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CURING AND THERMAL BEHAVIOUR OF EPOXY RESIN IN THE PRESENCE OF A MIXTURE OF IMIDE-AMINES

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The curing behaviour of diglycidyl ether of bisphenol-A (DGEBA) was investigated by the dynamic differential scanning calorimetry using varying molar ratios of aromatic imide-amines and 4,4'-diaminodiphenylsulfone (DDS). The imide-amines were prepared by reacting 1 mole of naphthalene 1,4,5,8-tetracarboxylic dianhydride (N) and 4,4'-oxodiphthalic anhydride (O) with 2.5 moles of 4,4'-diaminodiphenyl ether (E) or 4,4'-diaminodiphenyl methane (M) or 4,4'-diaminodiphenylsulfone (S) and designated as NE/OE or NM/OM or NS/OS. The mixture of the imide-amines and DDS at ratio of 0:1, 0.25:0.75, 0.5:0.5, 0.75:0.25 and 1:0 were used to investigate the curing behaviour of DGEBA.

A single exotherm was observed on curing with mixture of imide-amines and DDS. This clearly shows that the two amines act as co-curing agents. Curing temperatures were higher with imide-amines having sulfone linkage irrespective of anhydride. Curing of DGEBA with mixture of imide-amines and or DDS resulted in a decrease in characteristic curing temperatures. The thermal stability of the isothermally cured resins was also evaluated using dynamic thermogravimetry in a nitrogen atmosphere. The char yield was higher in case of resins cured imide-amines based on N and E. The activation energy of decomposition and integral procedural decomposition temperature were also calculated from the TG data.

Keywords: *curing, DGEBA, 4,4'-diaminodiphenylsulfone, imide-amines, thermal stability*

Introduction

Epoxy resins have been used in many industrial applications such as in surface coatings, adhesives, structural insulating materials for electronic devices because of their good chemical resistance and superior electrical and mechanical properties [1, 2]. However, the conventional epoxy resins are inefficient to satisfy the required properties in the field of advanced materials, which require high thermal and flame resistance [3–5]. Several approaches were used in the past to improve thermal stability and flame retardancy of epoxy resins. The properties can be varied by the modification of epoxy backbone, i.e., either at the time of synthesis or during cure using curing agents of varying structures.

Imide compounds and polymers [6–10] that contain aromatic and/or heterocyclic groups can offer desirable high temperature stability. Polyimide/polyamide-acid endcapped with amines have been used to impart higher thermal stability and flame resistance. Hay *et al.* [11] studied the molding of linear unreactive polyimides for toughening of epoxy–diaminodiphenyl sulfone system. They achieved increase in the fracture toughness with no loss of modulus but there was a little influence on the thermal capability across the range of modifying polyimides. Polyimide end-capped with amine or polyimide-siloxane [12]/polyamide-acid [13, 14] have been used to impart toughness and improve thermal and chemical resistance of epoxy resin. The curing process in the epoxy resins can be initiated by using a wide range of curing agents such amines, anhydrides and amino formaldehyde-resins [15–18].

In the present work, the curing and thermal behaviour of diglycidyl ether of bisphenol-A (DGEBA) has been investigated using mixture of imide-amines and 4,4'-diaminodiphenylsulfone (DDS). Curing of DGEBA in the presence of stoichiometric amounts of imide-amines was investigated using differential scanning calorimetry (DSC). Thermal behaviour of isothermally cured resins was evaluated using dynamic thermogravimetry in nitrogen atmosphere.

Experimental

Materials

Diglycidyl ether of DGEBA (grade LY556, having an epoxide equivalent of 177) was procured from Hindustan Ciba Geigy Ltd. 4,4'-Diaminodiphenyl sulfone (S), 4,4'-diaminodiphenyl ether (E), 4,4'-diaminodiphenyl methane (M) and naphthalene 1,4,5,8-tetracarboxylic dianhydride (NTDA) and

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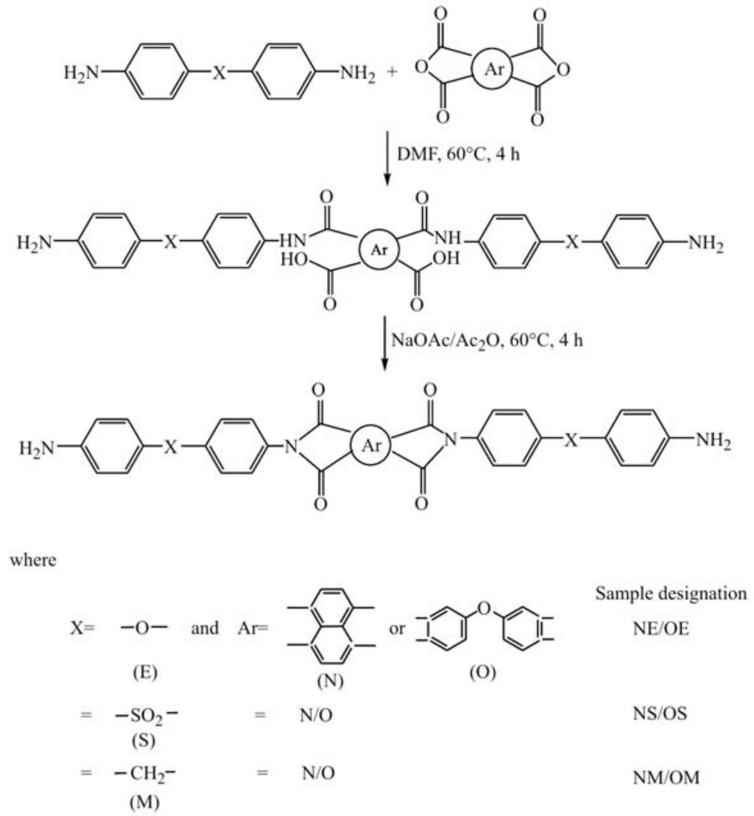

Scheme 1 Synthesis of aromatic imide-amines

4,4’-oxodiphthalic anhydride (ODPA) – all purchased from Aldrich and were used as received. N,N’-dimethyl formamide (DMF, Qualigens) was dried by keeping it over phosphorous pentoxide for 72 h followed by distillation under reduced pressure.

