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Recyclable thermosets based on modified epoxy-amine network polymers†

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The development of high performance, recyclable thermoset materials for applications in plastics, composites, coatings and adhesives requires a synthetic approach where recyclability is designed into the molecular structure of the material. This paper describes a single stage process for the creation of materials from simple, low-cost molecular building blocks, where the polymerisation of liquid epoxy resins and aliphatic amines in the presence of an *n*-butyl diboronic ester, delivers epoxy-amine-dioxazaborocane materials with tunable physical properties including glass transition temperature (T_g). Mechanical (thermal) recycling and reprocessing of the epoxy-amine-dioxazaborocane thermoset is demonstrated, with retention of Young's modulus and ultimate tensile strength. Most notably, an efficient and low-cost process for the chemical recycling, disassembly and dissolution of the thermoset is demonstrated via two complementary processes using either pinacol (diol) or mono-functional phenylboronic ester.

New concepts

This paper describes a simple, single stage process for the creation of a recyclable, high performance thermoset material from readily available epoxy resins and aliphatic amines, crosslinked with diboronic esters. Our approach to recyclable thermoset (network) polymers is distinguished from others in this field in creating an opportunity to form high performance, recyclable materials from established molecular building blocks, common to the plastics, composites, coatings and adhesive industries. The epoxy-amine-dioxazaborocane thermoset materials can be synthesised via a single stage mixing and casting process, though exploitation of a scarcely reported latent cross-linking reaction. Most notably, an efficient and low-cost process for the chemical recycling, disassembly and dissolution of the thermoset material is demonstrated via two complementary synthetic processes, using either pinacol (diol) or mono-functional phenylboronic esters. We anticipate that this approach to designing high performance thermoset materials from simple, accessible building blocks, with recyclability built into the molecular architecture, will inspire other researchers working across the field of material science.

Introduction

High performance thermoset plastics and polymers deliver lightweight, durable and mechanically robust materials for a wide range of industrial applications.¹ These include high durability coatings,^{2,3} structural composites^{4,5} (some of which are used in the renewable energy sector), lightweight components for aerospace and automotive application, and structural adhesives.^{6,7} Whilst focus is dominated on the challenge of plastic recycling of commoditised (low-cost) thermoplastic materials used in bottles, packaging and films,⁸ the significant environmental and scientific challenge of recyclable thermoset materials has received proportionately less attention. Thermoset polymers, which make up ~20% of all polymeric materials,⁹ are

based on high density crosslinked polymer networks traditionally assembled using non-reversible covalent bonding. However, their covalent network structure renders them single-use, and non-recyclable in their current form. Epoxy-resins are one of the leading thermoset technologies for industrial applications, due to their excellent mechanical strength, versatility and robustness with over 323 000 tonnes of epoxy resin produced in Europe per annum.¹⁰

The increasing focus on sustainable development goals¹¹ and Net Zero Strategy,¹² (including sustainable and recyclable materials), highlights a major limitation of epoxy-resin technology, in the limited opportunity for chemical or mechanical recycling.^{13,14} This issue is highlighted in applications such as wind turbine blades, where it is estimated that ~250 000 tonnes of epoxy thermoset polymers reside, that are destined for landfill or incineration, once the wind turbines have reached the end of their useful life.¹⁰ Without lightweight, structural epoxy-thermosets renewable wind-energy would not

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MiniMIX-D columns in sequence, using a THF mobile phase and a flow rate of 0.6 mL min⁻¹. Analysis was performed against a calibration curve of polystyrene standards (EasiVial PS-M supplied by Agilent).

Dynamic mechanical thermal analysis (DMTA)

DMTA analyses were conducted for network polymers on a PerkinElmer DMA 8000 and analysed in Pyris software (version 11.1.1.0492). Temperature scans were performed on 30 mm × 5 mm × 0.3 mm samples and were analysed in tension geometry with a fixed length of 11.52 mm, a frequency of 1 Hz and a constant temperature ramp of 3 °C per minute from -80 °C to 200 °C. Storage modulus (E'), loss modulus (E'') and tan delta (tan δ) signals were recorded.

Tensile testing

Thin film polymer samples were laser cut to a modified ASTM standard 638-14-type V⁴⁰ and experiments were conducted on either an Instron 5969 or Instron 3343 with a displacement ramp rate of 10 mm per minute.

