J Mater Sci (2007) 42:8342–8349
DOI 10.1007/s10853-006-0547-0

Epoxy-imide resins from *N*-(4-carboxyphenyl)trimellitimide: effect of imide and aromatic content on adhesive and thermal properties

Ginu Abraham · Shanmugam Packirisamy · Sugu Surya Bhagawan · Ganapathy Balasubramanian · Rajagopala Iyer Ramaswamy

Received: 9 August 2005 / Accepted: 8 June 2006 / Published online: 23 February 2007

Abstract The effect of imide and aromatic content on the adhesive and thermal properties of epoxy-imide resins obtained through the reaction of epoxy resins viz., Araldite GY 250 (DGEBA; difunctional), Araldite EPN 1138 (novolac; polyfunctional) and epoxidized hydroxyl-terminated polybutadiene (EHTPB) with an imide-diacid viz., *N*-(4-carboxyphenyl)trimellitimide (IDA-I) was studied for different carboxyl equivalent to epoxy equivalent ratios (C/E ratios). The glass transition temperature of epoxy-imides varies from 80 to 144 °C, 120 to 184 °C and 41 to 143 °C for GY 250-IDA-I, EPN 1138-IDA-I and EHTPB-IDA-I respectively with the variation in C/E ratio from 0.5 to 2. For C/E ratio up to 1, the overall thermal stability of epoxy-imides follow the trend, EPN 1138-IDA-I > GY 250-IDA-I > EHTPB-IDA-I and above this ratio considerable improvement in overall thermal stability is observed only for EHTPB-IDA-I. The optimum C/E ratio for obtaining maximum room temperature and elevated temperature adhesive strength was found to be 1.25. For the optimum ratio, GY 250, EPN 1138 and EHTPB-based systems give adhesive strength of 29, 23 and 18 MPa respectively at room temperature and retain 78.3, 56.2 and 44.4% of the room temperature adhesive strength at 150 °C. The influence of C/E ratio or in otherwords, imide or aromatic content on the adhesive strength, thermal stability and glass transition temperature was more pronounced for aliphatic epoxy, i.e., EHTPB-based system.

Introduction

Aromatic polyimides are well accepted as high performance polymeric materials due to their excellent mechanical strength and thermal stability. They are widely used as films, coatings and adhesives on a wide variety of substrates ranging from metals to glasses [1–5]. However, they pose processing problems due to their insolubility and infusibility. For most applications, an easily processable polyamic acid precursor is used and thermal or chemical imidization is carried out to obtain polyimides. Volatiles are evolved during the imidization process, which limit the end-uses of polyimides as films and coatings. In order to overcome the above processing problems, research work on polyimides has been focused in the following directions: (i) synthesis of copolyimides [6–8] which are soluble in the fully imidized state, (ii) synthesis of processable

G. Abraham · S. Packirisamy · Rajagopala Iyer Ramaswamy
Polymers and Special Chemicals Division, Propellant and Special Chemicals Group, PCM Entity, Vikram Sarabhai Space Centre, Thiruvananthapuram 695022, India

Present Address:
G. Abraham (✉)
The Centre for Advanced Macromolecular Design (CAMD), School of Chemical Sciences and Engineering, Kensington, Sydney, NSW 2052, Australia
e-mail: abraham.ginu@gmail.com

Sugu Surya Bhagawan · G. Balasubramanian
Propellant Engineering Division, Propellant and Special Chemicals Group, PCM Entity, Vikram Sarabhai Space Centre, Thiruvananthapuram 695022, India

Present Address:
Sugu Surya Bhagawan
Department of Polymer Engineering, Amritha School of Engineering, Ettimadai, Coimbatore 641105, India

addition-type imides [2, 9, 10] through the reaction of aromatic diamines with maleic, itaconic, citraconic and nadic anhydrides and (iii) synthesis of soluble polyimides using diamines and dianhydrides containing flexible linkages [11, 12].

Of late, researchers have focused their attention towards developing epoxy-imide resins, which are copolyimides, obtained by curing epoxy resins with imide group containing curatives [13–23]. The advantage of this approach is that the versatility of epoxy resins such as low shrinkage, chemical resistance and ease of fabrication, and the rigidity and high temperature stability of imide groups are combined in a single system. As no volatile is evolved during curing of epoxy resins with imide-group containing curatives, epoxy-imide resins can be conveniently used as adhesives and matrix resins for composites.

