

Solvent-Base Mismatch Enabling the Deconstruction of Thermoset Epoxy Polymers into Base Chemicals

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Abstract: Fiber-reinforced epoxy composites are key materials for the construction of wind turbine blades and airplanes due to their remarkable properties. On the flipside, their physical and chemical inertness also results in a lack of viable recycling technologies. Recently, tailored resins have been introduced, which allow controlled fragmentation of the polymer matrix and thus the recovery of embedded fibres. However, for the separated thermoset epoxy fragments there is no recycling solution available, resulting in loss of molecular complexity. Here we report a chemical process for recovering bisphenol A (BPA) from epoxy resins, using a mismatched base-solvent system at an elevated temperature. We demonstrate a combinatory disassembly processes / chemical deconstruction strategy on a commercial tailored composite sample, isolating both fibres and the polymer building block. The recovered BPA could be reused in established polymer production chains, thus closing the recycling loop and reducing the need for virgin resources.

Thermoset epoxy resins are a class of polymers that are cured on-site by blending short-chained epoxy resins with a curing agent, resulting in crosslinked molecular networks dominated by σ bond linkages and aromatic backbones. The liquid resins are commonly centred on bisphenol A (BPA) and prepared by reacting its phenol hydroxyl groups with epichlorohydrin forming strong C(alkyl)–O bonds. Multifunctional alkyl amine mixtures are widespread curing agents, and in turn create C(alkyl)–N linkages through nucleophilic substitution with the epoxide electrophiles during the hardening process.¹ Alternatively, carboxylic acid anhydrides can be used to cure epoxides, forming carboxylate ester networks (Fig. 1a).¹ Besides coatings and laminates, fibre-reinforced epoxy composites are a major application for these resins.² For the production of composites, glass or carbon fibres are embedded in a thermoset polymer matrix, resulting in favourable mechanical properties, which allow appliance as high performance materials. These composites have become crucial for the production of wind turbine blades, as well as for uses in the automotive, aeronautical and maritime industries.^{2,3}

Wind energy contributes to about 6% of the global energy supply in 2020. Due to the environmental ramification of burning fossil fuels, as well as geopolitical concerns obstructing the procurement of such fuels, wind energy is widely regarded as instrumental for securing our future energy supply.⁴ Nevertheless, recycling solutions for end-of-use composite materials are critically underdeveloped,^{2,5} and in turn the overwhelming majority of wind turbine blades are landfilled after their lifetime. Projections estimate that 43 million tons of decommissioned blades will have accumulated by 2050,⁶ with each one containing 80 to 90% composite material by weight.⁴ Landfilling of turbine blades is undesirable as their size renders

this manner of disposal highly volume inefficient. Therefore, landfilling as a means of disposal has been banned in several European countries, with more being expected to follow latest by 2025.⁴ However, implementing such bans can only be expedient if sustainable alternatives are available. Related industries are looking to develop recycling solutions for wind energy systems,^{4,7,8} however, processing these highly durable thermoset composites is notoriously challenging.²

In the last decades, attempts to valorise end-of-use composites have been focused on recovering fibres *via* harsh treatments that destroy the polymer matrices. *E.g.* pyrolysis has been shown to be efficient in removing the epoxy resins, however such processes are energy intensive and result in the recovery of fibres of lowered quality.^{2,5,9} An alternative disassembly concept for composite materials is based on tailored resins containing chemically labile functionalities, which can be cleaved under milder conditions, disassembling the polymer matrices into shorter, but still crosslinked polymer fragments.^{2,10,11} *E.g.* the incorporation of ester moieties allows selective cleavage *via* thermal¹² or basic¹³ treatment. Alternatively, epoxy resins can be designed to effectively fracture through physical processes upon exposure to certain triggers, such as acetic acid.^{14,15} In 2021, the wind turbine producer Siemens Gamesa commercialised turbine blades based on a resin system that allows clean separation of the fibre mesh from the resin using acetic acid,¹⁶ thanks to a ketal containing diamine (recyclamine) in the hardener.^{17,18} While this is an elegant way to recover fibres from composites, the reuse of the separated epoxy fraction is limited to filler material at best,¹⁹⁻²⁴ meaning that these novel resin formulations fall short of introducing a circular economy for these important structures.

