



Preparation of a one-component epoxy adhesive using PET bottle waste derived terephthalic dihydrazide as latent curing agent

Neena George^{a,*}, T.K. Bindu Sharmila^b, Thomas Kurian^a

^a Department of Polymer Science and Rubber Technology, Cochin University of Science and Technology, Kochi, 682022, Kerala, India

^b Department of Chemistry, Maharaja's College, Kochi, 682011, Kerala, India

ARTICLE INFO

Keywords:

Epoxides
Lap-shear
Peel
Adhesive
Terephthalic dihydrazide

ABSTRACT

The present work discusses the effect of PET bottle waste derived terephthalic dihydrazide (TDH) as a latent curing agent in diglycidyl ether of bis-phenol A (DGEBA) based epoxy resin. Differential scanning calorimetric studies were conducted to examine the latent character of TDH and its curing pattern in DGEBA and FTIR spectroscopy was used to confirm the completion of curing. A latent accelerator (Ajicure PN-40) was used to reduce the extremely high cure temperature of DGEBA-TDH system to a feasible range. Cure characteristics, barrier and aluminium to aluminium (Al/Al) adhesive properties of the synthesized adhesive with varied TDH content were studied to optimize the dosage of TDH. Pot life of the synthesized adhesive was determined through brookfield viscosity measurements. Ageing studies of the bonded Al/Al samples were carried out to study the practical utility of the newly synthesized adhesive. The properties of TDH cured one-component epoxy adhesive were compared with a commercial dicyandiamide (DICY) cured one-component epoxy adhesive. TDH cured adhesive displayed comparable properties to the DICY cured adhesive.

1. Introduction

Epoxy resins constitute one of the most important classes of structural adhesives by virtue of their good adhesive properties coupled with high thermal and mechanical strength [1]. Epoxy adhesives find wide applications in the automotive, aerospace, construction, electrical and electronic industries. They are used to bond a variety of materials, including metals, wood, concrete, glass, ceramic, plastics, etc and the bonded substrates remain intact over a broad range of conditions [2]. The global epoxy adhesives market is estimated to grow from USD 5.33 billion in 2015 to USD 10.13 billion by 2026, at a compound annual growth rate (CAGR) of 6.63% during 2016–2026 [3].

Epoxy adhesives are usually produced as one or two component, ambient or heat curing liquids, pastes or films. One-component epoxy adhesives are easier to use and they offer the highest shear strengths and the greatest service temperature capabilities in comparison to two-component epoxy adhesives. The application of two-component epoxy adhesives may sometimes result in fragile bonds if curing of the adhesive commences much ahead of complete application of the adhesive and proper joining of the adherends. But in a one component epoxy adhesive, the dispersed latent curing agent remain dormant at ambient

conditions and reacts only when triggered by external stimuli such as heat, photoirradiation or ultrasonication [4]. Thus they can be applied on the adherend continuously without the threat of early curing at room temperature. The one-component based epoxy adhesives market is estimated to grow at the highest CAGR during 2016–2021, in terms of volume. The excellent properties of the one-component type adhesives such as, quick curing time, a solvent-less system, and consumer friendly usage, is driving the demand in this segment [3].

Thermal latent curing agents employed in one-component epoxy adhesives act mainly by two different mechanisms; the first one is based on their insolubility in epoxy resin, due to their crystalline character, which on melting becomes soluble and active. The second mechanism is based on the use of a precursor compound, inactive at room temperature, but under stimuli gets converted into an active curing agent [5]. Examples for the latter type include Boron trifluoride-amine complex, dicyandiamide, and organic acid dihydrazides. Among these, organic acid dihydrazides are generally considered as non-toxic and safe for industrial applications but its application still remains obscure since only a few scientific reports regarding the application of dihydrazides in epoxy resins are published till date. In 2001, Holbery et al. [6] has studied the cure history of different combinations of dihydrazide,

* Corresponding author.

E-mail address: neenapattamana@gmail.com (N. George).

<https://doi.org/10.1016/j.ijadhadh.2019.102524>

dicyandiamide, urea and imidazole for accelerated cure of E-glass fiber reinforced epoxy composites. In 2012, Tomuta et al. [7] have reported the synthesis and suitability of adipic dihydrazide, dodecanoic dihydrazide, isophthalic dihydrazide and 1,4-cyclohexyl dihydrazide as latent curing agent in diglycidyl ether of bis-phenol A (DGEBA) coatings.