The imide-amines [19] were prepared according to the reaction scheme (Scheme 1).

Curing and thermal analysis

TA 2100 thermal analyzer having 910 DSC module was used for recording DSC traces at heating rate of 10°C min^{-1} and 5±2 mg of sample was used in each experiment. For curing studies, the samples were obtained by mixing stoichiometric amounts of imide-amines and DDS with DGEBA in the varying molar ratio of 0:1, 0.25:0.75, 0.5:0.5, 0.75:0.25 and 1:0 using minimum amount of low boiling solvent. After thorough mixing, the solvent was evaporated under vacuum and the freshly prepared samples were used for recording DSC traces in the static air atmosphere at a programmed heating rate from room temperature upto 350°C. The epoxy samples have been designated by adding prefix E to NE/OE or NM/OM or NS/OS followed by a numerical suffix. For example, epoxy cured using mixture of NS:DDS in the ratio of 0.25:0.75, 0.5:0.5, 0.75:0.25 and 1:0 has been designated as ENS-1, ENS-2, ENS-3 and ENS. The sample with DGEBA and DDS has been designated as ED. Similarly samples cured using mixture of other imide-amines and DDS were designated.

Thermal stability of the epoxy resins cured isothermally by heating 180±20°C for 3 h in an air oven was evaluated by recording TG/DTG traces in nitrogen atmosphere (flow rate 60 $\text{cm}^3\ \text{min}^{-1}$) using Rheometric Scientific Module. A heating rate of 20°C min^{-1} and powdered samples of 10±2 mg were used in each experiment.

Results and discussion

Structural characterization of imide-amines

The synthesis and structural characterization of the imide-amines were performed with the FTIR, ^{1}H-NMR, ^{13}C-NMR spectroscopic techniques and elemental analysis and the results were reported in an earlier work [19].

Curing studies

Figures 1 and 2 show the DSC scans of DGEBA in the presence of imide-amines ENS, ENE and ENM. The curing reaction of epoxy resins depends on the structure and reactivity of the curing agent. In the DSC traces of all the samples, a broad exothermic transi-

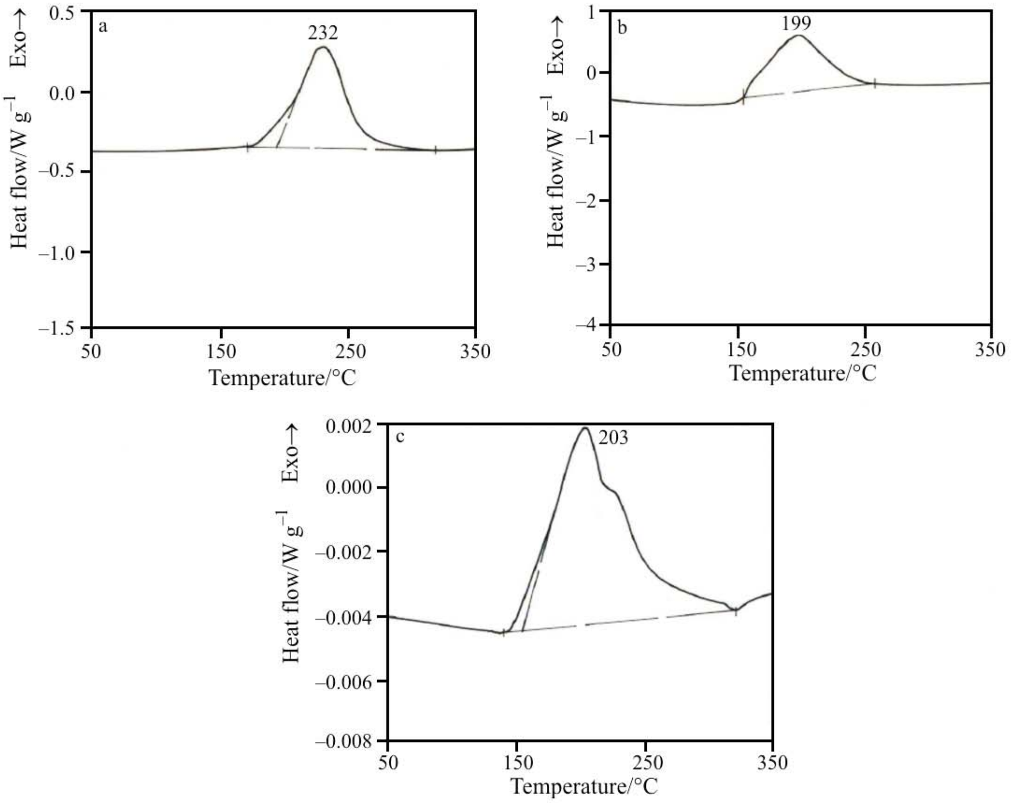

Fig. 1 DSC scans of DGEBA in the presence of imide-amines a – ENS, b – ENE and c – ENM