Results

Method Development

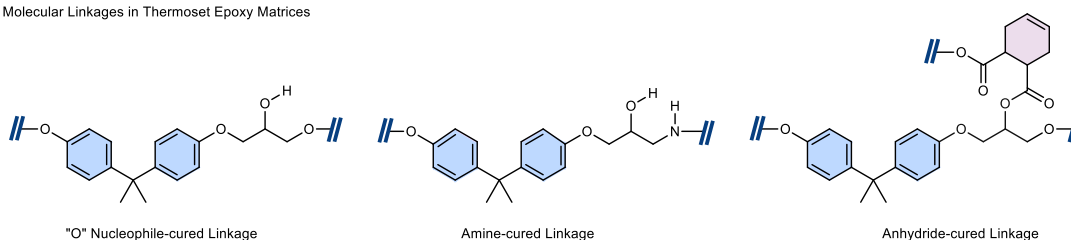
Recently, our group reported a selective disconnecting C–O bonds in the polymer matrix, allowing the recovery of chemical building blocks and high quality fibres directly from legacy materials.²⁵ While performing the chemical deconstruction directly on the composite is desirable, the current catalytic system on the solid-liquid interface of composite samples requires reaction times in the order of days. Therefore, we were interested in investigating alternative approaches for deconstructing relevant epoxy composites. In this work, we report on the development of a robust and simple protocol for cleaving the molecular linkage motifs in the epoxy resins after they have been separated from the fibres. Using a reaction system consisting of an alkaline base mismatched with an unpolar organic solvent at elevated temperatures, the important polymer building block bisphenol A (BPA)^{26,27} can be recovered from different epoxy samples. With both the recovery of fibres and polymer building blocks through a combinatory disassembly processes / chemical deconstruction strategy, a circular economy^{28,29} for composite-based structures such as wind turbine blades would be achievable (Fig 1b).

Reported protocols for separating fibres from the resins rely on acidic or basic treatments, and therefore identifying a methodology tolerating acid or base residues in the epoxy fraction is a critical success criterium. Recently, a method disconnecting C–C bonds in epoxy polymers has been reported.^{30,31} Although mild and elegant in its nature, a subsequent ether cleavage using BBr₃ is necessary in order to release the polymer building block BPA. We decided to target the C(alkyl)–O bonds formed during the reaction of BPA with epichlorohydrin, as cleaving these would directly liberate BPA (Fig.1b). Computational studies on epoxy linkages revealed that the bond dissociation energy of α -phenoxyketones is 10.4 kcal/mol lower than for the corresponding α -phenoxyalcohols.²⁵ During our studies on epoxy resin disconnections, we discovered that bases are competent at efficiently liberating phenols from dehydrogenated

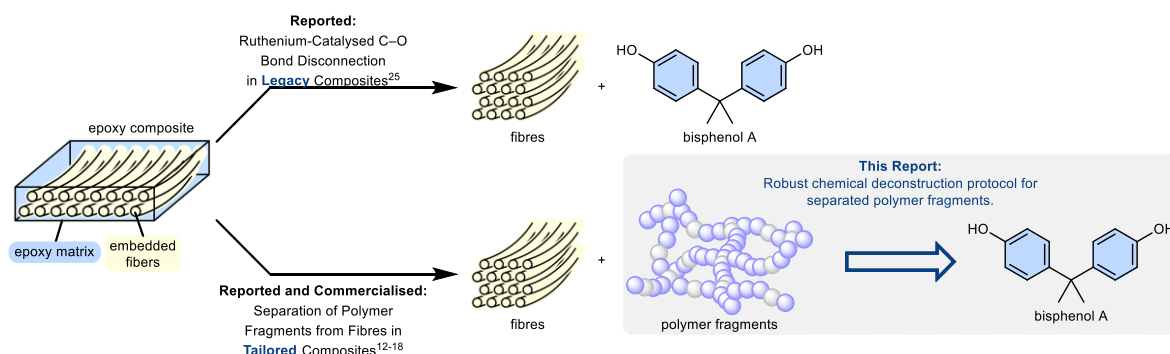
epoxy models, such as ketone **1**, which presents the oxidised form of the linkage formed from BPA and epichlorohydrin. Based on this discovery, we envisioned applying a robust alcohol oxidation process with stoichiometric amounts of base with respect to the epoxy resins (Fig. 1c). We recognised that processes that require base would neutralise residual acid in an epoxy fraction recovered from composite materials, thus avoiding extensive drying processes after the separation from the fibres.

Due to its atom efficiency, we proceeded with a homogeneously catalysed acceptorless dehydrogenation^{32,33} for the first step. Several protocols rely on the use of stoichiometric amounts of bases, we identified as potent for cleaving the C–O bond in ketone **1**.³⁴⁻³⁷ Stoichiometric or excess amounts of base have been shown to inhibit deactivations of the catalysts.³⁷