In our laboratory, we have synthesized different epoxy-imide resins by curing difunctional and polyfunctional epoxy resins with imide group containing dicarboxylic acids and evaluated their adhesive strength on stainless steel substrate [16–23]. For epoxy-imide resins obtained from diimide-diacids, it is observed that the adhesive strength, retention of room temperature adhesive strength at elevated temperature and thermal properties are influenced by the carboxyl eqv to epoxy eqv ratio (C/E ratio) [17]. Recently, we have reported the adhesive and thermal properties of epoxy-imide resins [19, 23] obtained by curing epoxy resins such as Araldite GY 250 (difunctional; diglycidyl ether of bisphenol-A (DGEBA)), Araldite EPN 1138 (polyfunctional; novolac epoxy) and epoxidized hydroxyl-terminated polybutadiene (EHTPB; polyfunctional) with *N*-(4-carboxyphenyl)trimellitimide in 1:1 C/E ratio. In the present paper, we report the effect of variation of C/E ratio on the adhesive and thermal properties of the above systems. Variation of C/E ratio in effect causes variation of imide and aromatic content of epoxy-imide resins. The C/E ratio has been varied from 0.25:1 to 2:1 and for each ratio the adhesive strength at room temperature and at 100, 125 and 150 °C, glass transition temperature and thermal stability have been evaluated to understand the effect of variation of imide and aromatic content.

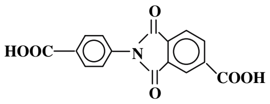
Experimental

Materials

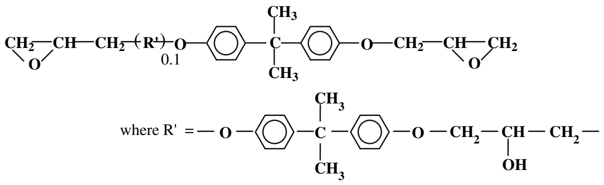
Epoxy resins, Araldite GY 250 (DGEBA, epoxy value: 5.0–5.5 eqv/kg) and Araldite EPN 1138 (Novolac-epoxy, epoxy value: 5.5–5.7 eqv/kg) manufactured and supplied by M/s Hindustan Ciba-Geigy, India Ltd., Mumbai, India were used. EHTPB having epoxy value of 5.2 eqv/kg was synthesized from hydroxyl-terminated polybutadiene (HTPB) [23, 24]. *N*-(4-carboxyphenyl)trimellitimide (IDA-I) was synthesized by the procedure reported earlier [19]. Structures of epoxy resins and IDA-I used in the present study are shown in Fig. 1.

Surface preparation of stainless steel coupons

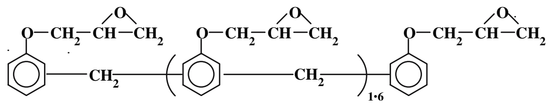
AISI-304 cold roll stainless steel coupons of dimensions 100 mm × 25 mm × 1.6 mm were used. Stainless steel coupons were first degreased by wiping with *n*-hexane. The area to be bonded was abraded with emery paper No. 36 and cleaned with detergent and tap water. It was then chemically etched using chromic acid solution (36 g of saturated solution of sodium dichromate in 100 g of conc. H_2SO_4) for 60 min at 60 °C as per ASTM D 2651. Coupons were then thoroughly washed with tap water to remove all traces of acid. The effectiveness of the surface was checked by waterbreak-free test, in which properly cleaned surface would hold a continuous film of water rather than a series of isolated droplets. The coupons were dried in an air oven at 110 °C for 1 h, cooled to room temperature and then used for bonding. The whole process of cleaning, etching and drying was carried out



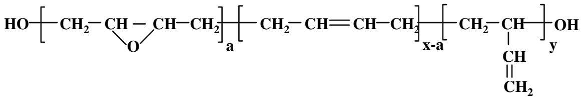
Imide-diacid (IDA-I)



Araldite GY 250



Araldite EPN 1138



Epoxidized hydroxyl terminated polybutadiene (EHTPB)

Fig. 1 Structures of imide-diacid (IDA-I) and epoxy resins just prior to adhesive application in order to ensure the availability of freshly prepared metallic oxide surface for bonding.

Curing of epoxy resins with imide-diacid and adhesive formulation

Imide-diacid (IDA-I) and epoxy resin (Araldite GY 250, Araldite EPN 1138 or EHTPB) in 1:1 C/E ratio were mixed thoroughly in a pestle and mortar. The cure reaction was followed by DSC. From the DSC data, the initial, maximum and final cure temperatures (T_{ic}, T_{maxc} and T_{fc} respectively) were determined (Table 1). The epoxy resin-imide-diacid mixture was applied on cleaned stainless steel substrates and the steel coupons were then bonded to get an approximate bonded area of 3 cm^2. The bonded specimens were stacked together and kept in an air oven under a contact pressure of 3.3 MPa. The temperature of the oven was raised to T_{ic} over a period of 30 min and maintained at this temperature for 30 min. The temperature of the oven was further raised to T_{maxc} over a period of 15 min and maintained at this temperature for 1 h and finally the oven temperature was raised to T_{fc} over a period of 15 min and the specimens were kept at this temperature for 30 min. The oven was allowed to cool to room temperature before the specimens were removed. The above cure schedule was used for preparing epoxy-imide resins using C/E ratios lower or higher than 1 as very little variation in T_{ic}, T_{maxc} and T_{fc} values was observed with the change in C/E ratios.

