

Reprocessable Epoxy Networks with Tunable Physical Properties: Synthesis, Stress Relaxation and Recyclability

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Abstract In order to extend the application of epoxy vitrimer, 1,4-cyclohexanedicarboxylic acid (CHDA) was used as a co-curing agent and structure modifier for sebacic acid (SA) cured diglycidyl ether of bisphenol A (DGEBA) epoxy vitrimer to tailor the mechanical properties of epoxy vitrimers with 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) as a transesterification catalyst. The glass transition temperature (T_g) of vitrimer increased gradually with the increase in CHDA content. Vitrimers behaved from elastomer to tough and hard plastics were successfully achieved by varying the feed ratio of CHDA to SA. Both the Young's modulus and storage modulus increased apparently with the increase in CHDA content. Stress relaxation measurement indicated that more prominent stress relaxation occurred at elevated temperatures and the stress relaxation decreased with the increase of CHDA content due to the reduced mobility of the vitrimer backbone. The vitrimers showed excellent recyclability as evidenced by the unchanged gel fraction and mechanical properties after compression molded for several times. With tunable mechanical properties, the epoxy vitrimers may find extensive potential applications.

Keywords Epoxy vitrimer; Mechanical properties; Stress-relaxation; Recyclability

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INTRODUCTION

The permanent network structures endow the thermoset polymers with superior dimensional stability, mechanical properties and environmental resistance, thus make them irreplaceable in numerous applications such as in aircraft industry, high performance composite and solar cell encapsulant^[1]. However, thermoset polymers cannot be reshaped, processed or recycled due to the permanent crosslinking structures^[2]. To construct the reversible thermoset polymer by dynamic covalent bonds provides an attractive way to impart processability, since the networks with reversible crosslinks are able to be rearranged under suitable conditions similar to thermoplastic^[3–5].

Processable thermoset polymers, also known as covalent adaptable networks^[6, 7], can be fabricated by incorporation of various reversible covalent bonds, such as Diels-Alder (D-A) linkages^[8–11], disulfide crosslinks^[5, 12–14], ionic crosslink^[15] and dynamic exchangeable crosslinks^[16–18]. These processable network polymers are simply classified into two groups according to the reversible mechanisms, namely, dissociative and associative^[16, 18]. For the dissociative mechanism, the covalent bonds firstly break and then reform, while for the associative one, the former covalent bond is only

broken when a new covalent bond has been formed. Polymer networks containing Diels-Alder (D-A) structures are a typical example of dissociative and exchangeable network with processibility^[10]. The D-A linkage is a reversible covalent bond which breaks/reforms quickly upon heating, and the D-A linkage tends to dissociate due to the chemical equilibrium shifting relatively towards the endothermic side^[7, 19]. Thus, the polymer with this type of networks undergoes fast topology rearrangement to show plasticity because of a decrease in connectivity and a sudden drop in viscosity. Transesterification between ester group and hydroxyl group in acid/anhydride cured epoxy represents a typical associative exchange mechanism^[19]. The term “vitrimer” is introduced to describe such a processable epoxy resin as its viscosity behaves like a distinctive vitreous silica upon heating, namely, decreasing gradually^[1].

Epoxy resin represents a kind of versatile network polymer with diverse industrial applications such as elastomer, coating, adhesive, electrical/electronic laminates and high performance composites^[20, 21]. The global production of epoxy is projected to reach 3 million tons by this year, which may result in drastic thermoset waste accumulation after discarding, since they are neither thermally recyclable nor degradable due to their permanent network structure^[21–23]. Therefore, the development of reprocessed epoxy resins is very attractive from the viewpoint of waste utilization and environmental protection.

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Since Leibler and coworkers reported the zinc acetate catalyzed processable epoxy vitrimer in 2011^[1], a number of epoxy vitrimers and their composites have been documented^[24–32]. For example, Ji *et al.*^[24, 25, 31] synthesized a series of elastic sebacic acid cured epoxy vitrimers and composites with 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) as a transesterification catalyst. Legrand and Soulié-Ziakovic^[29] reinforced the rubbery epoxy vitrimer by incorporation of modified nano-silica to form silica-vitrimer nanocomposites. Yang *et al.*^[32] prepared thermally reconfigurable shape memory vitrimer composites by incorporation of graphene into TBD catalyzed sebacic acid cured diglycidyl ether of bisphenol A (DGEBA) vitrimer. It is noted that the vitrimers in these systems are usually elastic epoxy resins, which are cured with long-chained dicarboxylic acid. Thus, the obtained epoxy resins showed elastic mechanical behavior at room temperature due to their relatively low glass transition temperature^[24, 25, 29, 31, 32]. The elastic epoxy vitrimers cannot meet the requirements for important applications, particularly for the electrical/electronic laminates and high performance composites. Therefore, to tailor the physical properties of epoxy vitrimers for expanding the practical applications of such reprocessing epoxy resins remains a challenging project.