a Molecular Linkages in Thermoset Epoxy Matrices



b Reported Chemical Recycling Concepts for Fiber-Reinforced Epoxy Composites



c Envisioned Approach to Chemical C–O Bond Cleavage in Epoxy Fragments

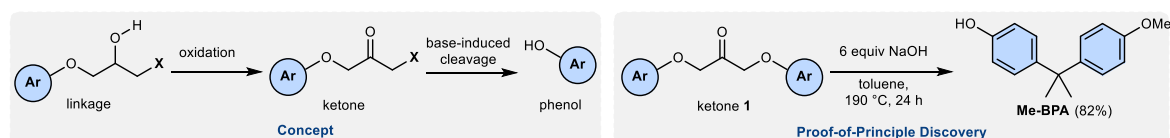
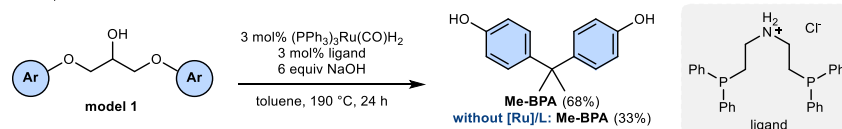


Fig. 1 | Concept for deconstruction of epoxy resins recovered after separation from fibres. **a**, Schematic illustration of a FREC and its reported separation into a fibre and epoxy fraction and the here reported deconstruction of epoxy resins into BPA. **b**, Linear linkage motifs in amine-cured epoxy resins. **c**, Project idea for a chemical deconstruction of epoxy resins.

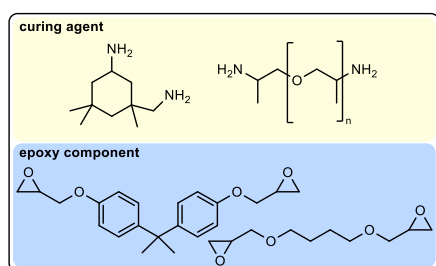
Beller *et al.* reported a highly efficient dehydrogenation of isopropanol using an *in situ* formed Ru-MACHO complex in the presence of stoichiometric amounts of NaOH/Pr.³⁷ The dehydrogenation in tandem with the base-induced C–O bond cleavage were optimised on **model 1**. In accordance with the conditions reported by Beller *et al.*³⁷, 3 mol% of (Ph₃P)₃Ru(CO)H₂ together with 3 mol% of bis[(2-diphenylphosphino)ethyl]ammonium chloride were chosen as the catalytic system (Fig. 2a, see SI for optimisation). To our delight, it was possible to use 6 equiv of powdered NaOH in the presence of the ruthenium catalyst at 190 °C to deconstruct the model compound over the course of 24 h, affording **Me-BPA** in a yield of 68% (Fig. 2a). While using stoichiometric reagents in excess generally can be considered a

flaw regarding atom economy³⁸ and the concept of green chemistry,³⁹ it has to be considered that NaOH is a commodity chemical that is exploited in several industrial processes in large quantities and is formed as a by-product during the production of chlorine.⁴⁰ To our surprise, a control experiment conducted in absence of the catalyst system afforded 34% of **Me-BPA** starting from **model 1**.

a Tandem deconstruction developed on **model 1**



b Components of Airstone 760E/766H



c Temperature resolved comparison of Tandem Reaction and Base Only Process

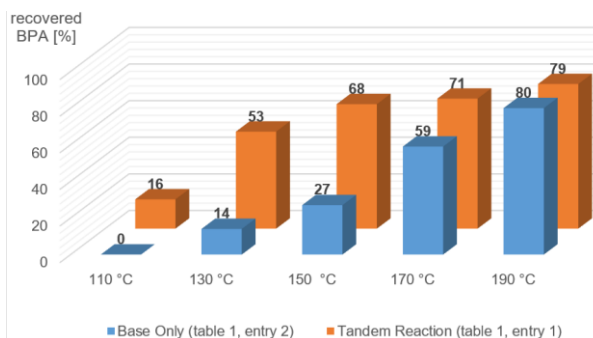


Fig. 2 | Method development and application to thermoset epoxy resin. a, Method for C–O bond cleavage developed on **model 1**. **b**, Formulation of the epoxy resin used as benchmark system. **c**, Comparison of the deconstruction of Airstone through the tandem reaction using a ruthenium catalyst and base with the base only protocol across different temperatures. All yields are given for isolated BPA. Reaction conditions are based on the those described in Table 1, entries 1 and 2, respectively.