Hot press recycling

Mechanical recycling was conducted using a 2" × 3" Mini Manual Hot Press Machine (TTLIFE) with LCD Controller at 700–1000 lbs pressure. Ground polymer samples were placed between PTFE sheets before pressing at 170 °C for 30 minutes, then allowed to cool to room temperature (~25 °C) overnight. Samples were then removed from the press and prepared for testing.

Results and discussion

The preparation of epoxy-amine-dioxazaborocane thermosets requires controlled sequential epoxy-amine polymerisation, followed by dioxazaborocane cross-linking. To achieve a single stage synthesis process suitable for film casting or resin infusion processes, multifunctional boronic esters must be designed and synthesised for optimal miscibility with epoxy resins, whilst maintaining a balance of storage stability and chemical reactivity to allow latent cross-linking of the developing epoxy-amine polymeric network. Previous work in our group^{24–26} reported latent reactivity in epoxy-amine-borate (EAB) complexation which could potentially be exploited in the formation of a thermoset/network polymer. In the previous work, epoxy-amine polymerisation resulted in the formation of β -amino diol functional groups, which underwent latent complexation with borate esters. It was hypothesised that this chemo-selective and latent reactive sequence could be applied to the 'one-pot' synthesis of epoxy-amine polymers, with latent crosslinking through diboronic esters, to form epoxy-amine-dioxazaborocane thermosets. Since dioxazaborocane functional groups are known to undergo reversible bond exchange with adjacent β -amino diol functionalities,⁴¹ this presented an attractive potential route for the preparation of high-performance dynamic thermosets, from low-cost commodity

starting materials, such as epoxy resins and aliphatic amines. To explore this concept, the preparation of suitable boronic esters that combined miscibility (in general purpose epoxy resins), stability and chemical reactivity (with a β -amino diol) was required. A range of mono-boronic esters (**1a–d**) were prepared from readily available starting materials (Fig. 2)⁴² and their physical and chemical reactivity properties were studied.

To explore the relative reactivity of the selected boronic esters with a β -amino diol functional group, the model β -amino diol **3** was prepared²⁴ and reacted (1:1 molar ratio) with the series of mono-boronic esters (Fig. 2, **1a–d**) in CDCl₃ (5 minutes at 25 °C). Direct analysis of the equilibrium reaction mixtures was conducted using ¹H-NMR. The reaction of β -amino diol **3** with cyclic boronic ester **1a** and **1b** did not deliver a high reaction yield of the desired dioxazaborocane **4**, even after extended reaction times. The non-substituted cyclic ethane-1,2-diol derived boronic ester (**1a**) achieved partial conversion to dioxazaborocane **4** (**1a**:**4** ratio, 27:73, Fig. 1, entry **1a**) whilst the more hindered 2,3-dimethyl-2,3-butanediol (pinacol) based ester (**1b**)⁴³ achieved a poor conversion of 78:12 (**1b**:**4** ratio) after 96 hours (Fig. 1, entry **1b**, comparison of electron-rich aromatic signal at 7.7 ppm (**4**) vs. 7.81 ppm (**1a/b**)). Equally, cyclic boronic ester **1a** and **1b** were found to be crystalline solids with relatively poor solubility in general purpose epoxy-resins such as diglycidylether of bisphenol-A (DGEBA, DER-331) and butane-1,4-diglycidyl ether. The linear (non-cyclic) esters **1c** (iso-propyl) and **1d** (*n*-butyl) in-contrast, showed quantitative conversion to the desired dioxazaborocane **4**, indicating the required reactivity for the proposed 'one-pot' latent network assembly. They were also found to be liquids and miscible with general purpose epoxy-resins. Based on this study, the *n*-butyl diboronic ester **2d** was selected for further studies, and it was found to deliver the ideal combination of