In the present paper, we report our effort to prepare a one-component metal to metal epoxy adhesive using terephthalic dihydrazide as latent curing agent. Differential scanning calorimetric studies were conducted to scrutinize the latent cure pattern of the synthesized one-component epoxy adhesive. All adhesive properties of the prepared adhesive were optimized with respect to the dosage of TDH and cure conditions. The ageing studies of the bonded metal joints were also conducted at different environmental conditions. The properties of TDH cured DGEBA are also compared to that of a similar system cured in the presence of a commercial latent curing agent viz., dicyandiamide. The significance of our study lays on the fact that we have synthesized the virgin TDH through chemical recycling of PET bottle waste by aminolysis reaction with hydrazine monohydrate [8]. Thus this scientific report may be highly beneficial to the society in the perspective that it helps in sustainable plastic waste reclamation too.

2. Experimental

2.1. Materials

General purpose unmodified solventless epoxy resin B11 (Specific gravity: 1.15–1.20; Viscosity at 25 °C: 1250 cP; Epoxy Equivalent Weight: 190–200) and Isophenyl fumed silica (Aerosil 200) were supplied by Ms Sharon Engineering Enterprises, Cochin. The latent curing agent, terephthalic dihydrazide was synthesized from PET bottle waste flakes adopting a procedure described in our previous work [8]. The accelerator (Ajicure PN-40) was supplied by Ajinomoto Fine-Techno Co., Inc., Functional Chemicals Div., Japan.

2.2. Preparation of latent epoxy adhesive

Solventless DGEBA epoxy resin was accurately weighed out into a dried beaker. 2 phr antisetling agent (isophenyl fumed silica) was dispersed thoroughly in the resin by mechanical stirring at 60 °C. Varying dosages (22–26 phr) of synthesized latent curing agent, terephthalic dihydrazide (TDH) was then added and the ingredients were mechanically stirred for an hour at 70 °C. The mixture was then ultrasonicated for 30 min in an ice bath to achieve a good level of dispersion. After ultrasonication, the mixture was placed in a vacuum oven for 30 min at 60 °C to remove bubbles trapped in the mixture during the stirring and sonicating steps. Finally the accelerator, (Ajicure PN-40 (amine epoxy adduct)) was added to the mixture under stirring at 40 °C to ensure fine and uniform dispersion without aggregates. The prepared adhesive was then stirred thoroughly at 70 °C and poured into preheated Teflon moulds sprayed with a suitable mold releasing agent and cured under vacuum for 18 h at 90 °C followed by 3 h at 130 °C. The cure, thermal and adhesive properties of the optimum formulation was also compared with a commercial latent epoxy adhesive prepared using 7 phr dicyandiamide as curing agent and 4 phr Ajicure PN-40 as accelerator and cured by a similar procedure. The different formulations prepared were named as 'TaAb', where 'a' represents the amount of TDH added in phr units and 'b' represents the amount of Ajicure PN-40 added in phr units.

2.3. Mechanism of curing of DGEBA-TDH adhesive formulation

In epoxy resins, dihydrazides are typically formulated to ¼ of dihydrazide to each epoxy equivalent. Therefore all the four primary amine hydrogen's of TDH will react, each with one epoxide group [9]. DGEBA acts as a difunctional monomer and dihydrazide as tetrafunctional in a step growth polymerization mechanism (Fig. 1), similar to the

conventional epoxy/primary diamine system. It should be commented that only the NH₂ groups of TDH are nucleophilic enough to attack the oxirane rings of DGEBA resin [7].

2.4. Characterization techniques

FTIR spectrometer (Thermo Nicolet, Avatar 370 FTIR) with a resolution of 4 cm⁻¹ in the transmittance mode was used to monitor the curing behavior of epoxy-hydrazide formulations at 130 °C in a spectral range of 400–4000 cm⁻¹. The disappearance of the absorbance peak at 912 cm⁻¹ was used to monitor the epoxy equivalent conversion. Differential scanning calorimetry (DSC) was performed using Mettler Toledo DSC 822e at a heating rate of 5 °C/min under nitrogen atmosphere. For dynamic DSC scans, uncured samples (5–8 mg) mixed at a stoichiometric epoxide/hydrazide ratio was placed in hermetically sealed aluminium pans, and heated from room temperature to 300 °C. The glass transition temperature (T_g) was calculated after the complete dynamic curing, by means of a second scan, as the temperature of the half way point of the jump in the heat capacity when the material changed from the glassy to the rubbery state. The precision of the determined temperatures is estimated to be ±1 K. Thermogravimetric analyses (TGAs) were carried out in a TGA Q 50 Thermal Analyzer (TA Instruments) under N₂ atmosphere (50 mL/min). Pieces of the cured samples with an approximate mass of 8 mg taken in aluminium pans were degraded between room temperature and 800 °C at a heating rate of 20 °C/min.