With a proof-of-principle of the tandem disconnection process at hand, we moved onto the testing of the reaction conditions on commercial thermoset epoxy resins. Airstone 760E/766H is an amine-cured resin system developed and marketed for the construction of wind turbine blades. It consists of bisphenol A diglycidyl ether and ethylene glycol diglycidyl ether as the liquid epoxy resin and isophorone diamine and poly(oxypropylene) diamine as the curing agents (Fig. 2b). The cured resin contains approximately 43 wt% of BPA, and isolated yields were calculated based on this value. As such crosslinked resins are not soluble, and chemical reactions are limited to the surface of the material. Therefore, a block of the cured resin was grinded into a powder. 100 mg of the powdered resin were suspended in toluene under argon in a pressure tube, and 50 wt% of powdered reagent grade NaOH, as well as 6 wt% precatalyst and ligand each were added. The 50 wt% of NaOH roughly equates 6 equiv of base for each BPA bound in the polymer. The reaction was stirred in a closed system at 190 °C for 24 h (Table 1). To our delight, using the catalyst system and powdered sodium hydroxide allowed for the recovery of 79% of the BPA from Airstone (entry 1). Surprisingly, the yield is significantly higher than that for **model 1**. Due to this discrepancy, we were interested in examining the base-mediated background reaction that was observed in the absence of catalyst on the polymeric sample. Astonishingly, without the ruthenium complex and ligand, 80% of BPA was recovered from the resin (entry 2). Next, the reaction was repeated under air using non-dried solvent (309 ppm water content) resulting the same yield (entry 3).

Table 1

Airstone 760//766H \longrightarrow BPA

Reaction conditions
 6 wt% (PPh₃)₃Ru(CO)H₂
 3 wt% ligand, 50 wt% NaOH
 toluene, 190 °C, 24 h, 300 rpm

Entry	Recovered BPA
1	no variation 79% (34.1 mg)
2	no [Ru]; no ligand 80% (34.5 mg)
3	no [Ru]; no ligand; no inert gas technique ^[a] 81% (34.7 mg)

[a] Reaction set up under air, non-dried toluene used as solvent.

Table 2

Airstone 760//766H \longrightarrow BPA

Reaction conditions
 50 wt% NaOH
 toluene, 190 °C, 24 h, 300 rpm

Entry	Recovered BPA
1	no variation 81% (34.7 mg)
2	no NaOH 0%
3	25 wt% NaOH 33% (14.3 mg)
4	KOH instead of NaOH 61% (26.4 mg)
5	<i>o</i> -xylene instead of toluene 75% (32.1 mg)
6	<i>p</i> -cymene instead of toluene 75% (32.5 mg)
7	DMF instead of toluene traces
8	water instead of toluene 0%
9	5 vol% water 0%
10	2 d reaction time 0%
11	12 vol% MeOH, 170 °C 45% (19.1 mg)
12	10 mol% TBAC, 170 °C 45% (19.3 mg)
13	10 mol% 15-crown-5, 170 °C 55% (23.5 mg)

Table 1. Tandem reaction and background reaction on polymeric sample. **Table 2.** Optimisation and Control experiments for the base-induced C–O bond cleavage.

At this point we assumed that two reaction pathways are simultaneously deconstructing Airstone: a base only process, as well as a ruthenium-catalysed tandem reaction as we had envisioned originally. In order to test this hypothesis, we compared the influence of the catalyst across different temperatures. The conditions described in Table 1, entries 1 and 2 were chosen as benchmark (Fig. 2c). In the absence of catalyst (blue), the reaction is highly efficient at 190 °C with 80% of BPA being recovered. However, already at 170 °C, the yield plummets to 59%. The trend continues almost linearly, with no free BPA being detected after 24 h at 110 °C. On the other hand, in the presence of a ruthenium catalyst (orange), the amounts of liberated BPA also decrease slightly as the temperature is reduced. However, even at 150 °C, 68% of BPA could still be isolated. Below 130 °C, the yields drop significantly with 16% of BPA isolated at 110 °C. This shows that both reactivities are viable, with the tandem approach allowing for a reduction of the reaction temperature. However, in light of the simplicity of a catalyst free protocol for the deconstruction of epoxy resins, we continued this study on the base promoted method.

Despite the high temperature and the relatively high base loading, this catalyst-free protocol is simple and robust, and therefore attractive for large scale applications. Due to the relatively high pressure that toluene will build up at 190 °C, similar solvents with higher boiling points were considered (table 2). Both *o*-xylol (bp 144 °C) and *p*-cymene (bp 177 °C) gave comparable yields of 75% of recovered BPA each after 24 h (entries 5 and 6). *p*-Cymene is