Measurements

IR spectra of the samples were recorded on a Nicolet 510 P FTIR in KBr pellets or as a smear on NaCl plate. DSC studies were made with a Mettler DSC TA 3000 at a heating rate of 10 °C/min. Glass transition temperature (T_g) of epoxy-imides was determined by DSC (TA; Q 100) at a heating rate of 10 °C/min. TG curves were recorded with a Dupont 900 thermal analyzer in conjunction with 951 thermogravimetric analyzer at a heating rate of 10 °C/min in nitrogen atmosphere. Tensile lap shear strength of bonded SS specimens was measured using Instron (Model No: 4469) at a crosshead speed of 10 mm/min as per ASTM D-1002. Tensile lap shear strength at 100, 125 and 150 °C was measured after equilibrating the test specimens at the required temperature for a period of 10 min in a temperature cabinet (Model No. EC 100) attached to Instron machine. To obtain lap shear strength value at a particular temperature, five bonded coupons were tested. The lap shear strength value reported is an average of three measurements which fell in the range of ±1 MPa.

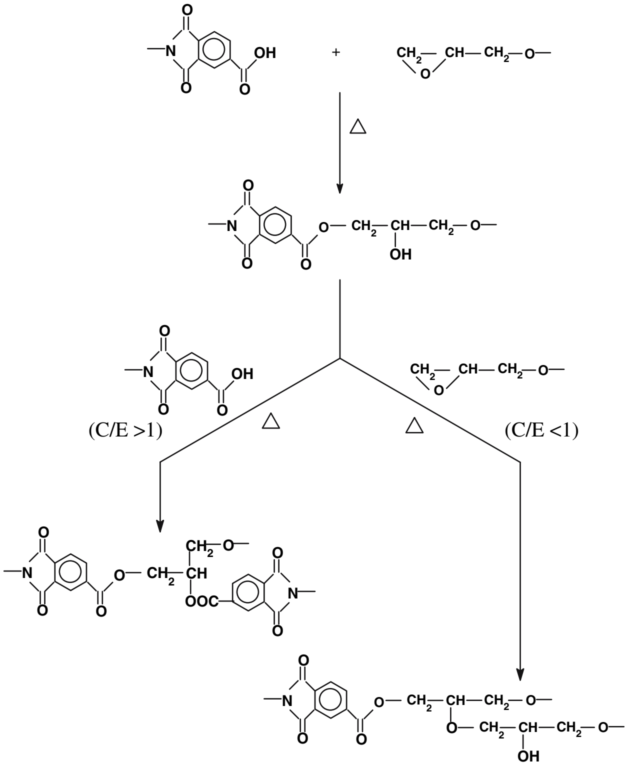
Table 1 Curing of epoxy resins with IDA-I: DSC data

System	T_{ic} (°C)	T_{maxc} (°C)	T_{fc} (°C)
GY 250-IDA-I	150	200	230
EPN 1138-IDA-I	170	230	270
EHTPB-IDA-I	180	220	250

Results and discussion

Curing of epoxy resins with imide-diacid

The curing of epoxy resins with imide-diacid proceeds through carboxyl-epoxy addition (Scheme 1) reaction at elevated temperatures. The structures of epoxy-imide resins thus obtained for Araldite GY 250-IDA-I, Araldite EPN 1138-IDA-I and EHTPB-IDA-I for 1:1 C/E ratio are given in Fig. 2. The curing of GY 250 with IDA-I (for C/E ratio = 1) is expected to result in the formation of a linear polymer whereas in the case of EPN 1138-IDA-I and EHTPB-IDA-I, crosslinked epoxy-imide resins are formed due to the polyfunctional nature of Araldite EPN 1138/EHTPB. This basic