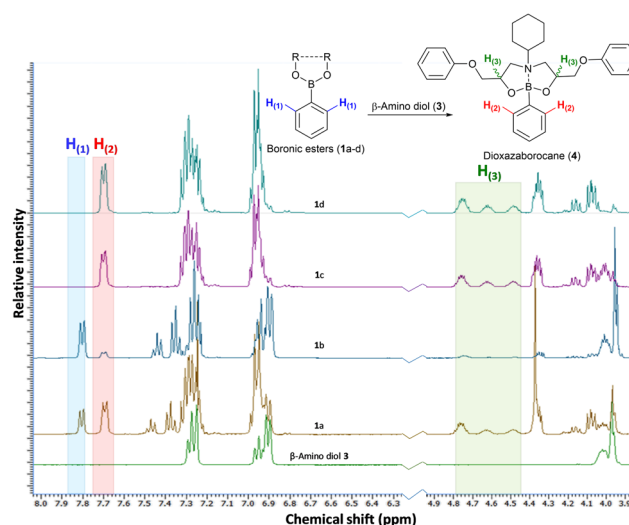


Fig. 1 ¹H-NMR analysis of the reaction mixtures of boronic esters **1a–d** with model β -amino diol **3**. Hydrogen environments of interest are indicated as according to the colours H-atom (H^1 , H^2 and H^3) labelled on both the molecule structures and spectra.



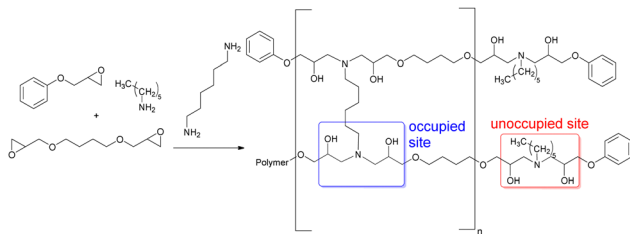


Fig. 4 The synthesis of epoxy-amine (A series) 'non-dynamic' materials with structural synergy to epoxy-amine-dioxazaborocane (D series) materials.

DMTA analysis supported the T_g trends observed by DSC, with apparent higher T_g 's for the D60–D100 vs. the A60–A100 series, as characterised by either T_{gonset} (sharp decline in E' , Fig. 5a) or peak of $\tan \delta$ (Fig. 5c).⁴⁶ Both the storage and loss modulus curves for the epoxy-amine-dioxazaborocane (D60–D100) materials show additional transitions after T_{gonset} , in comparison to the A60–A100 epoxy-amine materials (Fig. 5a and b, non-smooth decline in E' and E''). Given the well-established exchange reaction of boronic esters with diols and amino-diols^{18,38,41} it is proposed that these transitions are related to rapid bond exchange reactions and related rapid topological rearrangements, between 'occupied' (dioxazaborocane) and 'unoccupied' (β -amino diol) groups, enabled by the transition of the materials from the glassy to rubbery material state. It has been suggested that, when the topological freezing temperatures (T_v) is significantly lower than T_g , facile topological rearrangements would be expected at $T > T_g$.³³ In the case of the dioxazaborocane materials described here, as temperatures progressed beyond T_{gonset} the D-series materials softened and at $T_g + 30$ °C demonstrated flow behaviour, indicative of a material with potential for mechanical recycling (*vide infra*). This behaviour also resulted in a significant broadening of the $\tan \delta$ in the case of D60–D100 materials, accounting for the apparent large increase in T_g value when compared to the corresponding DSC result (Table 1). Equivalent post- T_g transitions are not present in the control epoxy-amine materials (A60–A100), presumably due to the absence of dynamic bond exchange reactions in these 'non-reversible' covalently cross-linked materials.

To address the significant difference in T_g between the D-series and A-series materials (which was considered important for comparison of material properties), higher T_g epoxy-amine thermoset materials were prepared as control materials. The A²-series were based on 4,4'-methylenedicyclohexanamine (4,4'-MDH) as the crosslinking agent, replacing the more flexible 1,6-diaminohexane (DAH). The bicycloaliphatic structure of 4,4'-MDH (see ESI[†]) resulted in an increase in material T_g (Table 1, A²20–A²100), reducing the ΔT_g between D100 and A²100 to ~ 8 °C. The A³ series utilised 4,4'-MDH (crosslinking) and DER-332 (bisphenol-A epoxide from Olin Epoxy) with cyclohexylamine for the epoxy-amine polymerisation to deliver materials with T_g 's in the region of 87.8–97.1 °C (Table 1, A³60–A³100).