In this study, we incorporate a rigid dicarboxylic acid, namely 1,4-cyclohexanedicarboxylic acid (CHDA) into sebacic acid cured epoxy resins in the presence of TBD as a transesterification catalyst. We find that the glass transition temperature and the mechanical properties are easily regulated by variation the weight ratio of CHDA and SA. The obtained epoxy vitrimers show the mechanical properties with the similarities of soft elastomer and tough or hard plastics. Although sporadic study has reported the shape memory performance regulation of epoxy vitrimer by combining mixed carboxylic acids as curing agents^[33], the example referred to tailoring the mechanical properties and recyclability of epoxy vitrimers is rarely reported.

EXPERIMENTAL

Materials

Diglycidyl ether of bisphenol A (DGEBA, DER 332) with epoxy equivalent of 174 was procured from Dow Chemical Company. Sebacic acid (SA, 98.5%), 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD, 97%) and 1,4-cyclohexanedicarboxylic acid (CHDA, *cis/trans* isomer, AR grade) were obtained from Micxy Chemical Co., Ltd (Chengdu, China). Chloroform was purchased from Chuandong Chemical CO., LTD (Chongqing, China).

Synthesis of Epoxy Vitrimer from DGEBA, CHDA and SA

Epoxy vitrimer was synthesized *via* bulk curing of DGEBA with SA and CHDA by using TBD as the transesterification catalyst at 170 °C for 30 min. The molar ratio of –COOH/epoxy was kept at 1:1 for all the samples. Four different vitrimers containing CHDA and SA with weight ratio of 1/9, 2/8, 3/7 and 4/6 were synthesized. They were named as EVC₁S₉, EVC₂S₈, EVC₃S₇ and EVC₄S₆, respectively. The detailed synthetic procedures for the vitrimers were as follows: DGEBA, SA and CHDA with

predetermined amounts were added into a 100 mL round-bottomed flask which was vacuumed and purged with N₂ for 3 times and then was put into a 170 °C silicon oil bath. Magnetic stirring bar was used to mix the reactants. TBD (2.5 mol% of –COOH group) was added into the flask when homogeneous mixtures were obtained and the reaction was terminated after 30 min. The products were taken out of the flask with a tweezer before cooling. The vitrimers consisting of DGEBA with CHDA or SA alone were prepared as the controls and abbreviated as EVC and EVS, respectively.

Tensile Tests

Dumbbell-shaped specimen with width of 4 mm for tensile test was cut from vitrimer sheet with thickness of 0.5 mm that was prepared by compression molding at 200 °C under 20 MPa for 10 min. The tensile test was performed on a MTS E44 universal testing machine at room temperature (~30 °C) with crosshead speed of 10 mm/min. The length between the two pneumatic grips was 25 mm. International standard ISO 527-3 was referenced during testing. Five measurements were carried out for each sample and the averaged result was reported.

Fourier Transform Infrared (FTIR) Spectroscopy

Fourier transform infrared (FTIR) spectra of the samples under reflective mode were recorded on a RF-5301PC spectrophotometer (Shimadzu, Japan) in the wavenumber range from 4000 cm⁻¹ to 400 cm⁻¹ with the resolution and scanning time of 4 cm⁻¹ and 32 times, respectively.

Gel Fraction Measurement

The gel fraction of the sample was measured by solvent extraction. About 1 g (*w*₁) of sample was immersed in 25 mL of chloroform at room temperature for 3 days to dissolve the un-crosslinked part. The insoluble residue was isolated from the solution by filtration and weighed (*w*₂) after drying at 80 °C for 12 h. The gel fraction (*G_f*) was calculated by the Eq. (1):

$$G_f(\%) = w_2/w_1 \times 100 \quad (1)$$

Stress Relaxation

Stress relaxation was performed on a TA DHR-1 rotational rheometer right after curing of the samples. After 10 min equilibration from the curing temperature to the selected temperature, a 1% strain step was employed and the plot of storage modulus versus time was recorded. Good contact between the sample and the plates was guaranteed by the *in situ* curing process.

Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry analysis was performed on a NETZSCH instrument DSC-214. The samples with ~6 mg in aluminum pans were heated from –40 °C to 120 °C at a scanning rate of 10 K/min under N₂ atmosphere. The curves were recorded for glass transition temperature analysis.

Dynamic Mechanical Analysis (DMA)

Thermo-mechanical properties of the samples were measured on a TA DMA Q800 dynamic mechanical analyzer under a tensile resonant mode at a heating rate of 3 K/min from –60 °C to 120 °C and at a frequency of 2 Hz.