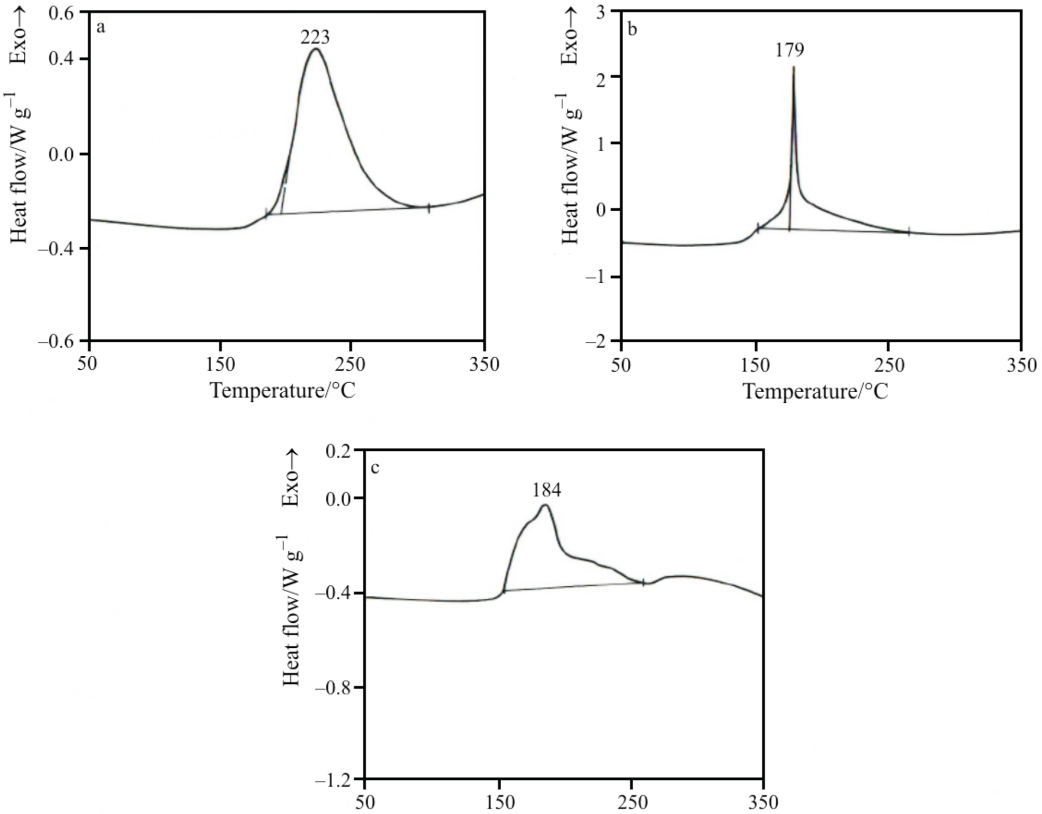

Fig. 2 DSC scans of DGEBA in the presence of imide-amines a – EOS, b – EOE and c – EOM

tion associated with curing was observed. The exothermic transition associated with curing was characterized by the following parameters:

- The initial temperature (T_i), where the curing starts.
- The temperature where the first detectable heat was released (T_{onset}).
- The temperature of the peak position of the exotherm (T_p).
- The temperature of the end of curing exotherm (T_f).
- The heat of curing calculated by the measurement of area under the exothermic (ΔH).

The results of DSC scans are summarized in Tables 1 and 2. A single curing exotherm was observed upon curing with a mixture of imide-amine and DDS. These results thus clearly showed that the two amines act as co-curing agents.

In all these samples, curing temperatures were higher with imide-amines having sulfone linkage irrespective of anhydride. This could be due to decreased nucleophilicity of amino group due to the presence of electron withdrawing sulfone groups. Several authors have already reported the increase in curing temperature with decreased nucleophilicity and hence reduces the reactivity of the amine. Comparing imide-amines of varying structure, T_p was found to be higher in ENS/EOS as compared to samples ENE/EOE or ENM/EOM.

NTDA based imide-amines showed higher T_p value. This indicates that NTDA is the least reactive and much higher temperature is needed to cure the resin. In NTDA, it is six membered ring imide whereas five-membered ring imide is present in other imide-amines. The lower reactivity could be due to its rigid structure due to the presence of naphthalene and more stability of six-membered ring as compared to five membered ring.

The DSC scans of DGEBA in the presence of mixture of NE/OM and DDS are shown in Figs 3 and 4. In the DSC scans of DGEBA cured using mixture of imide-amines (NM, NE, OM, OE), the presence of small amount of DDS in a mixture resulted in a significant increase in the peak exothermic temperature whereas in case of mixture of NS and OS, it

Table 1 Results of DSC scan of DGEBA in the presence of imide-amines NS/NE/NM, DDS and its mixture in varying molar ratios at 10°C min^{-1}

Sample designation	Molar ratios of NS/NE/NM:DDS	T_i/°C	T_{onset}/°C	T_p/°C	T_f/°C	ΔH/J g^{-1}
ED	0:1	131.1	174.6	224.0	295.7	264.4
ENS-1	0.25:0.75	143.2	173.2	214.3	297.3	277.3
ENS-2	0.5:0.5	147.1	148.9	214.5	297.3	248.2
ENS-3	0.75:0.25	140.6	180.8	207.1	299.4	293.7
ENS	1:0	170.8	194.0	231.7	317.2	179.8
ENE-1	0.25:0.75	158.8	175.4	218.8	269.9	195.7
ENE-2	0.5:0.5	165.9	175.4	214.4	257.1	286.6
ENE-3	0.75:0.25	169.2	181.5	216.7	268.3	150.2
ENE	1:0	156.7	182.2	198.9	260.0	137.8
ENM-1	0.25:0.75	142.2	176.6	217.8	297.3	104.4
ENM-2	0.5:0.5	141.8	191.8	216.3	284.0	283.6
ENM-3	0.75:0.25	167.1	201.9	224.4	265.8	211.2
ENM	1:0	140.1	155.2	203.0	320.5	112.4