The tensile lap shear tests were performed with a crosshead speed of 5 mm/min using Shimadzu Universal testing machine equipped with 10 KN load cell. Five samples for this test were prepared according to ASTM D1002. The procedure produced lap joint specimens with average adhesive thickness layer of 25 µm ± 2 measured by a micrometer gauge. 90° T-peel tests were carried out as per ASTM D 1876. 100 × 25 mm size of aluminium strips sliced from 0.8 mm thick sheets were used for peel studies while for lap shear strength studies strips of size 100 × 25 mm sliced from 1.6 mm thick sheets were used.

Tensile strength and elongation at break of epoxy specimens were tested on Shimadzu Universal Testing Machine (model-AG-1 series) equipped with 10 kN load cell using molded dumb-bell shaped specimens at a displacement rate of 50 mm/min as per ASTM D638. Impact strength of unnotched specimens were performed at room temperature by means of Ceast Resil Impact Analyser (Junior), according to ASTM D 4812–99. The pendulum employed has a capacity of 4 J and a striking velocity of 3.96 m/s, at room temperature. A rectangular sample of dimension 64 mm × 12.7 mm × 3.2 mm is clamped as a beam at the pendulum impact test fixture and struck at the middle. The energy required to fracture the sample was measured from the reading dial.

Water absorption studies of the epoxy castings with varied TDH dosage were performed according to ASTM D570. Standard specimens in the form of disks with a diameter of 50.8 mm and a thickness of 3.175 mm were dried at 80 °C in a vacuum oven until a constant weight was attained. Then they were immersed in water in a stainless steel water bath at 100 °C for an hour. The specimens were removed from the water bath, wiped with tissue paper and weighed on a balance with a precision of 1 mg. The percentage of water uptake was determined using equation (1).

$$\text{Water uptake (\%)} = \left(\frac{W_{\text{final}} - W_{\text{initial}}}{W_{\text{initial}}} \right) \times 100 \quad (1)$$

where W_{initial} and W_{final} denote the weight of dry specimens (the initial weight of specimens prior to exposure to the water absorption) and weight of specimens after water absorption, respectively.

3. Results and discussion

Equivalent stoichiometric amount of terephthalic dihydrazide (~26phr) required for curing the solventless DGEBA resin was

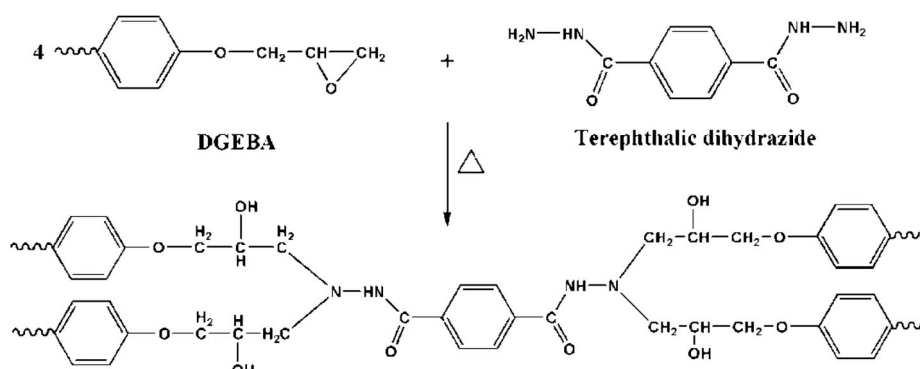


Fig. 1. General structure of the reagents and of the network formed [7].

calculated using Equation (2).

$$\text{Stoichiometric amount of TDH(phr)} = \frac{\text{Amine hydrogen equivalent of TDH}}{\text{Epoxy equivalent weight of DGEBA}} \times 100 \quad (2)$$

3.1. FTIR spectroscopy

The completeness of curing was investigated using FTIR spectroscopy. Fig. 2 shows the spectra of DGEBA resin, TDH and DGEBA-TDH thermosets with 26 phr TDH and varying (3–5 phr) dosage of the accelerator. The curing of all the thermosets were found to be completed within the stipulated curing schedule which was obvious from the disappearance of the epoxide peak at 912 cm^{-1} . It was well evident from the FTIR spectrum that the curing proceeds by nucleophilic attack of biguanide nitrogen's to epoxide groups since the broad peak at 3323 cm^{-1}