RESULTS AND DISCUSSION

Epoxy Vitrimer Synthesis

Epoxy vitrimer was synthesized by curing of DGEBA with SA and CHDA in the presence of TBD as a transesterification catalyst, as shown in Scheme 1(a). The epoxy network tends to undergo transesterification between ester group and hydroxyl group at elevated temperature with the help of TBD (Scheme 1b), leading to a dynamic network with the ability for rearranging topology to exhibit plasticity^[24, 31].

In order to show the processability of epoxy vitrimers, we thermally processed the samples through compression molding at 170 °C under 20 MPa for 10 min. Figure 1 shows the digital photos of EVS before and after processing. The vitrimer particles were changed into flat and transparent sheet after compression molding, indicating the good thermal processability. In order to prove the network structure of vitrimer, the gel fraction (G_f) was measured. All vitrimers showed gel fraction around 87%. The specific G_f values were ($88.9 \pm 1.2\%$), ($88.7 \pm 0.8\%$), ($86.8 \pm 1.4\%$), ($87.4 \pm 0.4\%$), ($86.9 \pm 1.6\%$) and ($87.8 \pm 0.6\%$) for EVS, EVC₁S₉, EVC₂S₈, EVC₃S₇, EVC₄S₆ and EVC, respectively. The results indicated that the processable vitrimer had the network structure and the variation of CHDA/SA ratio almost did not change the gel fraction of the resulting vitrimer.

FTIR spectroscopy was used to characterize the structure of epoxy vitrimer. Figure 2 shows the FTIR spectra of DGEBA, CHDA, SA and the typical vitrimer EVC₄S₆.

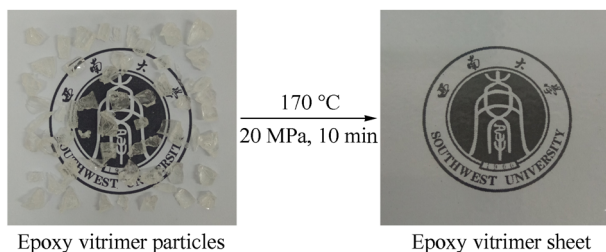


Fig. 1 Preparation of epoxy vitrimer sheet from its as-synthesized particles

Obviously, the characteristic epoxy band of DGEBA at 915 cm^{-1} disappeared in the spectrum of EVC₄S₆ after curing with CHDA and SA, and the strong absorption corresponding to the carbonyl group of $-\text{COOH}$ for CHDA and SA shifted to 1732 cm^{-1} due to the formation of ester groups by the reaction between epoxy and $-\text{COOH}$ groups. The combination of FTIR and gel fraction analysis indicated that the curing of DGEBA with CHDA and SA was achieved.

Glass Transition Temperature

The main purpose of this study is to incorporate rigid CHDA into SA cured DGEBA vitrimer for regulating the mechanical properties. The rigid CHDA not only affected the stiffness of vitrimer's backbone but also influenced its glass transition temperature (T_g), which in turn affected its mechanical properties because T_g determined the state of amorphous polymers at a specific application temperature. The effect of CHDA content on T_g of epoxy vitrimer was investigated by DSC. As shown in Fig. 3(a), the T_g values of EVS and EVC are 31.1 and 81.7 °C, respectively. With increasing content of CHDA, T_g of vitrimers shifted to a

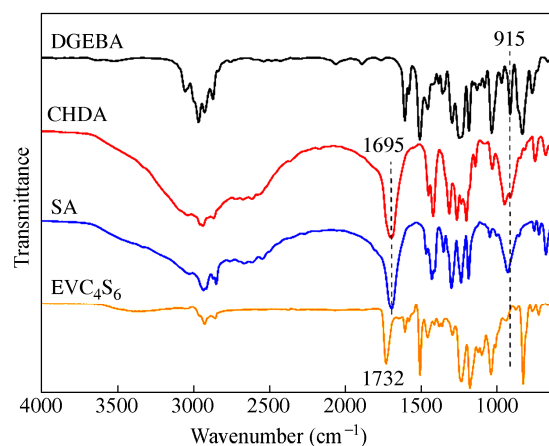
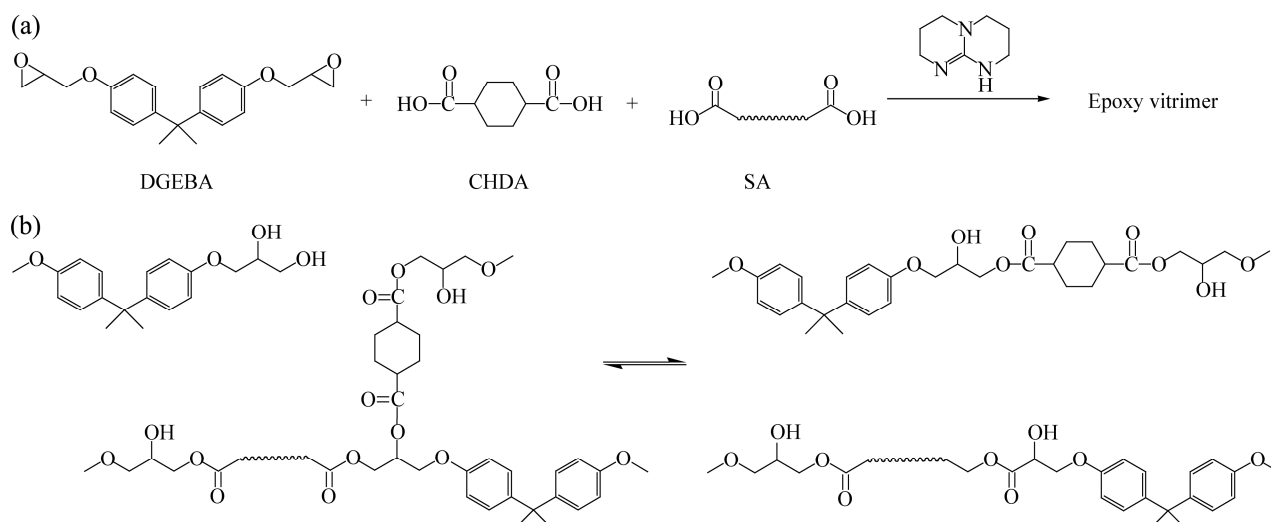