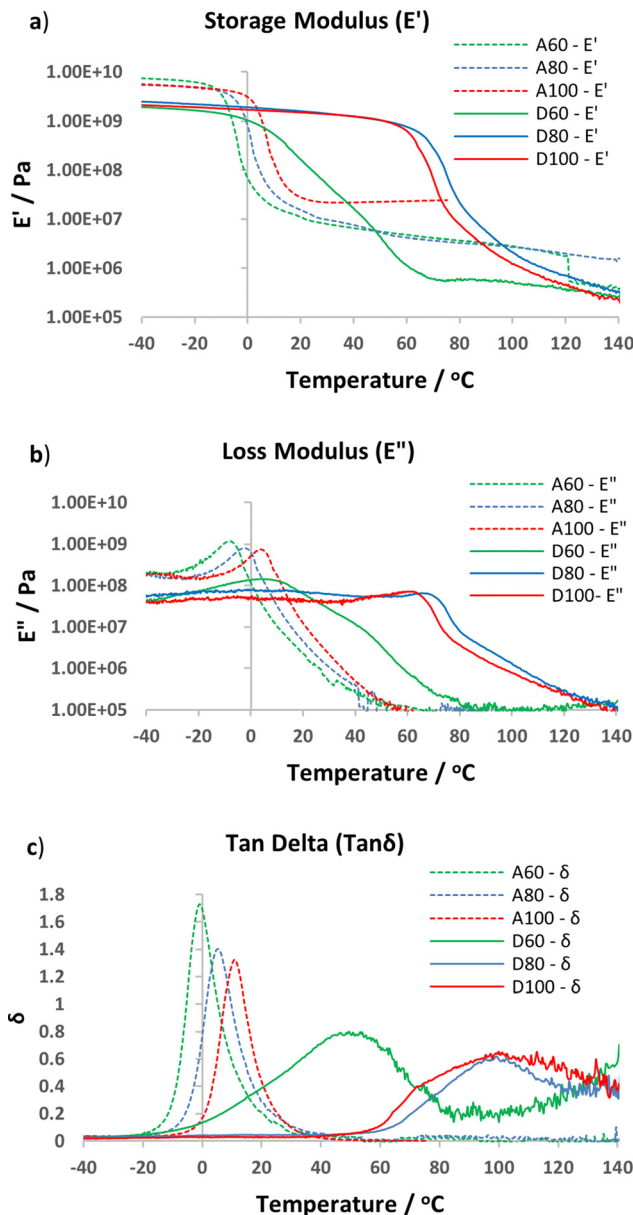


Fig. 5 DMTA analysis of D60–D100 and A60–A100 materials. (a) overlay of storage modulus curves (E'), (b) overlay of loss modulus curves (E'') and (c) overlay of $\tan \delta$ curves (E''/E').

Characterisation of the D20–D100 series and A²20–A²100 by infrared spectroscopy delivered insight into the structural difference between the dynamic and non-dynamic materials (Fig. 6). Increasing the dioxazaborocane content (*i.e.*, increasing %-occupied vs. %-unoccupied in D0–D100) results in a proportional decrease in OH concentration (Fig. 6a, indicated region). This can be assigned to conversion of β -amino diol (OH) groups into dioxazaborocane complexes (see Fig. 3 for illustration), consuming the OH functionality proportional to the degree of crosslinking. This trend is absent in the infrared spectra of the epoxy-amine series (A²0–A²100), where cross-linking does not consume hydroxyl groups, thus OH concentration should be approximately equal in all examples (Fig. 6b, indicated region).





Fig. 6 FTIR overlays of (a) epoxy-amine-dioxazaborocane materials (D0–D100, inset: representative molecular structure of D0 and D100) and (b) epoxy-amine thermoset materials (A²20–A²100, inset: representative molecular structure).