cm^{-1} due to N–H stretch vibrations of TDH got narrowed after curing. The terminal NH protons actively participate in the curing and are not available for intermolecular hydrogen bonding anymore whereas the participation of the inner N–H proton attached to C=O in any hydrogen bonding is limited due to steric factors. This justifies the narrowing of the broad N–H stretch band of TDH at 3323 cm^{-1} after curing. The band at 1620 cm^{-1} due to carbonyl stretch of TDH also becomes more visible after the curing has taken place [10]. A broad band due to intermolecular hydrogen bonded O–H stretch vibration (formed on opening of the epoxide ring during curing reaction) appears after curing at 3437 cm^{-1} . This broad O–H stretch band at 3437 cm^{-1} and narrow N–H stretch band at 3323 cm^{-1} was observed most prominently in the spectrum of T26A4 thermoset and thus it may be adjudged that optimum cure has taken place at 4 phr accelerator loading. Accordingly accelerator dosage was optimized as 4 phr throughout the TDH cured adhesive formulations.

3.2. Cure characteristics

Differential scanning calorimetry (DSC) was used to study the cure characteristics of the catalyzed and uncatalyzed epoxy/TDH formulations prepared incorporating 26 phr TDH. Fig. 3 shows the curing exotherms obtained for catalyzed and uncatalyzed epoxy/TDH compositions at a heating rate of $5 \text{ }^\circ\text{C}/\text{min}$. As can be observed, the cure curve obtained for uncatalyzed formulation was sharp and unimodal indicating that the reactions in the curing process take place abruptly once initiated. The cure process showed a latent character, as it react at a definite temperature much higher than $200 \text{ }^\circ\text{C}$. On addition of 4 phr

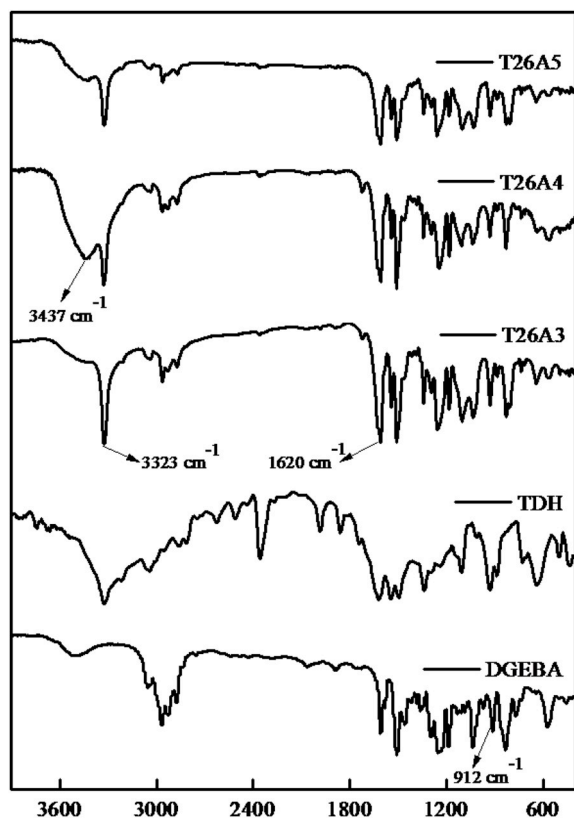


Fig. 2. FTIR spectra of DGEBA resin, TDH and cured DGEBA-TDH formulations prepared with 3, 4 and 5 phr accelerator.

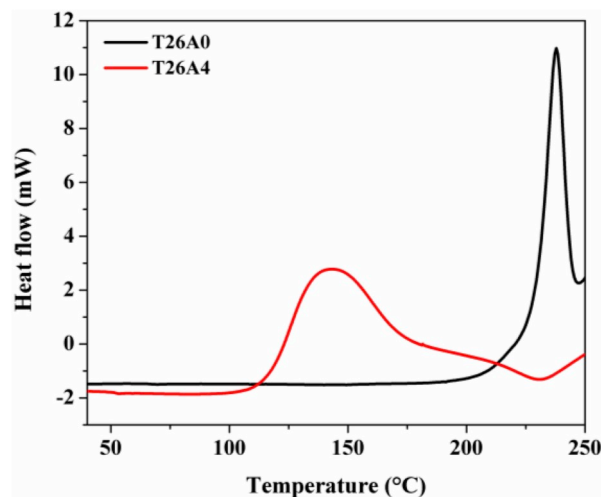


Fig. 3. DSC heat flow curves of epoxy adhesive formulation containing 26 phr TDH with and without 4 phr Ajicure PN-40 as accelerator at a heating rate of $5 \text{ }^\circ\text{C}/\text{min}$.

Ajicure PN-40 as accelerator, the cure curve got broadened and the peak cure temperature was decreased considerably to 142 °C, without any detrimental effect on its innate latent nature.