especially interesting, as this nontoxic solvent can be sourced from biomass.^{41,42} As epoxy resins are relatively polar, and with NaOH having poor solubility in toluene, we were interested in whether varying the polarity of the reaction medium could improve the reaction system. Exchanging the apolar solvent for an aprotic-polar and protic-polar one, respectively, was unrewarding (entries 7 and 8). Likewise, adding 5 vol% of water to the toluene completely inhibited the chemical deconstruction (entry 9). In order to follow up on the role of the polarity of the reaction medium, other variations were tested at 170 °C (entries 11 and 12). Without variation, 59% of BPA had been isolated as point of reference (Fig. 2c). The addition of methanol or tetrabutylammonium chloride (TBAC), which should ease the solvation of hydroxide ions in toluene, both resulted in reduced yields. Based on these results, the key to the base induced bond cleavage seems to be the mismatch between sodium hydroxide and toluene. The poorly solvated hydroxide ions can be expected to be highly reactive, thus enabling challenging bond cleavages. The C–O bond scission proceeds more efficiently on polymeric samples than on model compounds. In accordance with the solvent-base mismatch hypothesis, we considered the possibility that polyether functionalities found in the hardener of the resins might complex sodium ions,⁴³ thereby supporting the formation of poorly shielded hydroxide ions. We tested the addition of catalytic amounts of a crown ether (entry 13) at 170 °C. However no significant change was observed. Lastly, extending the reaction time to 2 days gave no improvement over the reaction time of 24 h (entry 10). We conclude therefore that 81% is the maximal amount of BPA that can be recovered from Airstone using this methodology. The remaining BPA might be adjacent to crosslinking motifs that are more challenging to cleave. This is in accordance with the highest BPA yield from this resin acquired using the ruthenium catalysed hydrogenolysis.²⁵

Scope of the Deconstruction Protocol

With this robust and convenient method at hand, testing the solvent-base mismatch-based deconstruction on a selection of other commercial thermoset resins was undertaken in order to scrutinize the generality of this deconstruction approach (Fig. 5). A universal two-component epoxy adhesive from UHU was cured, grinded into a powder and tested under optimised conditions. This glue uses a mixture of alkyl-based tetra- and pentaamines as the curing agent, which means that the cured resin should have a higher degree of crosslinking than Airstone. Nonetheless, to our delight 84% of BPA could be recovered. Next, a clear cast epoxy resin marketed for arts and crafts by Roizefar was subjected to the basic treatment. Here too, the deconstruction worked efficiently, yielding 86% of BPA. A partly bio-based epoxy resin polymer was examined, consisting of several epoxides and polymeric di- and triamines, and developed for maritime engineering by Sicomin. From this sample only a moderate yield of 58% BPA was recovered. At last, Lightstone 3100E/3102H was tested. This resin uses anhydrides as the curing agents instead of alkyl amines, being developed for pultrusion applications. As the central alcohol groups in the polymer are functionalised as esters (Fig. 1a), the chemical properties and behaviour of this resin is significantly different compared to the amine-cured resins. To our delight, here too BPA could be recovered from the resin in a yield of 57%. We postulate that the ester functionalities are cleaved by the sodium hydroxide before the C–O cleavage occurs.

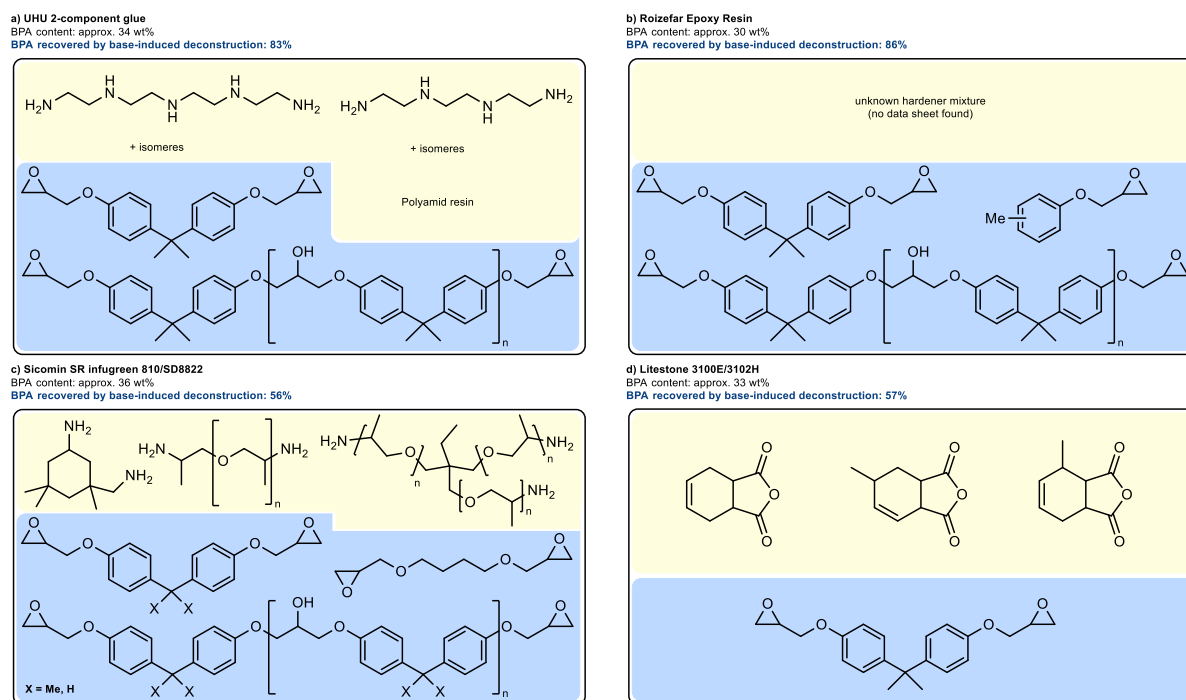
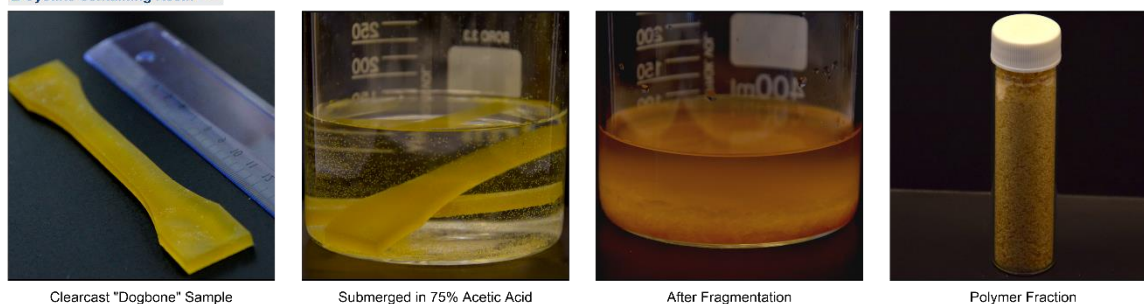