Chemical resistance and recycling

A chemical recycling process that allows selective disassembly and dissolution of epoxy-amine-dioxazaborocane could allow separation and purification of the molecular building blocks, ready for reuse. However, it is essential that high-performance epoxy-thermosets exhibit a broad spectrum chemical resistance, remaining undamaged and insoluble upon exposure to chemically aggressive liquids and solutions.^{47–49} Immersion of the epoxy-amine-dioxazaborocane D100, in a range of solvents including ethanol, DMSO, toluene, acetone, THF, distilled water and 1 M NaOH showed no material integrity loss with the expected excellent chemical resistance and insoluble properties of a ‘traditional/epoxy-amine’ thermoset, network polymer. The hydrolytic stability under aqueous conditions, can be attributed to the enhanced stability of boronic esters containing tetra-coordinate bonding *via* N-atom coordination,^{50,51} such as that seen in the bicyclic dioxazaborocane structures described here. The solvent resistance can be clearly seen in Fig. 9a (THF control), where both D100 and A100 remain unchanged and undissolved after 24 hours. Polymer-network disassembly was demonstrated with the dioxazaborocane D-series, using (mono-functional) *n*-butyl phenylboronic ester **1d** in THF. Using an excess of **1d**, the D100 polymer-network can be disassembled to form a soluble (non-crosslinked) epoxy-amine polymer as illustrated in Fig. 7. This is demonstrated visually in Fig. 9b (THF + **1d**) where, in contrast to THF only

(Fig. 9a), the D100 sample has fully dissolved after 24 hours. In all cases, the control epoxy-amine A100 remains undissolved, as expected for a ‘traditional’ epoxy thermoset. The same result was achieved using pinacol (2,3-dimethyl-2,3-butanediol) to disassemble the dioxazaborocane D100 polymer network, liberating the epoxy-amine polymer, with concomitant formation of di-pinacol boronic ester **2b** (Fig. 7, method b). ¹H-NMR analyses support this, with characteristic dioxazaborocane signals (Fig. 8, boronic ester model complex, 4.2–4.8 ppm) absent from the product ¹H-NMR spectra (Fig. 8, D100 + pinacol) indicating release of the non-complexed, linear epoxy-amine polymer (equivalent to polymer D0). The dipinacol ester of phenyl diboronic acid (**2b**) has a peak also clearly present in the product ¹H-NMR spectra (Ar-H at 7.8 ppm), further supporting our understanding of the chemical disassembly process.

GPC analysis of the chemical recycling solutions supported the results of the ¹H-NMR analysis, through comparison of molecular weight distributions from both disassembly methods (Fig. 7, method (a) and (b)) *vs.* authentic prepared samples of linear epoxy-amine polymer (*i.e.*, D100 without dioxazaborocane crosslinking = D0). The polymer molecular weight distributions of the two chemical disassembly methods (Fig. 10, ‘D100 post THF/**1d** immersion’ and D100 post THF/pinacol immersion’) show excellent correlation with the parent control polymers (Fig. 10, ‘D0 control’ and ‘D0 control with **1d**). In comparison, immersion of the epoxy-amine-dioxazaborocane in THF only (Fig. 10, D100 post THF immersion) did not result in dissolution of high molecular weight polymeric material, further evidencing the chemoselectivity of the disassembly process. These experiments not only demonstrate a viable mechanism for recovery of molecular building blocks from epoxy-amine-dioxazaborocane crosslinked polymer networks, but also provided evidence for the molecular structure of the epoxy-amine polymeric component within the epoxy-amine-dioxazaborocane crosslinked network.

Mechanical recycling

A covalent adaptable network, such as proposed with materials D60–D100, that undergoes rapid bond and topological rearrangement at $T > T_{\text{gonset}}$ would be an attractive material for mechanical recycling. Epoxy-amine-dioxazaborocane materials D60–D100 were ground into small particles, then heated under compression at 170 °C for 30 minutes, using a simple bench-top hot press, before slowly cooling overnight (see ESI† for details). In all cases this resulted in the formation of a cohesive sheet of polymeric material, where tensile testing of the mechanically recycled materials (D^R80–D^R100) *vs.* the parent materials (D80–D100), showed similar stress-strain curves (Fig. 12), across both D80 and D100 materials.