In addition to terminal active hydrogen's, inner NH hydrogen's in TDH also have an equal probability to enter in the curing process at high temperature once one each of the terminal hydrogen's on both sides is used up for crosslinking reaction. Thus the actual functionality of TDH should be assumed greater than four and this justify the necessity to optimize TDH in the prepared epoxy formulations. Dynamic DSC curves for the cure of DGEBA with 22, 24 and 26 phr TDH dosage at constant (4 phr) accelerator loading are shown in Fig. 4. The respective calorimetric data and the glass transition temperature (T_g) of the cured thermoset obtained in the second scan are given in Table 1. A single exotherm peak was obtained for all the three TDH cured formulations and the peak shifted towards lower temperatures due to enhanced cure rate with increased TDH content. T_g increased upon increase in TDH dosage from 22 to 24 phr but remained more or less constant on further increase in the TDH dosage.

3.3. Adhesive properties

Fig. 5(a) and (b) shows the effect of TDH loading on the lap shear strength and T-peel strength of the prepared adhesive. On varying the TDH dosage, the lap shear strength of the epoxy adhesive initially showed an increasing trend up to 24 phr TDH loading and thereafter the shear strength was decreased. Epoxy adhesive prepared with 24 phr TDH showed an optimum aluminium to aluminium (Al/Al) shear strength of 10.8 MPa which was 32% and 10% high as compared to the 23 and 25 phr counterparts respectively. The peel strength values also reflected a similar trend with 24 phr TDH loading showing optimum peel strength of about 1.8 N/mm. Mechanical interlocking was identified as the probable mechanism of adhesion, where the adhesive enters the pores or surround the projections on the aluminium substrate before solidification.

3.3.1. Optimization of Al/Al adhesive post cure temperature and post cure time

Among all the Al/Al adhesive formulations prepared, T24A4 was found to offer the best adhesive bond strength. Accordingly the post cure temperature and post cure time of T24A4 formulation was optimized based on the adhesive properties so as to fabricate the best possible adhesive joint. Based on the results (Table 2) 130 °C was found as the optimum post cure temperature. Thus the adhesive joints were cured at

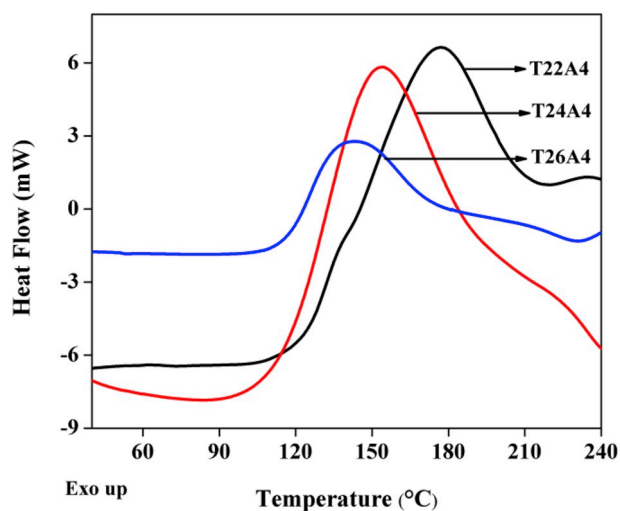


Fig. 4. DSC dynamic scanning curves for DGEBA-TDH formulations prepared with 22, 24 and 26 phr TDH in combination with 4 phr accelerator at a heating rate of 5°C/min.

Table 1

Calorimetric data of DGEBA-TDH formulations with varied TDH content.

TDH loading (phr)	Exothermic peak temperature (°C)	T _g (°C)
22	177	116
24	153	132
26	142	134

130 °C for 1–5 h and the lap shear strength and T-peel strength of the specimens were tested after each hour. The obtained results are tabulated in Table 3. An optimum Al/Al adhesive strength was obtained when the specimens were cured at 130 °C for 4 h.

3.4. Barrier properties

The percentage water absorption of the epoxy thermosets with varied TDH dosage was investigated as per ASTM D570 after immersing dried standard specimens in water at 100 °C for 1 h. When the TDH loading was increased from 22 phr to 24 phr, the percentage of water absorption of the epoxy thermosets got decreased from 0.75% to 0.64% as shown in Fig. 6.

On further addition of 2 phr dosage of TDH, the percentage water absorption of the thermosets got increased. At 24 phr of TDH loading, the crosslink density may have reached an optimum and likewise the percentage of water uptake of the polymer was also comparatively less. Further addition of TDH into the matrix may have created agglomerates in the matrix lowering its barrier properties considerably.