Scheme 1 Epoxy-carboxyl addition reaction

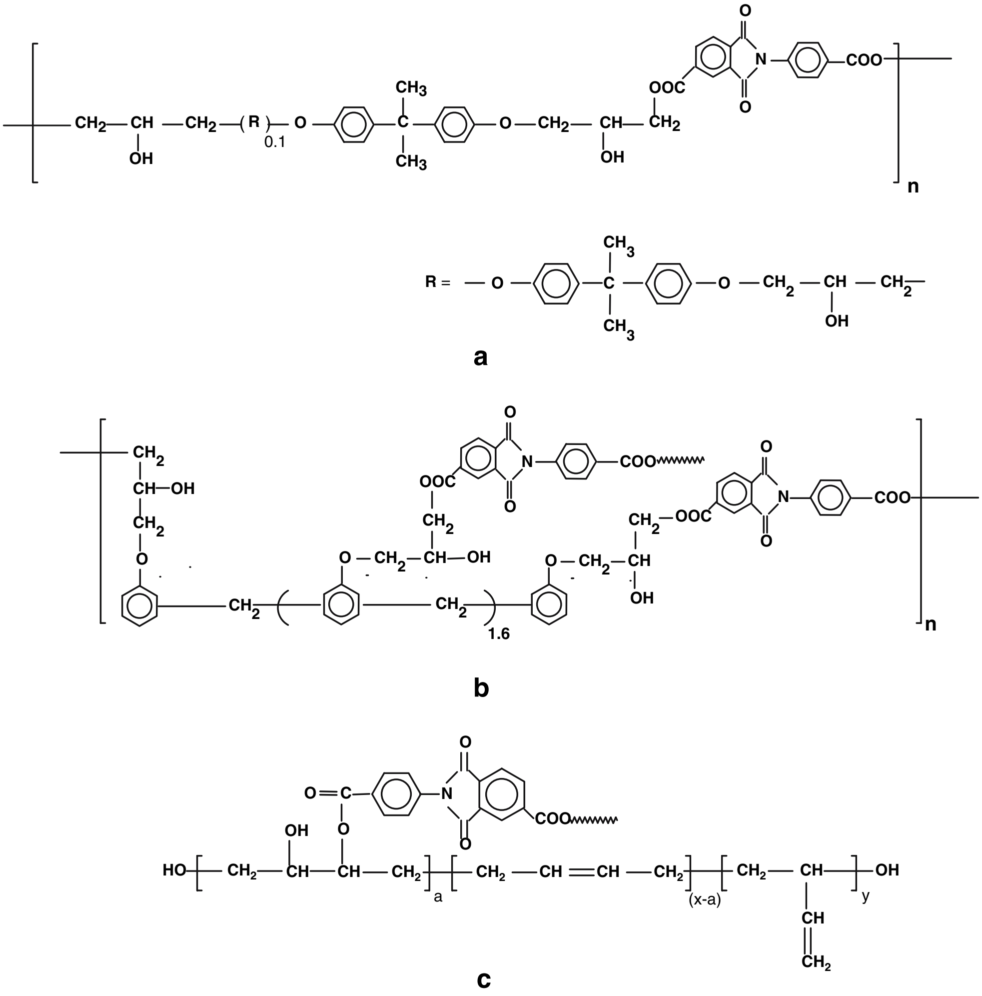


Fig. 2 Structures of different epoxy-imides (**a**) GY 250-IDA-I, (**b**) EPN 1138-IDA-I and (**c**) EHTPB-IDA-I.

difference in the epoxy-imide resins derived from Araldite GY 250 and Araldite EPN 1138 resins is found to influence the thermal as well as adhesive properties.

The cure reaction of epoxy resins, Araldite GY 250, Araldite EPN 1138 and EHTPB, with IDA-I was followed by DSC and IR spectra. As outlined in the experimental section, the cure schedule for each system was fixed based on the DSC data. The absence of the peak at 915 cm^{-1}, corresponding to epoxy group, in the IR spectra of the cured resins confirmed the completion of curing. The IR spectra of cured resins show absorptions at 1,780, 1,719, 1,385 and 725 cm^{-1} arising due to imide group. The broad band around 3,000 cm^{-1} is due to –OH group obtained as a result of esterification reaction of epoxy and –COOH group. The characteristic ester peak has probably merged with the imide absorption at 1,719 cm^{-1}.

When C/E ratio is 1, it is likely that no free acid or epoxy group is left out in the cured resin. However, when the ratio is less than unity, unreacted epoxy groups are expected to be present in the cured resins. The peak at 915 cm^{-1} corresponding to epoxy group is absent in the IR spectra of epoxy-imide resins prepared by using C/E ratio less than 1, suggesting that there is no unreacted epoxy group. It is reported [25] that dangling hydroxyl groups formed by the carboxyl-epoxy addition reaction react with the excess epoxy groups and similar reaction is possible in the present systems (Scheme 1) when C/E ratio is less than 1. If carboxyl group is in excess (for C/E ratios higher than 1) it can undergo esterification with the dangling hydroxyl group present in the growing polymer chain [26, 27]. No meaningful information could be obtained from the IR spectra of epoxy-imide resins prepared using C/E ratio higher than 1 as both –OH peaks corresponding to carboxylic group and hydroxyl group of the growing polymer chain appear in the same region (3,000–3,500 cm^{-1}) in the IR spectra. The dangling hydroxyl-epoxy reaction and the esterification reaction as shown in Scheme 1 would contribute to additional crosslinking. Thus, for one epoxy equivalent, a maximum of two carboxyl equivalent can be used to make a highly crosslinked polymer. In the case of EHTPB-based systems, in addition to the esterification of dangling hydroxyl groups, esterification of end-functional –OH groups of EHTPB with imide-diacid may take place when the C/E ratio is above 1.