Table 2 Results of DSC scan of DGEBA in the presence of imide-amines OS/OE/OM, DDS and its mixture in varying molar ratios at 10°C min^{-1}

Sample designation	Molar ratios of NS/NE/NM:DDS	T_i/°C	T_{onset}/°C	T_p/°C	T_f/°C	ΔH/J g^{-1}
EOS-1	0.25:0.75	139.3	187.3	214.2	294.4	332.8
EOS-2	0.5:0.5	184.5	195.1	224.4	308.5	165.9
EOS-3	0.75:0.25	181.6	184.6	212.2	278.3	105.9
EOS	1:0	185.8	196.4	223.1	308.5	177.1
EOE-1	0.25:0.75	158.8	176.1	196.7	253.8	226.2
EOE-2	0.5:0.5	163.4	177.6	189.0	243.8	131.3
EOE-3	0.75:0.25	136.4	159.6	194.2	248.4	179.7
EOE	1:0	150.9	176.4	179.2	265.8	181.5
EOM-1	0.25:0.75	163.4	172.1	195.8	264.1	85.2
EOM-2	0.5:0.5	159.2	169.2	189.9	260.4	63.2
EOM-3	0.75:0.25	156.7	173.1	179.6	248.4	111.1
EOM	1:0	149.3	154.2	183.9	260.0	115.3

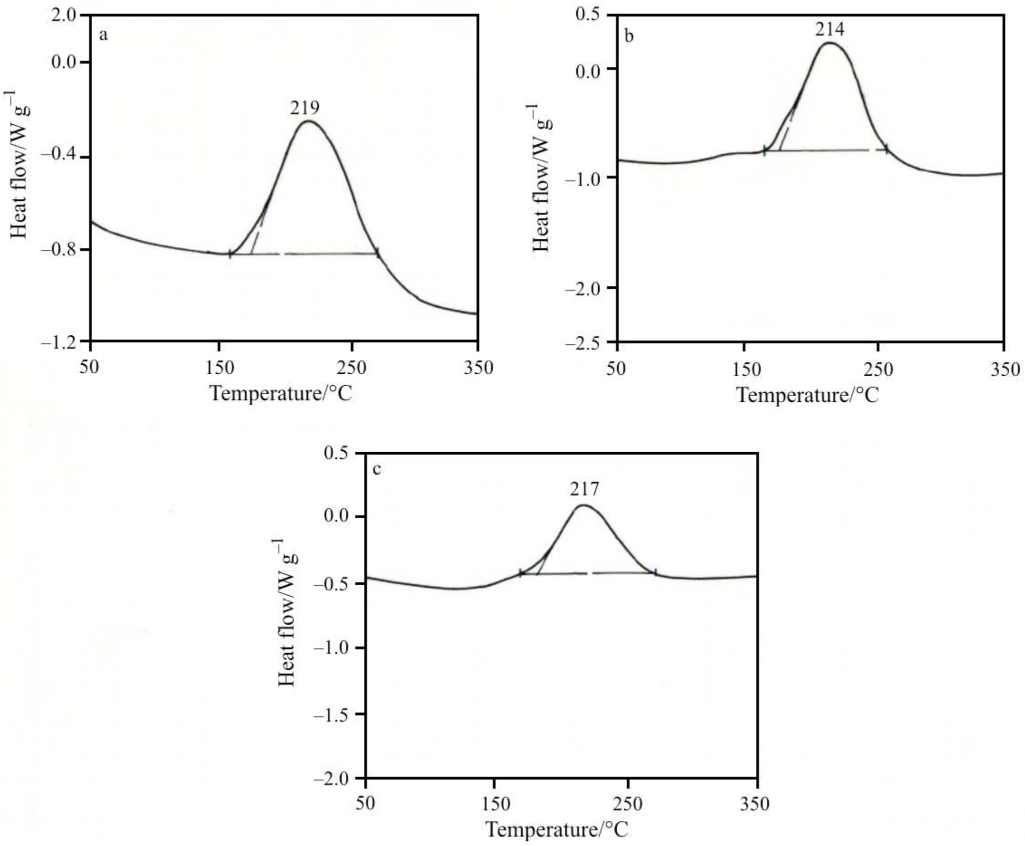

Fig. 3 DSC scans of DGEBA cured using mixture of NE and DDS a – ENE-1, b – ENE-2 and c – ENE-3 at 10°C min^{-1}