3.5. Ageing studies of T24A4 adhesive bonded joints

The percentage retention of Al/Al adhesive bond strength in different environmental conditions was studied and is tabulated in Table 4. The optimum composition T24A4 was selected for the ageing studies. It was concluded from the ageing studies that the bonded specimens remained more or less intact when aged under ambient temperature (303 K). But when the bonded joints were exposed to elevated temperatures, they showed a much reduction in the adhesive strength. This may be due to the decomposition of weak N–N bond of the dihydrazide without any mass loss (since no mass loss was recorded in TGA trace (Fig. 7) below 100 °C) at temperatures below 100 °C. The two exothermic decomposition peaks obtained below 100 °C at 70 °C and 88 °C for T24A4 thermoset in the second derivative plot of DSC thermogram (Fig. 8) is a clear evidence for this decomposition. The peak at 132 °C corresponds to its T_g. The decomposed N–N fragments can get recombined in a similar or different fashion on cooling. A much more decrease in the adhesive strength occurs when the samples were kept under water at 100°C. This may be due to the plasticization effect caused by the diffused water molecules at the bonded interface, increasing risks of mechanical debonding of the adhesive from the adherend. The application of high temperature makes the issue more critical. The refrigerated samples also displayed much less retention of properties, since at low temperatures the epoxy thermosets become more brittle decreasing the interfacial adhesion between adhesive and adherend.

3.6. Pot life of the prepared T24A4 adhesive

Pot life of an epoxy adhesive is defined as time during which the viscosity of the adhesive gets doubled [11]. Pot life plays a vital role in a latent adhesive selection for a specific manufacturing process since it reflects the desired shelf life of the prepared adhesive at a specific temperature. In the present study, pot life of the prepared latent adhesive kept at 30 °C was determined by measuring the brookfield viscosity of the resin at specific time schedules and the obtained results are plotted in Fig. 9. The initial brookfield viscosity of the prepared adhesive was 5400 cP, which on storage for 30 days at 30 °C got doubled to 10700 cP. Thus the effective pot life of the adhesive was found to be 30

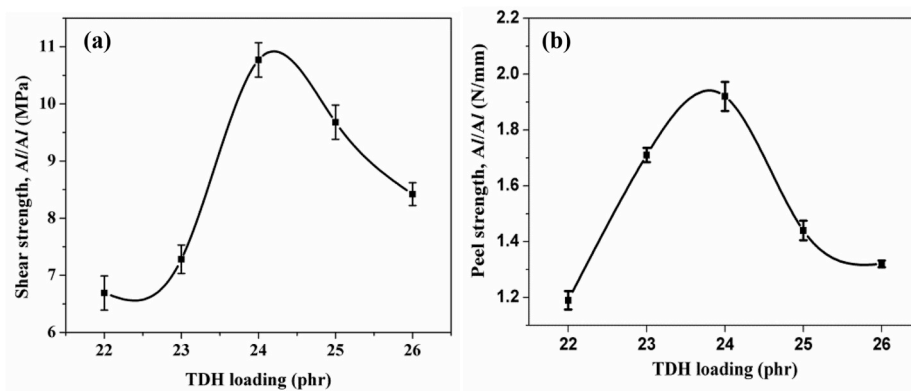


Fig. 5. Effect of TDH loading on Al/Al (a) lap shear strength and (b) T-peel strength of the prepared adhesive at constant accelerator loading.

Table 2
Optimization of the post cure temperature of the prepared T24A4 adhesive formulation.

Post cure temperature (°C)	Lap shear strength (MPa)	T-peel strength (N/mm)
120	9.39 ± 0.33	1.79 ± 0.04
130	10.77 ± 0.30	1.92 ± 0.02
140	10.12 ± 0.29	1.82 ± 0.03
150	8.53 ± 0.40	1.68 ± 0.09

Table 3
Optimization of the post cure time of T24A4 adhesive formulation at 130 °C.

Post cure time (h)	Lap shear strength (MPa)	T-peel strength (N/mm)
1	8.10 ± 0.27	1.25 ± 0.03
2	9.55 ± 0.33	1.50 ± 0.07
3	10.77 ± 0.30	1.92 ± 0.02
4	11.53 ± 0.23	2.09 ± 0.05
5	10.05 ± 0.37	1.73 ± 0.04

Table 4
Percentage retention of shear and peel strength of Al/Al adhesive joints aged at different conditions.