Fig. 3 | Base-mediated deconstruction of a small selection of commercial thermoset epoxy resins. All yields are given for isolated BPA. 100 mg of powdered resin were reacted with 50 wt% powdered NaOH in 4.2 ml toluene at 190 °C for 24 h.

In order to have a perspective for application, the developed protocol needs to tolerate residues from treatments that can separate fibers from the resin. In order to investigate this, tailored resins designed for separation from fibres at their end-of-use were tested. The first is a clear cast sample formulated according to the report by Hinge *et al.*, using L-cystine as an additive in the polymer.^{14,15} The second is a composite sample using the so called “recyclamine technology”¹⁸ from Aditya Birla, which is also used for commercial wind turbine blades.¹⁶ Both samples were each submerged in 75% acetic acid at 70 °C overnight. Over the course of this time, the L-cystine containing resin fragmented into a fine granulate, while the recyclamine-based resin fragments dissolved, leaving behind the glass fibre mesh. The solution of the later was then neutralised, upon which the resin precipitated as a fine powder. After drying, 100 mg of each sample was reacted with 50 wt% of NaOH in toluene under air at 190 °C for 24 h. To our gratitude, BPA was recovered in all cases. Unfortunately, a yield in percentage cannot be reported, as the swelling of the epoxy resins does not allow approximations regarding the wt% of BPA in the resin. For these resins designed for disassembly, preliminary upscaling experiments were conducted. 1 g of each material was reacted with NaOH in a steel autoclave fitted with a Teflon liner. Gratifying, satisfactory yields were observed.