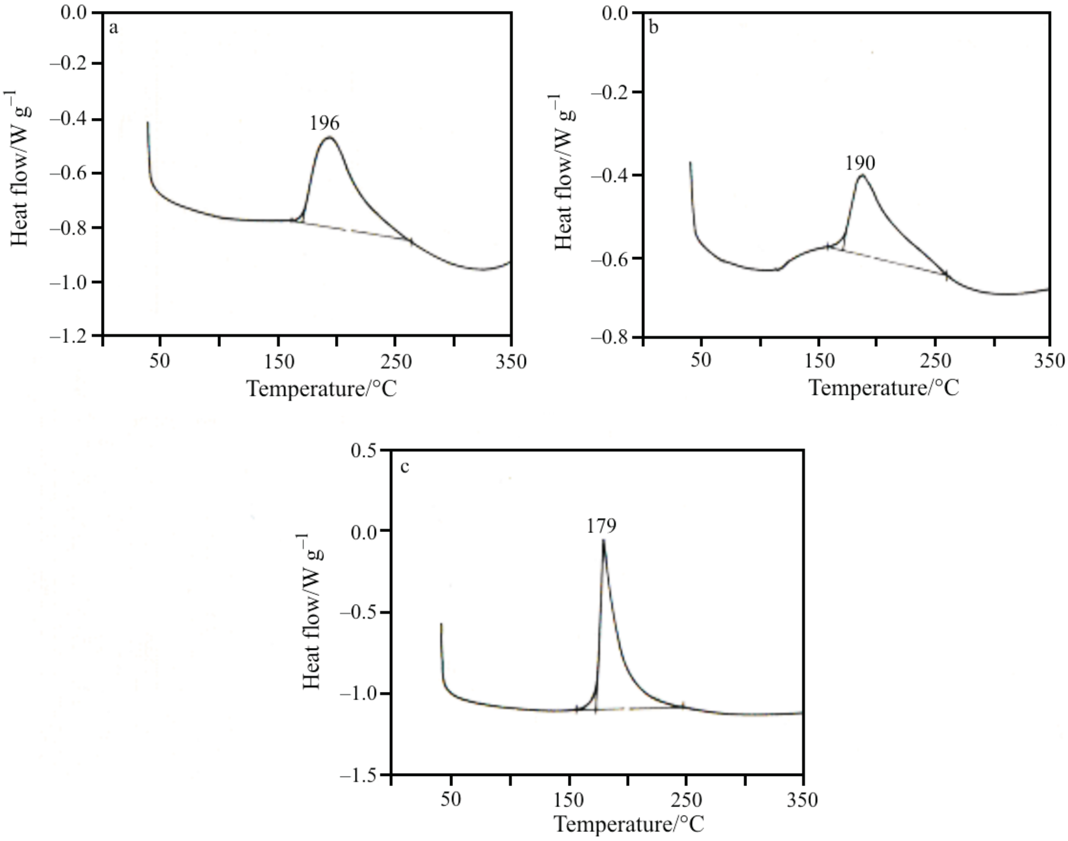

Fig. 4 DSC scans of DGEBA cured using mixture of OM and DDS a – EOM-1, b – EOM-2 and c – EOM-3 at 10°C min^{-1}

showed decrease in peak exotherm temperature. All the temperatures are lower than either of the curing agent used. ΔH values showed no definite trend.

In the DSC scans of samples cured isothermally (by heating at 180±20°C for DDS: NS/NE/NM/OS/OM/OE in an air oven for 3 h), absence of exothermic transition indicated the completion of curing. However, DSC was not sensitive enough to determine the glass transition temperature as no shift in the baseline was observed.

Thermal stability

TG/DTG traces of DGEBA cured isothermally using imide-amines in nitrogen atmosphere ENE, EOS, ENM at heating rate 20°C min^{-1} is shown in Fig. 5. TG traces were obtained by plotting percent residual mass *vs*. temperature. Percent char yield at 800°C and following characteristic temperatures were noted from TG/DTG traces:

- IDT=Initial decomposition temperature, is the temperature at which the sample starts decomposing. It was determined by extrapolation using the steep portion of mass loss curve and the initial baseline.
- T_{max}=It is the temperature at which the rate of mass loss is maximum. It was obtained from DTG traces.
- FDT=Final decomposition temperature, is the temperature at which the mass loss was complete. It was also obtained by extrapolation.
- Char yield=It is the residual mass left after recording the TG trace upto 800°C.

The results of TG/DTG scans are summarized in Tables 3 and 4. From the char yield, limiting oxygen index (LOI) was calculated according to Van Krevelen and Hoftyzer method [20] (Eq. (1)).

$$\text{LOI}=17.5+0.4\text{CR} \tag{1}$$

where CR=char yield.

The char residue from pyrolysis is linearly proportional to the oxygen index of halogen free polymers.

In case of NTDA and ODPA based imide-amines, a single step decomposition was observed in all the samples except in EOS. All the samples were stable upto 350°C and a significant mass loss occurred only beyond this temperature. The char yield was found to be dependent on the structure of imide-amines and it was highest for ENE and lowest for EOS. Lower char yield observed in case of EOS, ENS could be due to the loss of SO_2, which is present in the