Fig. 2 FTIR spectra of DGEBA, CHDA, SA and EVC₄S₆, respectively



Scheme 1 (a) Synthesis of epoxy vitrimer from DGEBA, SA and CHDA with TBD as a catalyst; (b) Exchange process via transesterification in the epoxy network

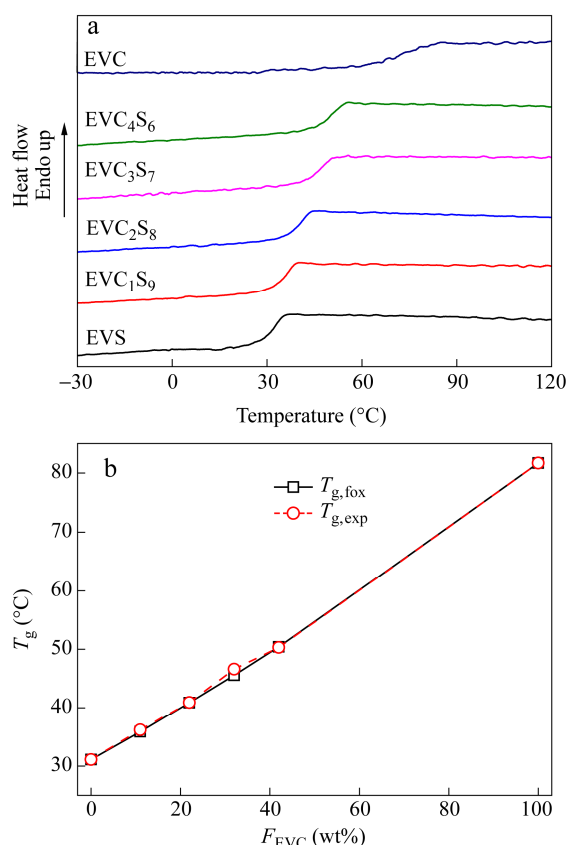


Fig. 3 (a) DSC heating scans of the epoxy vitrimers with different compositions; (b) Glass transition temperature (T_g) versus weight fraction of EVC (F_{EVC})

higher temperature range due to the increased chain stiffness. The values were 36.2, 40.9, 46.6 and 50.3 °C for EVC₁S₉, EVC₂S₈, EVC₃S₇ and EVC₄S₆, respectively.

If we define EVS and EVC as homo-vitrimer, the vitrimers containing both SA and CHDA can be supposed to be co-vitrimers. The EVS and EVC were miscible in the co-vitrimers as they all showed a single T_g . For the miscible blends or copolymers, the T_g could be evaluated by the Fox equation (Eq. 2):

$$1/T_{g,fox} = F_1/T_{g,1} + F_2/T_{g,2} \quad (2)$$

where $T_{g,1}$ and $T_{g,2}$ are the glass transition temperatures of EVS and EVC, F_1 and F_2 are the weight fractions of EVS and EVC^[34]. The $T_{g,fox}$ and $T_{g,exp}$ versus weight fraction of EVC (F_{EVC}) are graphically shown in Fig. 3(b). The $T_{g,exp}$ almost completely overlapped with $T_{g,fox}$, indicating that EVS and EVC were fully miscible in the co-vitrimers.