a Acidic fragmentation of tailored epoxy resins

L-Cystine-containing Resin^{14,15}



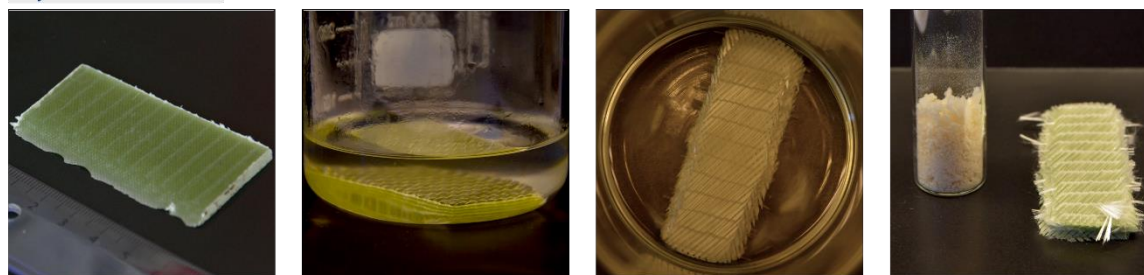
Clearcast "Dogbone" Sample

Submerged in 75% Acetic Acid

After Fragmentation

Polymer Fraction

Recyclamine-based Resin^{17,18}



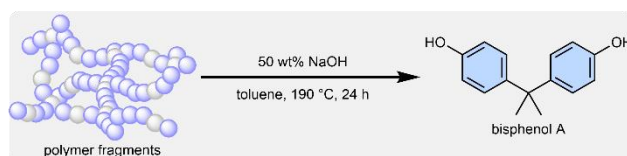
Composite Sample

Submerged in 75% Acetic Acid

After Fragmentation

Polymer Fraction and Glass Fibres

b Base-induced deconstruction of recovered polymer fraction



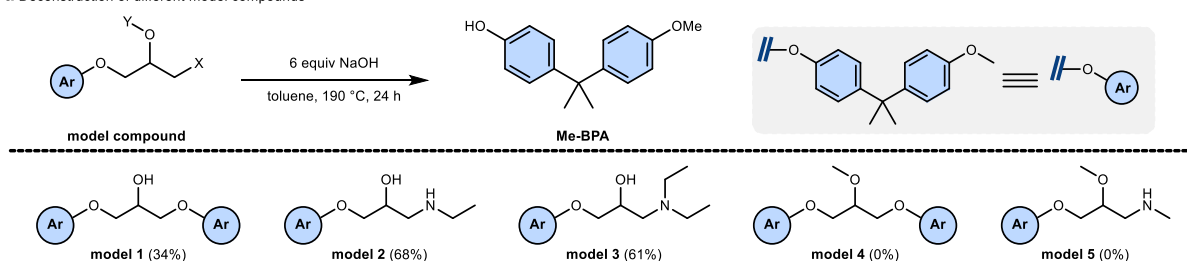
Resin	Scale	Recovered BPA
L-Cystine-based	100 mg	22 wt%
L-Cystine-based	1 g	28 wt%
Recyclamine-based	100 mg	15 wt%
Recyclamine-based	1 g	7 wt%

Fig. 4 | Proof-of-principle for combining acidic fragmentation of tailored resins with base-mediated deconstruction. a, Fragmentation of tailored epoxy resin samples in acetic acid. **b,** Base-induced deconstruction applied to resin samples obtained from separation process.

At last, mechanistic investigations were conducted to elucidate how the deconstruction proceeds on a molecular level. We recognised that the based-induced bond scission could operate through epoxide intermediates analogously to alkaline-based lignin pulping processes.⁴⁴ However, there are considerable chemical and physical differences between thermoset epoxy resins and lignin. Incidentally, based-mediated pulping of the latter does not require solvent-base mismatch conditions. We initiated the investigation by testing model compounds mimicking Amine-cured linkages (Fig. 5a). Both for secondary and tertiary amines, the reaction proceeded smoothly, yielding roughly the double amount of **Me-BPA** as for **model 1**. However, the latter contains two cleavable C–O bonds. Therefore, the bond scission rate for **model 1**, **2** and **3** is consistent under the applied conditions. In order to scrutinise whether the central alcohol moiety is involved in the deconstruction, the methyl capped **model 4** was tested. Here, no conversion was observed, which supports a mechanism proceeding the deprotonation of the alcohol followed by intramolecular nucleophilic attack forming an epoxide under release of a phenoxide. In linkages containing secondary amines, a sodium amide might form, which could be competent at attacking the C–O bond as well, forming an azetidin-3-ol instead of an epoxide. However, **model 5** with a protected alcohol and secondary amine proved unreactive, contradicting that amine moieties are involved in the deconstruction. The investigation was continued *in silico*, probing the free energy profiles of various mechanisms (see Supporting Information). A simplified version of **model 1** named model B (green) and **model 2** herein referred to as model D (red) will be discussed. The most

favoured reaction mode (Fig. 5b) was found to be initiated from an equilibrium between the alcohol and hydroxide and the alkoxide and water **3**. This acid-base reaction goes *via* intermediates **1** and **2** in which substrate and alkoxide interact with either OH⁻ or water via H-bond (B1 and B2 have O-H distances of 1.1 and 1.4 Å, respectively). For Model D, **3** is more stable than **2** by 1.3 kcal/mol due to the intramolecular hydrogen bond with the amine group that stabilises the anion (O-H(N) distance of 2.2 Å). This attractive interaction is also observed in the NCI-plot analysis of intermediate **3**. No such interaction is observed in the same intermediate with model B, explaining why **2** is lower in energy than **3** by 4.9 kcal/mol. The C–O bond cleavage step can occur from either **2** or **3**. For Model B, the pathway following TS2 ($\Delta G = 9.8$ kcal mol⁻¹) is slightly preferred over TS3 ($\Delta G = 10.6$ kcal mol⁻¹) because of the hydrogen bonding with water. Instead, for Model D, TS3 ($\Delta G = 6.4$ kcal mol⁻¹) is favoured over TS2 ($\Delta G = 10.4$ kcal mol⁻¹) thanks to the intra-molecular hydrogen bond with the amine group, which is entropically preferred than the inter-molecular interaction with water. These differences made that the overall barrier for model B is 26.3 kcal mol⁻¹, while for Model D is 25 kcal mol⁻¹.