Thermal properties of epoxy-imide resins

The glass transition temperature, initial decomposition temperature (T_i), maximum decomposition temperature (T_{max}), final decomposition temperature (T_f) and char residue at 900 °C of epoxy-imide resins for C/E ratios 0.5, 1.0, 1.5 and 2.0 are summarized in Table 2. TG curves of EHTPB-IDA-I system, where remarkable improvement in thermal stability is observed with the increase in C/E ratio, are compared in Fig. 3.

From Table 2 it is observed that for all the systems there is considerable increase in T_g (50–60 °C) when C/E ratio is increased from 0.5 to 1. This is attributed to the increase in imide and aromatic content of epoxy-imide systems when C/E ratio is increased from 0.5 to 1. For GY250-IDA-I and EPN 1138-IDA-I systems there is only a marginal increase (4–10 °C) when the C/E ratio is increased from 1 to 2. On the other hand, for EHTPB-IDA-I system, T_g increases from 93 °C to 112 °C with the increase in C/E ratio from 1.0 to 1.5 and with further increase in the ratio to 2, T_g increases to 143 °C. Unlike Araldite GY 250 and Araldite EPN 1138, EHTPB is an aliphatic epoxy resin and hence, increase in aromatic content and imide content by way of increasing the C/E ratio influences EHTPB-based system when compared to the other two systems.

For C/E ratio of 0.5, the thermal stability of epoxy-imide resins follow the trend,

EPN1138-IDA-I > GY 250-IDA-I > EHTPB-IDA-I

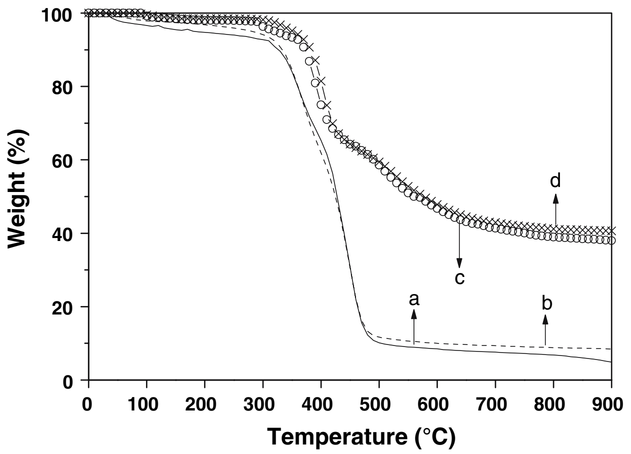


Fig. 3 Thermograms (in nitrogen atmosphere) of EHTPB-IDA-I system obtained using different C/E ratio: (**a**) 1:0.5, (**b**) 1:1.0, (**c**) 1:1.5 and (**d**) 1:2

Table 2 Comparison of glass transition temperature and thermal stability of epoxy-imides for different carboxyl eqv to epoxy eqv ratios

System	Composition (C/E ratio)	T_g (°C)	T_i (°C)	T_{max} (°C)	T_f (°C)	Char residue at 900 °C(%)
GY 250-IDA-I	0.5	80	380	424	570	24.0
	1.0	139	370	420	580	26.0
	1.5	140	360	420	570	31.4
	2.0	144	360	405	620	33.2
EPN 1138-IDA-I	0.5	120	355	398	615	41.2
	1.0	170	380	420	550	42.0
	1.5	175	375	410	620	39.0
	2.0	184	365	410	630	35.0
EHTPB-IDA-I	0.5	41	320	405	470	7.0
	1.0	93	330	404	470	9.0
	1.5	112	370	450	620	38.0
	2.0	143	370	450	665	41.0

The higher thermal stability of EPN 1138-based system when compared to other two systems is attributed to high aromatic content and polyfunctional nature of EPN 1138. Though EHTPB is a polyfunctional resin, the epoxy-imide resin derived from it has the least thermal stability and this is attributed to the high aliphatic content. With the variation in C/E ratio only a marginal variation in the overall thermal stability is observed for GY 250-IDA-I and EPN 1138-IDA-I. However, there is a remarkable improvement in thermal stability of EHTPB-IDA-I particularly when the C/E ratio is increased from 1 to 1.5. When the C/E ratio is above 1, in addition to epoxy-carboxy addition reaction, esterification of end-functional –OH groups of EHTPB with –COOH of IDA-I can take place resulting in additional crosslinking contributing to the improvement in thermal stability.