Key physical parameters such as Young’s modulus and ultimate tensile strength were statistically unchanged in the higher crosslink density D80 and D100 materials. Both series of materials measured Young’s moduli of 96–119 MPa for the D80/D^R80 examples, increasing to 128–136 MPa for the higher crosslinked D100/D^R100 example (Fig. 11b, entries 2 and 3), demonstrating the ability to both control material properties



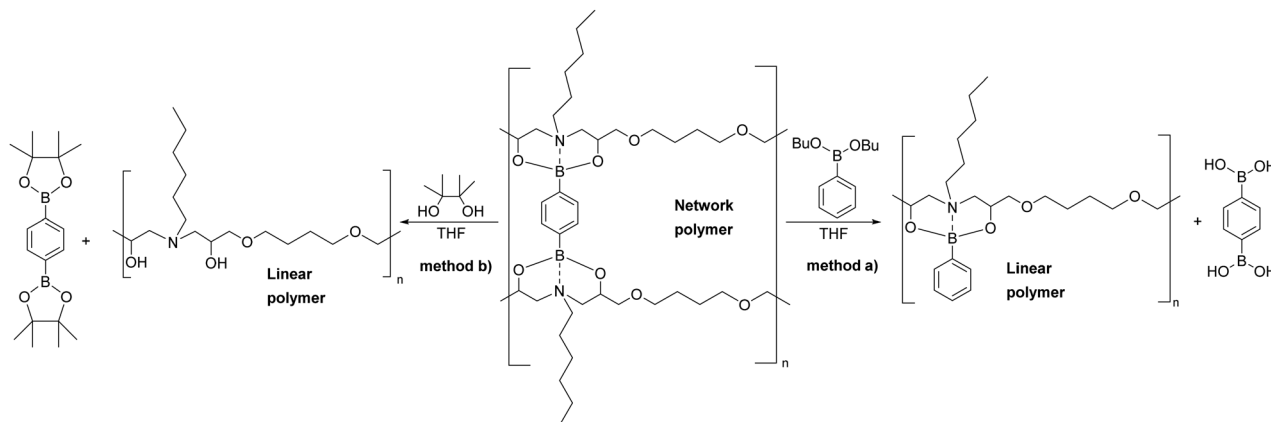


Fig. 7 Chemical recycling of epoxy-amine-dioxazaborocane D100 via either (a) phenylboronic ester or (b) 2,3-dimethyl-2,3-butanediol (pinacol) methodologies.



Fig. 8 $^1\text{H-NMR}$ spectra of epoxy-amine-dioxazaborocane thermoset D100 after chemical recycling via pinacol methodology (according to Fig. 7b).

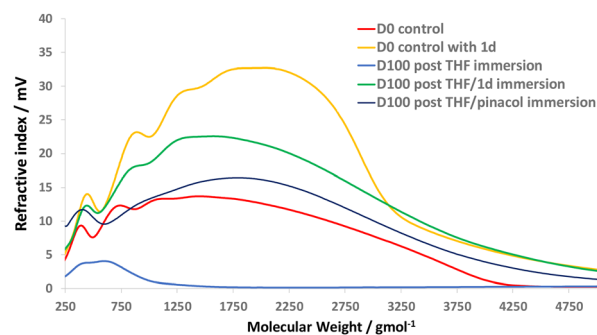


Fig. 10 GPC analysis of the solutions from **1d** and pinacol chemical recycling methodologies (according to Fig. 7) vs. D0 controls (with and without **1d**).

selected for analysis. Whilst the D-series materials were lower in Young's modulus than the corresponding A³-series (309–321 MPa), these results demonstrate significant promise for this new material class. Similar trends were observed for the ultimate tensile strength data of D^R100 (Fig. 11a) with the pre- and post-recycling results for D80/D^R80 and D100/D^R100 statistically indistinguishable from each other (30–38 MPa). Again, whilst the A³ control demonstrated higher strength (68–69 MPa), the results for the epoxy-amine-dioxazaborocane were very promising. These results support the hypothesis that the dioxazaborocane bond exchange results in sufficient rheological flow for material fragments to reform into a single cohesive polymer sheet, and that bond interchange restores the polymer network and associated mechanical properties.

Extended evaluation to study the impact of repeated mechanical recycling on the thermal, chemical and mechanical properties of dioxazaborocane material D80 was conducted. A virgin sample of D80 was prepared and subjected to three cycles of mechanical recycling, with characterization by infrared spectroscopy, GPC (after chemical disassembly in THF/pinacol), tensile testing and thermal analysis (T_g by DSC) at each stage of recycling (plus characterisation of the virgin D80).