Mechanical Properties

The static mechanical properties were measured by tensile test to understand the effect of vitrimer composition on the elongation at break (ϵ), tensile strength (σ) and Young's modulus (E). Figure 4(a) shows the stress-strain curves of the vitrimers with different compositions. Notably, the curve of EVC was not obtained because it was too brittle to prepare dumbbell-shaped specimen through cutting. Due to the particular poor mechanical properties, the thermo-mechanical properties and stress relaxation are also not discussed in the

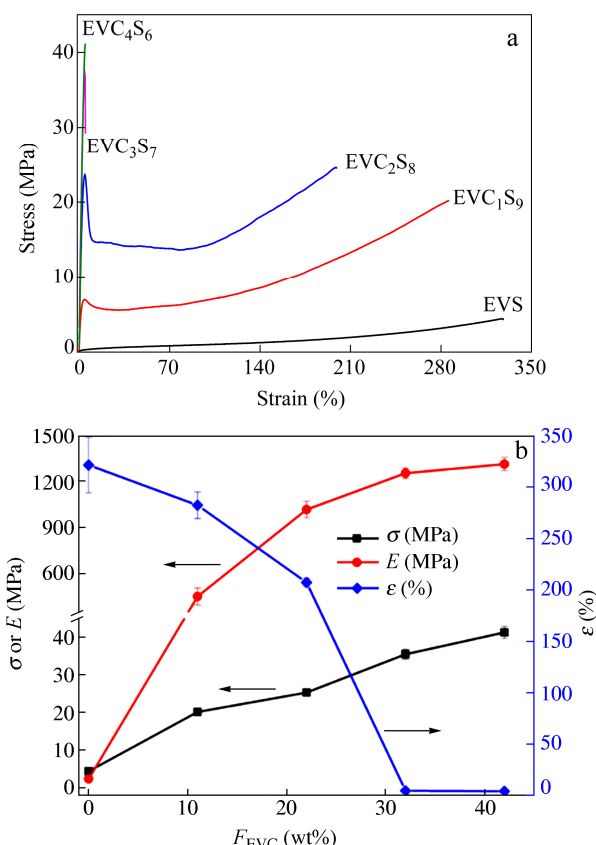


Fig. 4 (a) Stress-strain curves of epoxy vitrimers with different compositions; (b) Effect of weight fraction of EVC on the mechanical properties of epoxy vitrimers

following sections. EVS showed typical behavior of elastomer with a high elongation at break and a low tensile strength and Young's modulus. The values of ϵ , σ and E were $(321 \pm 27)\%$, (4.43 ± 0.26) MPa and (2.35 ± 0.14) MPa, respectively. The elastic vitrimer gradually changed to tough and hard plastics with incorporation of CHDA. The elongation at break decreased while tensile strength and Young's modulus increased with increasing content of CHDA (Fig. 4b). With the incorporation content of CHDA lower than 20 wt%, both EVC₁S₉ and EVC₂S₈ showed the tough plastic behavior with obvious yielding point, stable necking, cold drawing and strain hardening. The yield strength of EVC₁S₉ was (6.96 ± 0.23) MPa, while EVC₂S₈ increased to (23.63 ± 0.35) MPa. Due to the strain hardening behavior, the tensile strength values of both vitrimers were higher than the yield strength, with the values of (20.17 ± 0.56) and (25.33 ± 0.85) MPa, respectively. Compared to EVS, the E values enhanced by ~ 193 and 433 times to (452.9 ± 56.2) and (1017.4 ± 54.1) MPa for EVC₁S₉ and EVC₂S₈, respectively; meanwhile, ϵ values decreased gradually to $(282 \pm 13)\%$ and $(207 \pm 4)\%$, respectively. With further increasing CHDA content to 30 wt% or 40 wt%, EVC₃S₇ and EVC₄S₆ changed to hard and brittle plastics. Although both tensile strength and Young's modulus increased gradually to (35.49 ± 1.37) and (1253 ± 35.8) MPa for EVC₃S₇, and (41.32 ± 1.62) and (1312.4 ± 43.1) MPa for EVC₄S₆, the elongation at break decreased abruptly to less than 5%.