Adhesive properties of epoxy-imide resins

Adhesive lap shear strength values at room temperature, 100 °C, 125 °C and 150 °C for different C/E ratios for EPN 1138-IDA-I system are compared in Fig. 4 and those for GY 250-IDA-I and EHTPB-IDA-I are given in Table 3. For all the three systems maximum room temperature adhesive strength is obtained when the C/E ratio is 1.25 and thereafter a decrease in adhesive strength is observed with the increase in C/E ratio. It is noticed that the adhesive lap shear strength values at 100 °C, 125 °C and 150 °C are influenced by both C/E ratio and the nature of epoxy resin. As discussed earlier, the increase in C/E value from 0.5 to 1 increases the T_g of epoxy-imides by 50–60 °C

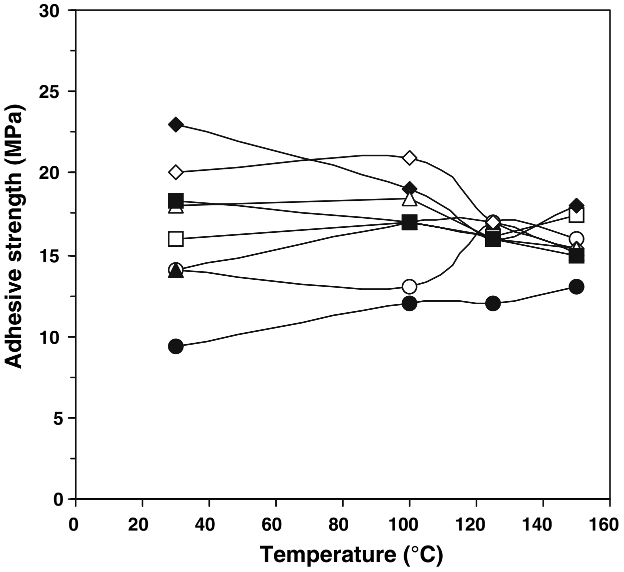


Fig. 4 Effect of C/E ratio on the adhesive strength at different temperatures of EPN 1138-IDA-I system: (●) 0.25, (○) 0.5, (▲) 0.75, (□) 1.0 (◆) 1.25, (◇) 1.5, (△) 1.75 and (■) 2.0

(Table 2). This increase in T_g with the increase in C/E ratio is responsible for the improvement in retention of adhesive strength at elevated temperature of epoxy-imides. For EPN 1138-IDA-I system 100% or more than 100% retention of the room temperature adhesive strength is observed only up to 1:1 ratio. EPN 1138-based system, being a highly crosslinked one, the esterification reaction between the unreacted or excess acid groups with the dangling hydroxyl group (Scheme 1) might not have been taken to completion and hence, unreacted acid groups may be present which in turn act as a weak layer in the glue-line and reduces the adhesive strength at elevated temperature.

The effect of nature of epoxy resin can be understood by comparing the percentage retention of adhesive strength for a chosen C/E value and the influence is expected to be more for lower C/E ratios than for higher C/E ratios where the influence of imide and aromatic content would be predominant. The percentage retention of room temperature adhesive strength at 125 °C and 150 °C of epoxy-imides for C/E ratio of 0.5 is compared in Fig. 5. It is noticed that EPN-1138-IDA-I is able to retain more than 100% of the room temperature adhesive strength at 125 and 150 °C. On the other hand, GY 250-IDA-I retains 36.5% and 11.5% of the room temperature adhesive strength at 125 and 150 °C respectively. For EHTPB-IDA-I, 9.3% and 6.7% retention of room temperature adhesive strength is observed at 125 and 150 °C respectively. Thus, it is evident that EPN 1138-based system is able to retain its room temperature adhesive strength at elevated temperatures even when C/E ratio is low. This has been attributed to the polyfunctional nature and higher aromatic content of EPN 1138. The poor retention of adhesive strength observed with EHTPB-IDA-I is due to the higher aliphatic content of EHTPB. In the case of GY 250-IDA-I, the percentage retention is higher than that of EHTPB and this is attributed to the higher aromatic content of the former system. The above conclusions are supported by T_g of these epoxy-mides (Table 2).

In order to understand the influence of aromatic and imide content on the adhesive properties, the relative percentage increase in adhesive strength at room temperature and at 150 °C of epoxy-imides when the C/E ratio is increased from 0.5 to 1.25 is compared in Fig. 6. It is noticed that the room temperature adhesive strength increases by 51, 64 and 20% for GY 250-IDA-I, EPN 1138-IDA-I and EHTPB-IDA-I respectively when the C/E ratio is increased from 0.5 to 1.25. Interestingly, for the same increase in the ratio, the adhesive strength at 150 °C increases by 641 and 500% for GY 250-IDA-I and EHTPB-IDA-I respectively whereas for EPN 1138-IDA-I only ~13% increase is observed. The above observation clearly brings out the fact that increase in the imide and aromatic content of epoxy-imide systems (by way of increasing the C/E ratio) influences the high temperature adhesive strength remarkably only for GY 250- and EHTPB-based systems. As explained earlier, among the epoxy resins, EHTPB is purely an aliphatic one and hence, increasing the C/E ratio increases the aromatic and