S. No.	Ageing Condition	Percentage retention of lap shear strength (%)	Percentage retention of peel strength (%)
1	Ambient temperature (6 months at 303 K)	99	98
2	Air-oven (100 °C-24 h)	89	92
3	Hot water (100 °C-24 h)	86	87
4	Salt water (100 °C-24 h)	84	84
5	Cold condition (0 °C-24 h)	84	82

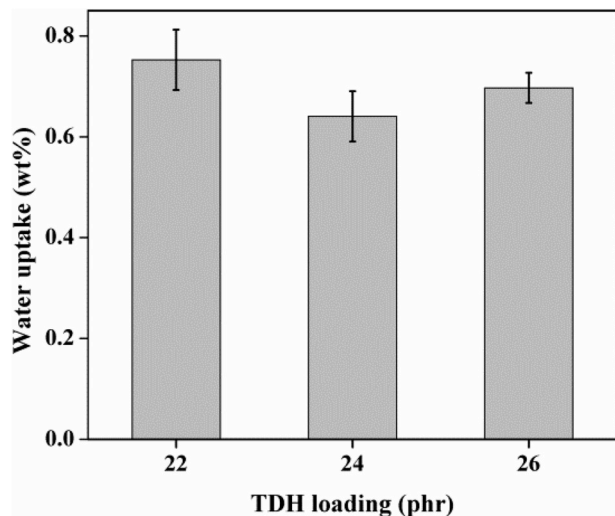


Fig. 6. Water absorption properties of epoxy/TDH thermosets with varied TDH dosage.

days.

3.7. Comparison of terephthalic dihydrazide with commercial dicyandiamide

A comparative study of the curing behaviour of DGEBA-TDH and

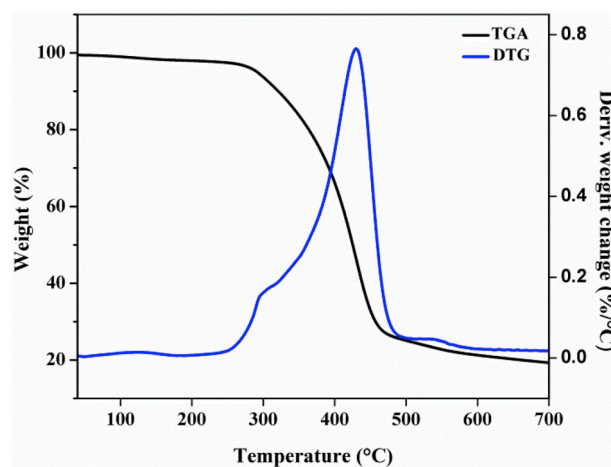


Fig. 7. TGA/DTG curve for T24A4 thermoset.

DGEBA/DICY combinations was conducted using differential scanning calorimetry and the dynamic scan curves are shown in Fig. 10. While an optimum dosage of 24 phr TDH was added as curing agent in epoxy/TDH system, only 7 phr DICY was added as curing agent in epoxy/DICY system as optimum dosage [12,13]. 4 phr Ajicure PN-40 was added as accelerator in both the systems. The curing performance of both the systems was quite comparable as evident from their cure curves. The mechanical/thermal properties, crosslink density and lap shear strength of both the systems were also compared and are tabulated in Table 5. As can be seen from the table, T24A4 thermosets displayed higher glass transition temperature, tensile and impact strength compared to the D7A4 thermosets. The lap shear strength and water uptake properties of both the thermosets were almost similar.

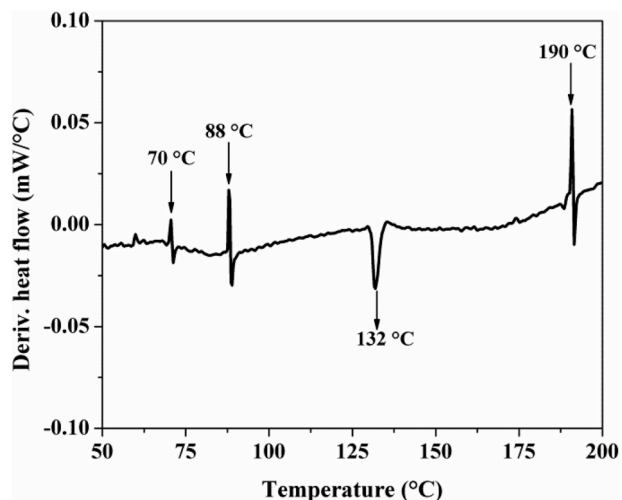


Fig. 8. The second derivative of DSC curve obtained for T24A4 thermoset specimen.