CHDA is a rigid dicarboxylic acid. It could increase the backbone stiffness and glass transition temperature of vitrimer, thus enhanced the strength and modulus, but reduced the elongation at break of vitrimer. EVS showed a T_g of 31.1 °C, which was very close to the temperature for tensile test. Therefore, segment motion of EVS was not frozen, as discussed later in DMA analysis, thus it showed tensile behavior of elastomer. For EVC₁S₉ and EVC₂S₈, the T_g increased to 36.2 and 40.9 °C, which were slightly higher than the testing temperature. Segment motion of EVC₁S₉ and EVC₂S₈ were frozen at testing temperature (~30 °C) since they were in the glassy state. The difference between T_g and testing temperature was approximately 10 °C or even smaller, which resulted in the sample with forced high-elastic deformation under loading^[35]. Both EVC₁S₉ and EVC₂S₈ showed high elongation at break, although the values were relatively lower than that of EVS. When the content of CHDA increased to 30 wt% and 40 wt%, the T_g s increased to 46.6 and 50.3 °C, which were much higher than the testing temperature. Consequently, forced high-elastic deformation was absent for both of EVC₃S₇ and EVC₄S₆, leading to rather low elongation at break. Except for the variation of T_g , the increased backbone stiffness also caused the reduction in elongation at break of vitrimers. Furthermore, the enhanced tensile strength and Young's modulus were attributed to the increased stiffness of vitrimers with increasing the content of CHDA. It enables the vitrimer in glassy state to exhibit high strength and modulus due to the frozen segment. The results of tensile testing proved our initial purpose for regulating the mechanical properties of epoxy vitrimers by incorporation of rigid CHDA moiety. Epoxy networks were successfully prepared, which showed the properties from elastomer to tough and hard plastics.

The dynamic mechanical properties of vitrimers were studied by DMA. Figure 5 shows the temperature dependence of storage modulus (E') and $\tan\delta$ of the samples. It is apparent that the storage modulus of vitrimer increased with increasing content of CHDA moiety at a given temperature. For example, the E' value of EVS at -20 °C was 1555 MPa, while that of EVC₁S₉, EVC₂S₈, EVC₃S₇ and EVC₄S₆ increased to 1696, 1765, 1854 and 2445 MPa, respectively. A prominent drop in E' plots occurred for all the vitrimers, corresponding to the α -relaxation (*i.e.*, glass transition) of the samples. The α -relaxation shifted to higher temperature range with increasing content of CHDA moiety. The changes in both storage modulus and α -relaxation were attributed to the increased stiffness of vitrimer backbone with increasing content of rigid CHDA moiety. The peak temperature of α -relaxation of $\tan\delta$ plots was denoted as the T_g of vitrimer. The values were 33.6, 39.5, 42.6, 51.7 and 57.9 °C for EVS, EVC₁S₉, EVC₂S₈, EVC₃S₇ and EVC₄S₆, respectively. These values were somewhat higher than those obtained by DSC measurement due to the different principles of the two methods.

The T_g s of all samples were higher than the temperature for tensile test. Thus, it seems that all the samples should be in the glassy state to exhibit hard and brittle tensile behavior during tensile test. However, it should be noted that the T_g is chosen

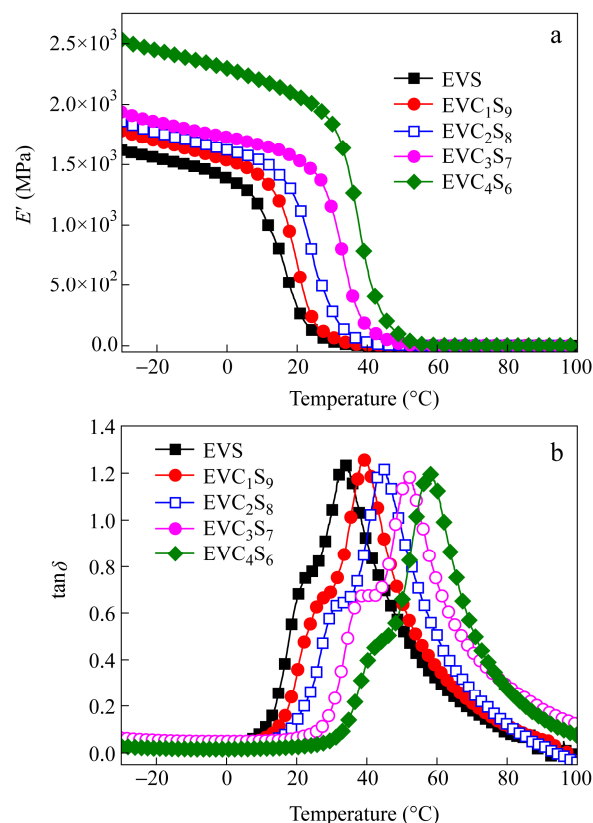


Fig. 5 (a) Storage modulus (E') and (b) $\tan\delta$ versus temperature for the epoxy vitrimers with different compositions