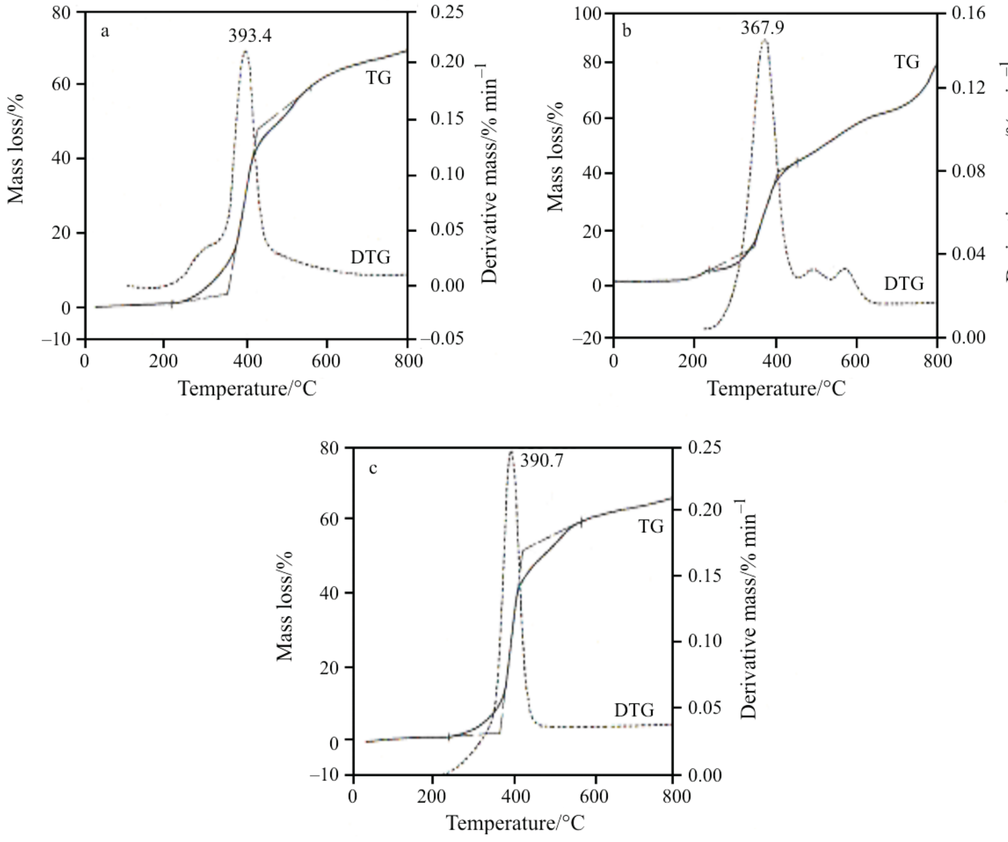

Fig. 5 TG/DTG traces of DGEBA cured isothermally using imide-amines in nitrogen atmosphere a – ENE, b – EOS and c – ENM (heating rate 20°C min^{-1})

Table 3 Results of thermal stability of DGEBA cured isothermally using NS/NE/NM, DDS or mixture in nitrogen atmosphere (heating rate=20°C min^{-1})

Sample designation	IDT/°C	T_{max}/°C	FDT/°C	Char yield/% at 800°C	LOI/%
ED	394.9	419.9	466.0	19.2	25.2
ENS-1	432.0	456.2	484.7	30.2	29.6
ENS-2	416.1	438.4	458.1	28.3	28.8
ENS-3	402.1	432.3	451.4	28.2	28.8
ENS	353.0	386.7	414.4	28.3	28.8
ENE-1	447.1	462.7	487.9	28.2	28.8
ENE-2	402.3	431.5	466.2	30.6	29.7
ENE-3	395.2	428.9	460.6	31.8	30.2
ENE	353.2	393.4	424.8	35.2	31.6
ENM-1	402.4	436.7	462.3	25.3	27.6
ENM-2	394.8	440.5	468.6	26.2	28.0
ENM-3	406.2	441.1	469.2	27.4	28.5
ENM	362.4	390.7	417.5	34.5	31.3

Table 4 Results of thermal stability of DGEBA cured isothermally using OS/OE/OM, DDS or mixture in nitrogen atmosphere (heating rate=20°C min^{-1})

Sample designation	IDT/°C	T_{max}/°C	FDT/°C	Char yield/% at 800°C	LOI/%
EOS-1	409.0	428.2	450.8	25.5	27.7
EOS-2	393.8	425.2	453.8	23.5	28.9
EOS-3	398.0	420.6	438.4	28.4	28.9
EOS	340.2	367.9	400.5	20.5	25.6
EOE-1	410.5	443.8	464.2	25.7	27.8
EOE-2	395.3	425.7	461.6	28.3	28.8
EOE-3	398.2	441.3	458.5	29.2	29.2
EOE	369.9	400.4	427.5	32.1	30.3
EOM-1	404.8	429.6	459.2	26.5	28.1
EOM-2	399.7	435.2	462.5	25.7	27.8
EOM-3	401.2	434.3	459.2	23.2	26.8
EOM	382.7	402.3	426.1	30.4	29.7

imide-amines. Imide-amines based on NTDA gave resins with higher char yield as compared to those based on ODPA. This can be explained on the basis of compact network structure in the presence of naphthalene-based imide-amines. Increased char formation can limit production of combustible carbo-containing gases, decrease the exothermicity due to pyrolysis reactions, as well as decrease the thermal conductivity of the surface of burning materials [21]. The solid-state aromatisation reactions in these imide-amines may be facilitated by the formation of additional cross-links through the reaction of carbonyl and trapped/unreacted amino groups at elevated temperatures. All the samples had LOI values calculated based on their char yield was higher than 28 except for EOS. On the basis of LOI values, such materials can be classified as self-extinguishing resin and flame resistant DGEBA resin can be obtained by using imide-amines as curing agents.

The samples cured using mixture of NS/NE/NM imide-amines: DDS were stable upto 410±25°C. IDT, T_{max}, FDT and char yield increased significantly when 0.25 mole of DDS was added to the mixture of NE/NM. Incorporation of 0.25 mole of DDS in mixture of NS and DDS increased the char yield, IDT, T_{max} and FDT.

The samples cured using mixture of OS/OE/OM imide-amines: DDS were stable upto 400±10°C. The relative thermal stability values of the cured resins were higher of either of the samples.

Integral procedural decomposition temperature (IPDT)

The integral procedural decomposition temperature which sums the shape of the primary thermogravimetric traces (TG) was calculated according to the method of Doyle [22]. IPDT provides the most valid comparison of diverse polymeric materials, when thermogravimetric traces of all polymer samples were obtained under identical conditions.

From the TG traces, the area under the curve was obtained by counting the squares under the curve. The ratio of the area under the curve and the area of rectangular plot bounded by the curve gave the value of A^*. A^* was correlated to IPDT by the following relation:

$$\text{IPDT}=A^*(T_f-T_i)+T_i \qquad (2)$$

where T_f=final decomposition temperature, T_i=initial decomposition temperature.

Activation energy of decomposition (E)

Activation energy of decomposition (E) can also be used as a criteria for comparing the thermal stability of polymers. The rate constant of a reaction depends upon energy difference between the transition and initial states. This shows that if the energy difference is higher (i.e. activation energy is higher) the compound will be more stable.