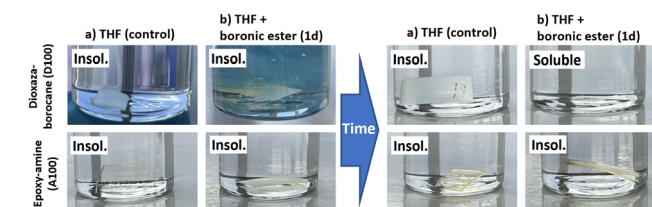


Fig. 9 Chemical recycling of D100, using *n*-butyl phenylboronic ester **1d**. (a) immersion of epoxy-amine-dioxazaborocane D100 and epoxy-amine A100 in THF at the start of the experiment and after 24 hours. (b) shows immersion of D100 and A100 in THF with *n*-butyl phenylboronic ester **1d** at the start of the experiment and after 24 hours.

and maintain such properties post-recycling. For comparison to a 'traditional' epoxy-amine control, the high T_g A³ series was



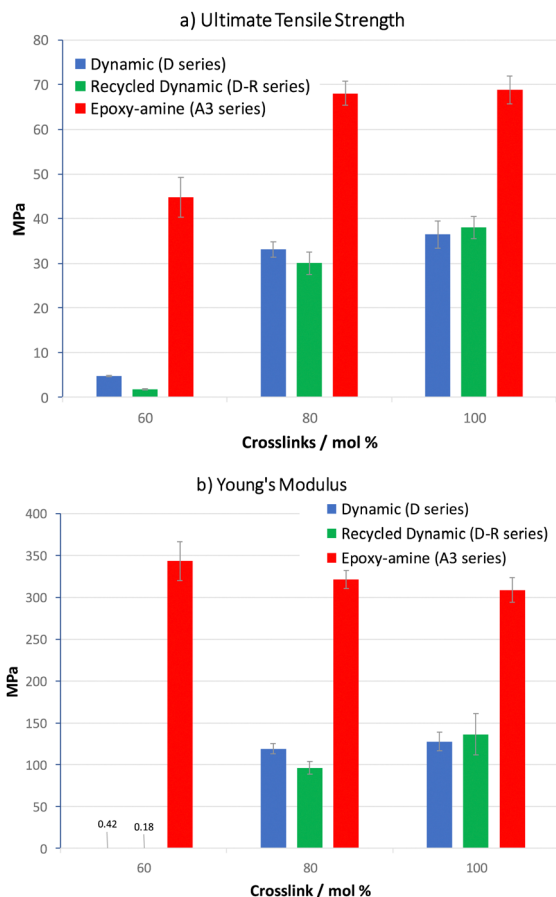


Fig. 11 (a) Ultimate tensile strength and (b) Young's modulus, data for D60–D100 and A³60–A³100 materials.

Analysis by infra-red spectroscopy revealed low levels of apparent oxidation upon subjecting the D80 material to repeated



Fig. 12 Stress over strain curves for D80/D^R80 and D100/D^R100 (pre and post mechanical recycling). Inset: Image of D100 before and after mechanical recycling, by hot press and laser cutting into dog bones for tensile testing.

mechanical recycling at elevated temperatures (170 °C). This is characterised by the presence of weak absorption bands at 1658 cm⁻¹, associated with amine oxidation and at 1738 cm⁻¹, characteristic of epoxy resin oxidation (Fig. 13a, highlighted region).⁵² To examine the impact of mechanical recycling on the molecular weight distribution of the dioxazaborocane D80 material, the same samples were subjected to chemical disassembly in THF/pinacol according to our experimental procedure (see previously described method) and analyzed by GPC. A slight reduction in the molecular weight distribution profile was observed upon mechanical recycling, proportional to the number of recycling processes to which the material was subjected. Since oxidation of polymers is a well-recognized route to degradation (including molecular weight reduction),^{52,53} it seems reasonable to assume that these properties are connected, and that the apparent oxidation is responsible for the modest reduction in the molecular weight distribution by GPC (Fig. 13b). The tensile properties of the