as the temperature of α -relaxation peak. As a matter of fact, the α -relaxation initiated from much lower temperature range, which meant the segment motion occurred at lower temperature. In order to gain the insight into the different tensile behaviors of vitrimers, the storage moduli of samples at 30 °C (close to ambient temperature for tensile test) were compared. The values were 34.9, 71.7, 281.5, 1126 and 1838 MPa for EVS, EVC₁S₉, EVC₂S₈, EVC₃S₇ and EVC₄S₆, respectively. The E' values of EVS, EVC₁S₉ and EVC₂S₈ dropped considerably at 30 °C compared to those in the glassy state such as at -20 °C, which indicated that the segment motion of the three samples was activated at 30 °C, corresponding to the relatively high elongation at break. The segment mobility decreased from EVS to EVC₁S₉ and EVC₂S₈ at 30 °C due to the increased backbone stiffness with increasing the content of CHDA moiety as evidenced by the increased E' values, which accounted for the increased tensile strength, Young's modulus and decreased elongation at break. The E' values of EVC₃S₇ and EVC₄S₆ at 30 °C remained at the same order of magnitude compared to the values at -20 °C, corresponding to the hard and brittle behavior of the two samples. It was noted that there was a shoulder peak before α -relaxation for all the vitrimers, which might be ascribed to the α -relaxation of non-crosslinked part of the samples since the gel fraction of all the samples was ~87 wt%. There was ~13 wt% non-crosslinked epoxy, which might show relatively lower glass transition temperature relative to the network vitrimer.

Rheology and Recyclability

The flow property of the epoxy vitrimers was studied by shear stress relaxation. The effect of temperature on the shear stress relaxation of vitrimer was studied by using EVC_2S_8 as an example (Fig. 6a). Relaxation occurred very slowly at 100 °C due to the fact that the topology of network was almost frozen at such a temperature. Obvious stress relaxation occurred when the temperature increased to 140 °C or higher, *i.e.*, the higher the temperature, the faster the relaxation. It is attributed to the fast arrangement in the topology of vitrimer network because of the accelerated transesterification reaction at elevated temperatures^[28]. Based on the Maxwell model for viscoelastic materials, the time required to relax 63% of the initial stress can be used as the relaxation time^[36, 37]. The relaxation times of EVC_2S_8 at 140, 170 and 200 °C observed from the stress relaxation plots were 830, 106 and 41 s, respectively. The effect of CHDA content on the stress relaxation of vitrimers was studied at the temperature of 170 °C, as shown in Fig. 6(b). It is obvious that the relaxation became slow with increasing content of CHDA owing to the increased stiffness to reduce the mobility of vitrimer backbones. The relaxation time of EVS at 170 °C was 71 s, and those of EVC_1S_9 , EVC_2S_8 , EVC_3S_7 and EVC_4S_6 increased to 75, 106, 178 and 507 s, respectively. The results indicated that the increased stiffness of epoxy vitrimers reduced the stress relaxation rate. However, they all showed obvious stress relaxation, indicating the thermal processability

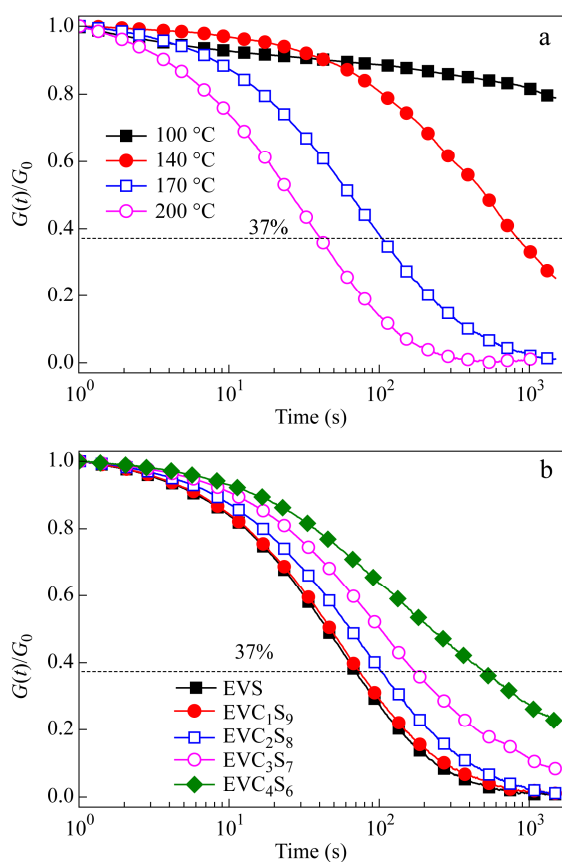


Fig. 6 (a) Stress-relaxation curves at different temperatures for EVS_2C_8 and (b) stress-relaxation curves of vitrimers with different compositions at 170 °C

for all the samples.

The recyclability of vitrimer was examined by compression molding of the cut sample pieces at 170 °C under 20 MPa for 10 min. The smooth and transparent sheet without crack was obtained (Fig. 7), indicating that the samples showed good thermal processability. To make sure the reversibility, the sample preparation was repeated for three times and the recycled vitrimer sheet was subjected to gel fraction measurement and tensile test. The gel fraction was all in the range of 85 wt%–89 wt% for the reprocessed samples, close to the values of as-synthesized vitrimers, indicating that reprocessing did not destroy the network structure of vitrimer.