Several expressions for the determination of activation energy from the TG data have been reported in [23–26]. Dharwadkar and Kharkhanawala [27] have given a modified equation for the calculation of activation energy, which is independent of sample size and heating rate.

$$\ln[\ln(1-\alpha)^{-1}] = \frac{100E\theta}{RT_i^2(T_f - T_i)} + C \qquad (3)$$

where α=fraction reacted, T_i=temperature of inception of reaction, T_f=temperature at the point of inflection on the curve, $\theta=(T\text{-}T_s)$ (where T is the temperature under consideration and T_s is the maximum temperature), C=constant and R=gas constant.

A plot of $\ln[\ln(1-\alpha)^{-1}]$ *vs*. θ gave a straight line. The slope of the line was equal to

$$m = \frac{100E}{RT_i^2(T_f - T_i)} \qquad (4)$$

From this equation E was calculated.

The IPDT values are summarized in Tables 5 and 6. Comparing imide-amines based on ether/methylene/sulfone amine and different anhydrides, the trend in IPDT was

ENE>EOE

ENM>EOM

EOS>ENS

This clearly shows that trend of thermal stability changes with the nature of anhydride/amine. For example with ether containing amine, IPDT value was highest with NTDA based anhydride. The trend with methylene or sulfone containing amines was different.

Table 5 Thermal behaviour of cured epoxy resin using mixture of NS/NE/NM:DDS in nitrogen atmosphere

Sample designation	Decomposition temperature/°C at various %age mass losses				IPDT/°C
	10	20	30	40	
ENS-1	429	444	454	466	634.2
ENS-2	396	416	441	453	612.4
ENS-3	384	417	433	446	598.3
ENS	348	372	390	400	601.1
ENE-1	441	453	465	473	678.1
ENE-2	399	419	426	445	669.8
ENE-3	393	406	421	436	618.6
ENE	332	355	386	400	702.1
ENM-1	400	424	436	449	669.8
ENM-2	393	403	441	456	664.4
ENM-3	404	413	425	436	662.4
ENM	350	386	400	412	765.8

Table 6 Thermal behaviour of cured epoxy resin using mixture of OS/OE/OM:DDS in nitrogen atmosphere

Sample designation	Decomposition temperature/°C at various %age mass losses				IPDT/°C
	10	20	30	40	
EOS-1	400	415	424	436	653.8
EOS-2	389	403	418	432	690.7
EOS-3	394	402	418	428	694.3
EOS	320	360	387	430	641.6
EOE-1	404	420	432	444	668.2
EOE-2	392	419	430	452	612.5
EOE-3	390	407	425	436	610.9
EOE	360	399	409	431	573.9
EOM-1	400	420	443	451	614.8
EOM-2	394	419	429	441	600.7
EOM-3	398	410	424	445	598.4
EOM	376	400	410	423	600.8

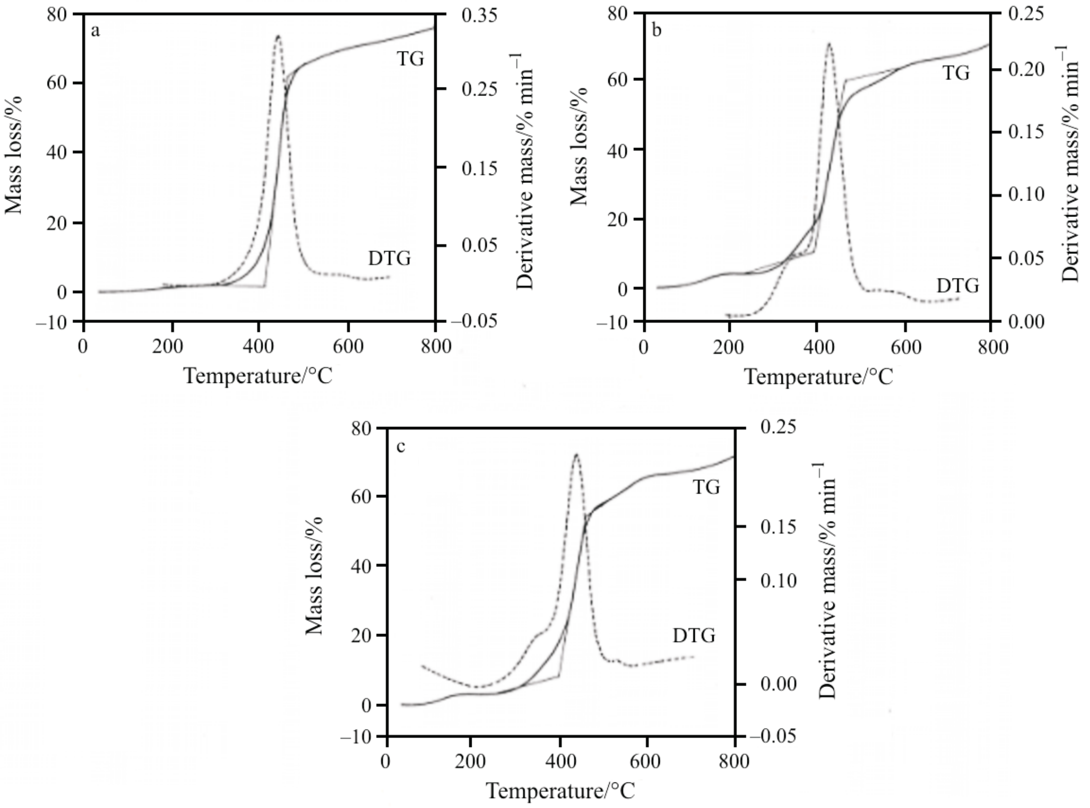

Fig. 6 TG/DTG traces of isothermallly cured epoxy resins using mixture of NS and DDS a – ENS-1, b – ENS-2 and c – ENS-3 at 20°C min^{-1}