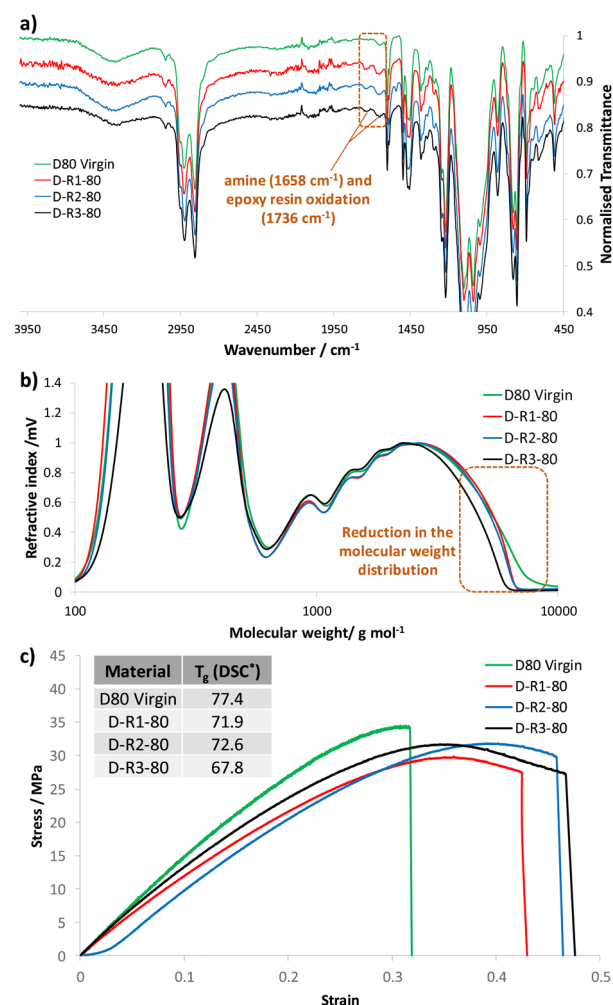


Fig. 13 Characterisation of dioxazaborocane D80 before mechanical recycling (virgin), and after one (D-R1-80), two (D-R2-80) and three (D-R3-80) recycling processes (at 170 °C) by (a) Infrared spectroscopy, (b) GPC after chemical recycling in THF/pinacol and (c) tensile testing with T_g by DSC (inset, *run at 100 °C min⁻¹).



dioxazaborocane D80 materials were generally maintained on repeated mechanical recycling (Fig. 13c). The recycled samples (R1, R2 and R3) all demonstrated similar stress-strain curves, Young's moduli and ultimate tensile strength compared to the D80 virgin sample (Fig. 13c). In addition to this, the recycled materials (R1–R3) all showed improved ductile behavior in comparison to the virgin material (D80), likely linked to the slow cooling of the recycled materials in the mechanical hot-press, allowing annealing of the polymeric networks. Finally, thermal analysis of the 4 samples by DSC, revealed a slight depression in T_g after repeated mechanical recycling (Fig. 13c, inset), again likely linked to the reduction in molecular weight distribution observed by GPC^{54,55} and associated polymer oxidation.⁵⁶

Conclusions

A new methodology for the synthesis of recyclable thermoset polymers has been described, based on epoxy-amine building blocks which are common to the coatings, adhesives, and composites industries. Practical limitations associated with miscibility and reactivity of boronic esters (used in crosslinking) has been overcome to allow the preparation of epoxy-amine-dioxazaborocane, covalent adaptive networks, via a single stage film casting process, which also demonstrated the inherent latent reactivity of dioxazaborocane crosslinking. Comparative 'traditional' epoxy-amine thermoset materials were also prepared and the physical, mechanical and recyclable properties studied, alongside the epoxy-amine-dioxazaborocane materials.

Two complementary synthetic methodologies for the subsequent chemical disassembly and dissolution of epoxy-amine-dioxazaborocane thermosets has been demonstrated. This process uses simple and readily available reagents (pinacol or phenylboronic ester) and highlights a potential route to recycling, separation and recovery of molecular building blocks. The dynamic nature of epoxy-amine-dioxazaborocane thermosets allows for facile topological rearrangement and flow when heated above T_g , delivering an effective mechanism for mechanical reprocessing or recycling of the materials. This recycling process has been shown to retain important material properties including Young's modulus and ultimate tensile strength. Overall, this work highlights the opportunity to create high performance thermoset materials for industrial applications, with mechanical and chemical recyclability designed into the molecular architecture.

Conflicts of interest

There are no conflicts to declare.

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