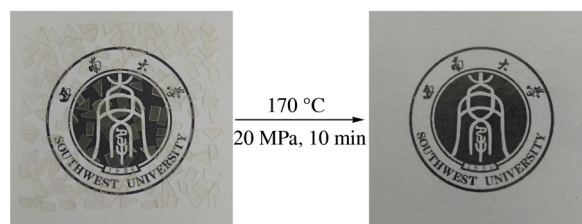


Fig. 7 Recycling of epoxy vitrimer network chips by compression molding

Figure 8 shows the stress-strain curves of EVS, EVC_2S_8 and EVC_4S_6 after being molded at 170 °C for 10 min for three times. It was obvious that the mechanical properties of all vitrimers recovered after recycling. Both gel fraction measurement and tensile test indicated that the epoxy vitrimers with different compositions showed excellent recyclability without loss in mechanical properties and gel fraction, which was ascribed to the dynamic nature of network of vitrimers.

CONCLUSIONS

Epoxy vitrimers with tunable mechanical properties from elastomer to tough and hard plastics were successfully synthesized by curing DGEBA with SA and CHDA using TBD as a transesterification catalyst. CHDA as a rigid moiety increased the stiffness, resulting in the epoxy vitrimer with higher glass transition temperature. The vitrimer without CHDA was almost in rubbery state at room temperature and showed the elastic mechanical behavior. Incorporation of CHDA with the content lower than 20 wt%, the T_g s of vitrimers were about 10 °C higher than room temperature, at which forced high-elastic deformation occurred for the vitrimers to exhibit typical behavior of tough plastic. When the content of CHDA increased to 30 wt% or higher, the vitrimer changed to hard and brittle plastic due to the much higher glass transition temperature and the stiffness of vitrimer's network. The content of CHDA affected the stress-relaxation behavior of the vitrimers significantly. The relaxation rate decreased with increasing content of CHDA due to the increased stiffness of vitrimer backbone. The vitrimers with different compositions showed excellent thermal recyclability without loss in mechanical properties and gel fraction. With tunable mechanical properties and excellent recyclability, the vitrimers may find wide applications from elastomer to plastics. By this way, it is

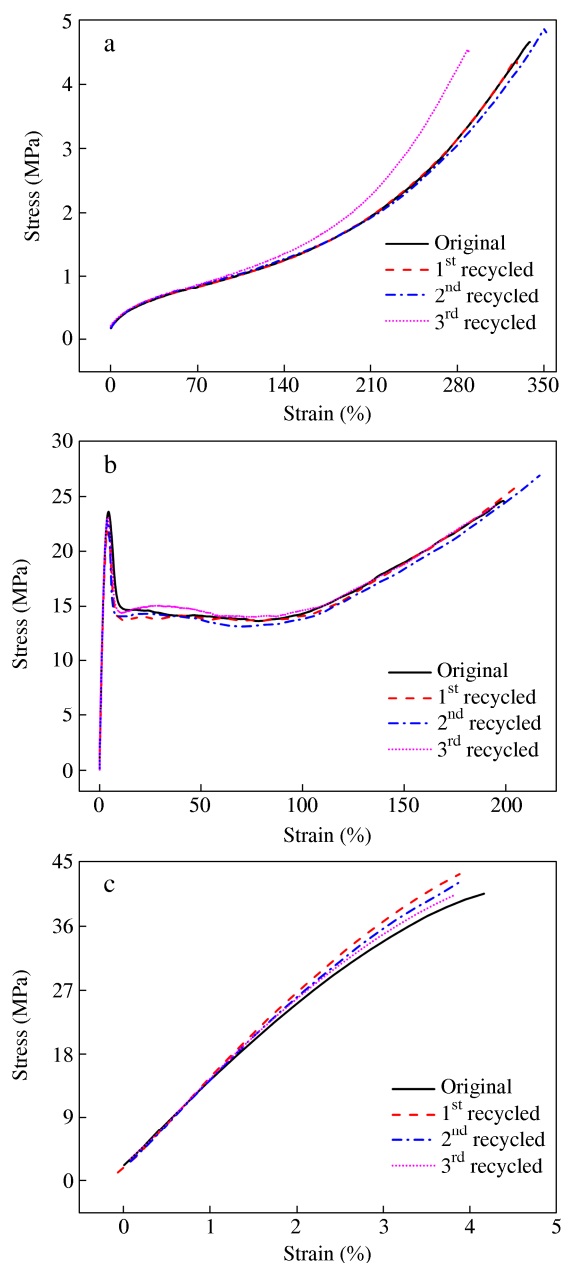


Fig. 8 Stress-strain curves of recycled samples (a) EVS, (b) EVC₂S₈, and (c) EVC₄S₆

without solid waste accumulation due to the easy reprocessability, which may promote the sustainable development of epoxy resins.

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