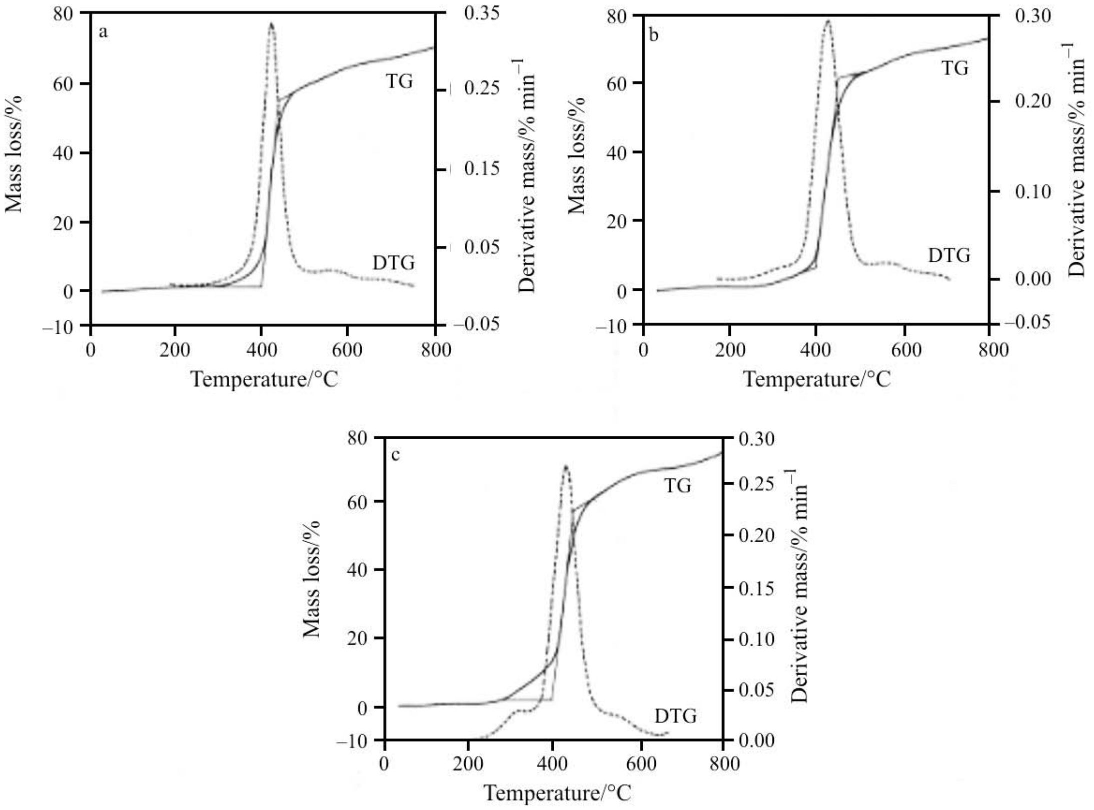

Fig. 7 TG/DTG traces of isothermallly cured epoxy resins using mixture of OS and DDS a – EOS-1, b – EOS-2 and c – EOS-3 at 20°C min^{-1}

Table 7 Activation energy values for the thermal decomposition of cured epoxy resin using mixture of NS/NE/NM:DDS

Sample designation	Temperature range/°C	E/kJ mol^{-1}
ENS-1	318–562	335.7
ENS-2	320–566	346.3
ENS-3	299–543	356.9
ENS	260–486	148.7
ENE-1	335–585	383.8
ENE-2	300–550	363.4
ENE-3	295–542	355.6
ENE	221–550	234.2
ENM-1	302–546	394.7
ENM-2	290–544	386.7
ENM-3	304–562	375.9
ENM	230–584	301.8

Table 8 Activation energy values for the thermal decomposition of cured epoxy resin using mixture of OS/OE/OM:DDS

Sample designation	Temperature range/°C	E/kJ mol^{-1}
EOS-1	299–550	358.7
EOS-2	297–552	364.8
EOS-3	302–555	369.7
EOS	230–451	174.4
EOE-1	319–563	343.6
EOE-2	301–547	338.3
EOE-3	304–555	306.4
EOE	226–570	247.5
EOM-1	309–554	289.8
EOM-2	303–549	298.4
EOM-3	308–560	310.3
EOM	250–552	223.6

The activation energy of decomposition (E) was calculated according to Eqs (3) and (4) and the values are given in Tables 7 and 8. On comparing the activation energy in a series of imide-amines based on DDM/DDE/DDS keeping anhydride constant the following trend in activation energy was seen.

ENM>ENE>ENS

EOE>EOM>EOS

In case of NTDA based imide-amines, activation energy was highest for DDM (ENM) based amines and lowest with sulfone containing amines (ENS).

IPDT values decreased significantly on adding increasing amount of imide-amines NS/NE to the mixture. There was not much change in IPDT values on adding NM to the mixture. The activation energy was found to be increased with NS and DDS mixture and decreased with NE/NM and DDS mixture.

IPDT and activation energy values increased on adding more moles of OS to the mixture whereas it decreased with OE imide-amine. In case of sample cured using mixture of OM and DDS, IPDT values decreased and activation energy increased.

Conclusions

From these results, it can be concluded that the crosslinking of epoxy resin by aromatic imide-amines containing aryl ether, sulfone and methylene linkages was influenced by the structure of imide-amines. The peak temperature of exothermic transition depends on the nucleophilicity of imide-amines and it was found to be highest for ENS and EOS due to the presence of electron withdrawing group (sulfone group). Higher char yields were obtained with epoxy resins cured with imide-amines based on NTDA as compared to ODPA based imide-amines. The highest char yield was observed in case of ENE and lowest with EOS. Thermal stability of the cured material was found to be dependent on the structure of network. Composition of the mixture had a large effect on the thermal behaviour of epoxy resin.

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