, 893.

29. Durga, G., Singh, D.; Kukreja, P.; Narula, A. K. Ind J Eng Mater Sci 2009, 16, 133.

30. Liu, Y. L.; Liu, Y. L.; Jeng, R. J.; Chiu Y. S. J Polym Sci Part A: Polym Chem 2001, 39, 1716.

31. Martel, B. J Appl Polym Sci 1988, 35, 1213.

32. Van Krevelen, D. W.; Hoftyzer, P. J. Properties of Polymers; Elsevier: New York, 1976.