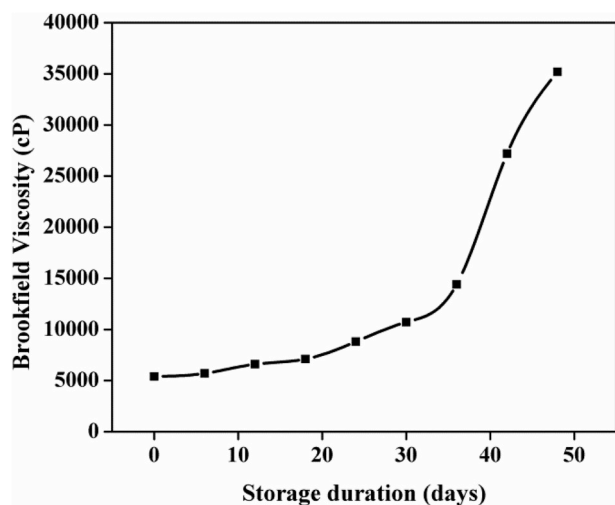


Fig. 9. Variation in brookfield viscosity of the prepared T24A4 adhesive with time.

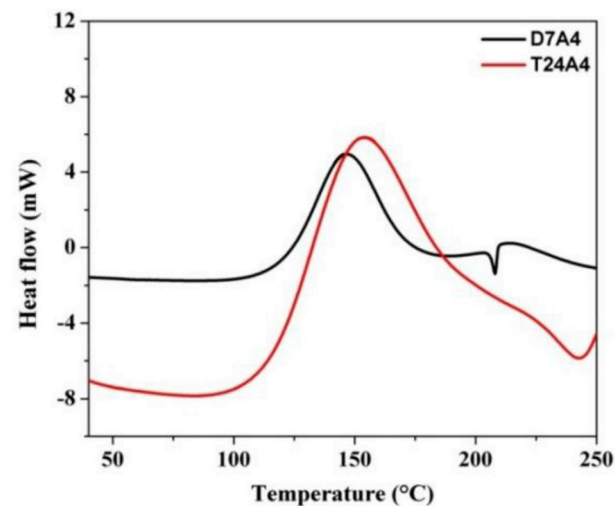


Fig. 10. DSC heat flow curves of DICY and TDH cured epoxy formulations.

Table 5

Properties of D7A4 epoxy thermosets versus T24A4 epoxy thermosets.

Properties	D7A4	T24A4
Tensile strength (MPa)	24.56 ± 0.43	28.54 ± 0.56
Elongation at break (%)	3.67 ± 0.19	3.87 ± 0.28
Impact strength (kJ/m ²)	7.61 ± 0.12	8.2 ± 0.20
Crosslink density (mol/g)	0.336 ± 0.006	0.157 ± 0.004
Glass transition temperature (°C) (from DSC)	110 ± 1 °C	130 ± 2 °C
Water uptake (wt%)	0.60 ± 0.06	0.64 ± 0.08
Lap shear strength (MPa)	11.24 ± 0.83	11.53 ± 0.76

4. Conclusions

DGEBA based one-component epoxy adhesives could be prepared using the PET bottle waste derived terephthalic dihydrazide as latent curing agent and Ajicure PN-40 as latent accelerator. The curing behaviour of the DGEBA-TDH systems could be studied by differential scanning calorimetry. Addition of 4phr Ajicure PN-40 as accelerator lowers the peak exothermic cure temperature of T26A0 adhesive from 238 °C to 142 °C and shifts to higher temperatures with subsequent decrease in dosage of curing agent. FTIR spectroscopy confirms optimum curing of the adhesive at 130 °C when 26 phr TDH and 4 phr accelerator is incorporated. The analysis of barrier and Al/Al adhesive properties with varied TDH content establishes the optimum dosage of curing agent in the formulation as 24 phr. All the cured DGEBA-TDH thermosets show thermal stability up to 246 °C. Cured T24A4 thermosets show a weight gain of about 0.64%, after 1 h of boiling in water. Post cure at 130 °C for 4 h results in higher lap shear strength and T-peel strength. Pot life of the synthesized adhesive (T24A4) was found to be 30 days. Terephthalic dihydrazide cured DGEBA adhesive shows improved Tg, tensile and impact properties and almost similar lap shear strength and % water uptake in comparison with dicyandiamide cured commercial one-component epoxy adhesive. However ageing studies has proved that the TDH cured epoxy adhesive gives best results only below a temperature of about 70 °C.

Acknowledgments

The authors would like to thank UGC (University Grants Commission, New Delhi, India) for providing funds for the research work. The authors gratefully acknowledge Ajinomoto Fine-Techno Co., Inc. Functional Chemicals Div., Japan for providing the latent accelerator (Ajicure PN-40) required for the work.

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