Table 3 Effect of carboxyl eqv to epoxy eqv ratio on adhesive strength of GY 250-IDA-I and EHTPB-IDA-I systems

C/E ratio	Adhesive lap shear strength of GY 250-IDA-I (MPa)				Adhesive lap shear strength of EHTPB-IDA-I (MPa)			
	R.T	100 °C	125 °C	150 °C	R.T	100 °C	125 °C	150 °C
0.25	19.0	14.0	5.0	2.5	–	–	–	–
0.50	19.2	18.2	7.0	2.2	15.0	6.3	1.4	1
1.00	23.1	21.0	18.0	11.4	18.0	12.0	7.0	4.1
1.25	29.0	22.2	21.0	16.3	18.0	14.0	8.0	6.0
1.50	27.0	25.3	21.1	15.3	17.0	12.3	10.3	7.0
2.00	22.0	21.4	22.0	15.0	13.0	12.0	8.0	6.4

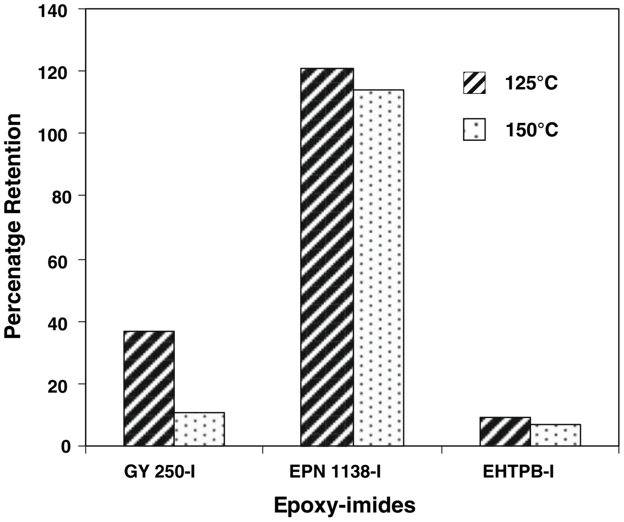


Fig. 5 Percentage retention of room temperature adhesive strength at elevated temperatures for the carboxyl to epoxy equivalent ratio 0.5

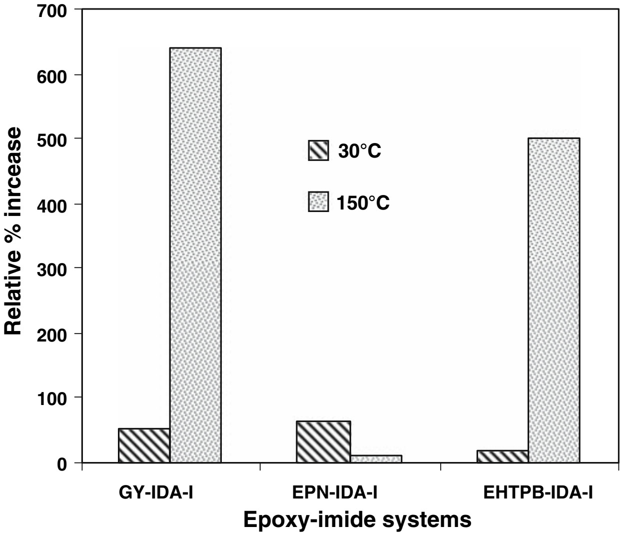


Fig. 6 Relative percentage increase in the adhesive strength at room temperature and at 150 °C of IDA-I-based systems with the increase in the ratio from 0.5 to 1.25

imide content of the system considerably, which in turn improves the high temperature adhesive strength. GY 250 being a difunctional epoxy resin, increasing the C/E ratio from 0.5 to 1.25 probably toughens the system in addition to increasing the imide and aromatic content. In addition, the presence of excess imide-diacid introduces crosslinks due to esterification reaction with dangling hydroxyl group. The above factors are responsible for the remarkable increase in high temperature adhesive strength of GY 250-IDA-I with the increase in the ratio. EPN 1138-based system being a highly crosslinked one with high aromatic content, the increase in the C/E ratio does not influence the adhesive strength to the same extent as in the other systems.

Conclusions

For epoxy-imides obtained by curing Araldite GY 250, Araldite EPN 1138 and EHTPB with *N*-(4-carboxyphenyl)trimellitimide, increase in T_g and thermal stability is more pronounced for EHTPB-based system than for the other two systems. Unlike Araldite GY 250 and Araldite EPN 1138, EHTPB is an aliphatic epoxy resin and hence, the effect of increase in aromatic content and imide content by way of increasing the C/E ratio is felt more for this system.