a Deconstruction of different model compounds



b Free energy profile for the C-O bond cleavage

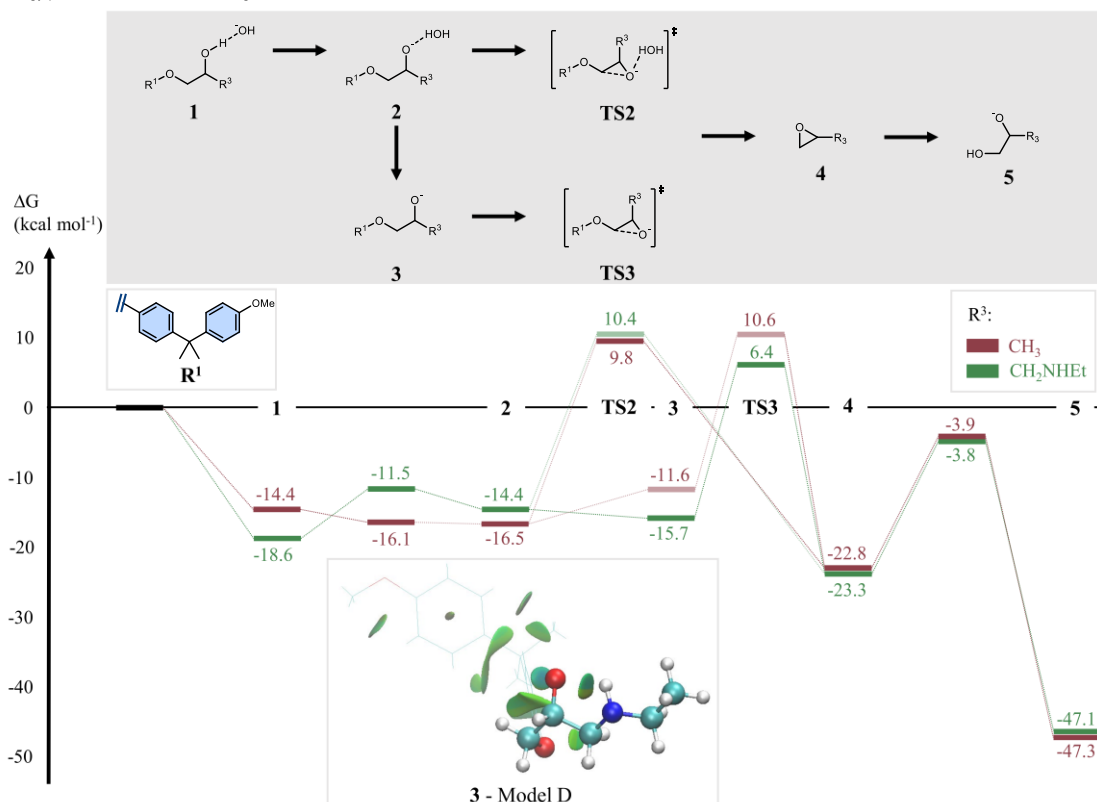


Fig. 5 | Mechanistic investigation on deconstruction process. a, Experimental investigation on different model compounds probing scope and limitations. **b**, Free energy

profile for the C–O bond cleavage reaction with models B (red) and D (green) with the NCI plot for intermediate D3 and free energies in kcal mol⁻¹.

Discussion

Disassembly processes for tailored epoxy composite materials, which enable circularity for their embedded fibres, have been reported and even commercialised. The here reported conveniently simple chemical deconstruction protocol has been shown to be competent at recovering the important polymer building block BPA from such resins after they have been separated from the fibre fraction. Furthermore, both amine-cured and anhydride-cured resins can be deconstructed. The recovered BPA could be reintroduced into existing production chains, leading to virgin polymers, thereby enabling circularity for the polymer fraction.

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Methods

See Supplementary Information for further methods.

Data availability

The authors declare that all other data supporting the findings of this study are available within the paper and its Supplementary Information files.

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Author contributions

Conceptualisation was carried out by HS, AA, AKS and TS. Experiment design was carried out by HS, AA and AKS. The experimental investigation was carried out by HS, AA, BSD and EVS. AA and TS supervised and directed the research, and wrote/revised the original draft. Density Functional Theory studies were conducted by GMFB and ANF. Funding acquisition was carried out by TS. All authors reviewed the final manuscript.

Competing interests

HS, AA and TS are inventors on the provisional patent application (EP22200007), submitted by Aarhus University, which covers the base-mediated deconstruction of epoxy resins. TS is co-owner of SyTracks A/S, which commercialises the COtubes.