The adhesive lap shear strength at room temperature and at 100, 125 and 150 °C is influenced by both the nature of epoxy resin and C/E ratio. For any given C/E ratio, the retention of room temperature adhesive strength at elevated temperature follows the trend, EPN1138-IDA-I > GY 250-IDA-I > EHTPB-IDA-I. This trend is attributed to the polyfunctional and aromatic nature of Araldite EPN 1138, aromatic nature of Araldite GY 250 and aliphatic nature of EHTPB.

For all the systems, optimum room temperature and high temperature adhesive strength is obtained when C/E ratio is 1.25. Relative increase in high temperature adhesive strength with the increase in C/E ratio is more pronounced for GY 250-IDA-I and EHTPB-IDA-I systems than for EPN 1138-IDA-I.

Acknowledgements The authors are thankful to the Director, Vikram Sarabhai Space Centre (VSSC) for granting permission to publish this work. Thanks are due to Mr. K. S. Sastri and Dr. K. N. Ninan for encouragement. Help received from the members of the Analytical and Spectroscopy Division, VSSC for the thermal and spectral analysis of the samples is gratefully acknowledged. The authors also thank the reviewer for valuable comments. One of the authors (G.A.) is thankful to UGC, New Delhi for granting her a Fellowship.

References

1. McGrath JE, Dunson DL, Mechan SJ, Hedrick JL (1999) Adv Polym Sci 140:62
2. de Abajo J, de la Campa JG (1999) Adv Polym Sci 140:23
3. Mittal KL (1984) In: Polyimides-synthesis, characterization and application, vol I & II. Plenum, New York
4. Wilson D, Stenzenberger HD, Hergenrother PM (1990) In: Polyimides. Glassgow, Blackie, London
5. St. Clair AK, Stemp WS, St. Clair TL (1979) Adhes Age 22:35
6. Lee H, Stoffey D, Neville K (1967) In: New linear polymers. Mc Graw Hill, New York
7. de Abajo J (1992) In: Kricheldorf HR (ed) Handbook of polymer synthesis. Marcel Dekker, New York, p 941

8. Serafini T, Delvigs P, Lithsey GR (1972) J Appl Polym Sci 16:905
9. Kinloch AJ (1986) In: Structural adhesives. Elsevier, London
10. May CA (1980) In: Resins for aerospace, ACS Symp. Series No. 132. American Chemical Society, Washington
11. Moon YD, Lee YM (1993) J Appl Polym Sci 50:1461
12. Stern SA, Liu Y, Feld A (1993) J Polym Sci B, Polym Phys 31:939
13. Lee CJ (1984) U. S. Pat. 4,487,894
14. Serra A, Cadiz V, Martinez PA, Mantecon A (1986) Angew Makromol Chem 138:185
15. Chin WK, Shau MD (1996) In: Salamone CJ (ed) Encyclopedia of polymeric materials, vol 5. CRC Press, New York Chap 3, p 2210
16. Adhinarayanan K, Packirisamy S, Suma P, Ramaswamy R (1991) In: Sivaram S (ed) Polymer science, contemporary themes, vol I. Chapman & Hall, New Delhi, p 76
17. Adhinarayanan K, Packirisamy S, George RS, Rao VL, Ramaswamy R (1991) J Appl Polym Sci 43:783
18. Packirisamy S, Adhinarayanan K, Jameela SR, Ramaswamy R (1991) In: Sivaram S (ed) Polymer science, contemporary themes, vol I. Chapman & Hall, New Delhi, p 150
19. Abraham G, Packirisamy S, Adhinarayanan K, Feby AG, Ramaswamy R (2000) J Appl Polym Sci 78:1729
20. Abraham G, Packirisamy S, Vijayan TM, Ramaswamy R (2003) J Appl Polym Sci 88:1737
21. Abraham G, Packirisamy S, Ramaswamy R, Thomas S (2005) Int J Polym Mat 54:1107
22. Abraham G (2001) Ph.D. thesis, Mahatma Gandhi University, India 2001
23. Abraham G, Packirisamy S, Ravindran PV, Thomas S, Ramaswamy R, to be communicated
24. Latha PB, Adhinarayanan K, Ramaswamy R (1994) Int J Adhes Adhes 14:57
25. Ichino T, Hasuda Y (1987) J Appl Polym Sci 34:1667
26. Louis V, Adams M, Crannon JH (1986) In: Mark HF, Bikales NM, Overberger CG, Menges G (eds) Encyclopedia of polymer science and engineering, vol 6. Wiley Interscience, New York, p 322
27. Bhatnagar MS (1996) In: Salomone CJ (ed) Encyclopedia of polymeric materials, vol 5. CRC Press, Boca Racton, p 2233