Solvent-base mismatch enables the deconstruction of epoxy polymers and bisphenol A recovery

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Fiber-reinforced epoxy composites are key materials for the construction of wind turbine blades and airplanes due to their remarkable mechanical strength 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 complex molecular structures at their disposal. 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 potentially be reused in established polymer production chains, thus closing the recycling loop and reducing the need for virgin resources.

1. Introduction

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, the major application for these resins are fibre-reinforced epoxy composites[2]. To produce such 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 are crucial for the production of wind turbine blades, as well as for uses in the automotive, aeronautical and maritime industries.[2,3]

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It is estimated that wind energy contributes to about 6% of the global energy supply in 2020.[4] 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 future energy supplies.[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% of 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. For example, pyrolysis has been shown to be efficient in removing the epoxy resins, however, such processes are energy intensive and result in the recovery of lower quality fibres.[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] As an example, the incorporation of ester moieties allows selective cleavage *via* thermal[12] or basic[13] 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 ketal containing diamines (recyclamine) in the hardener.[17,18] While this represents an elegant strategy to recover fibres from composites, the reuse of the separated epoxy fraction is limited to filler material at best,[19–24] and consequently these novel resin formulations fall short of introducing a circular economy for these important structures.

Recently, our group reported a catalytic disconnection process based on ruthenium for selective cleavage of 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 and complementary approaches for deconstructing relevant epoxy composites. In this work, we report on the development of a robust and simple protocol for cleaving specific molecular linkages in the epoxy resins after they have been separated from the fibres. Using a reaction system consisting of an alkali mismatched with an apolar organic solvent at elevated temperatures, the important polymer building block bisphenol A (BPA)[26,27] can be recovered from different commercial epoxy samples. The discrepancy in polarity controlling the solubility of the reagent in the reaction medium proved to be a key factor. 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 can now be achievable (Fig 1b).

2. Results and Discussion

Reported protocols for separating fibres from the resins rely on acidic or basic treatments of the composites, and therefore identifying a methodology for the disassembly of the epoxy polymer tolerating potential residual acid or base in the epoxy fraction from the fibre separation process is a critical success criterium. Recently, a photocatalytic process for disconnecting C–C bonds in epoxy polymers has been reported.[30,31] Although mild and elegant in its nature, a subsequent ether cleavage using BBr_3 is necessary to release the bisphenol A. As an alternative approach, we decided to target the C(alkyl)–O bonds formed during the reaction of BPA with epichlorohydrin, as its cleavage would directly liberate BPA (Fig.1b), analogously to the Ru-catalysed process for composites.[25] During our preliminary studies on epoxy resin disconnections, we discovered that certain bases are competent at efficiently liberating phenols from the dehydrogenated epoxy models, such as ketone **I**, representing the oxidised form of the linkage generated from BPA and epichlorohydrin. Additionally, computational studies on epoxy linkages revealed a significant reduction in the C–O bond dissociation energy by 10.4 kcal mol^{-1} upon oxidation of the α-phenoxyalcohols to their corresponding ketones.[25] Based on this finding, we envisioned combining a robust alcohol oxidation process with stochiometric

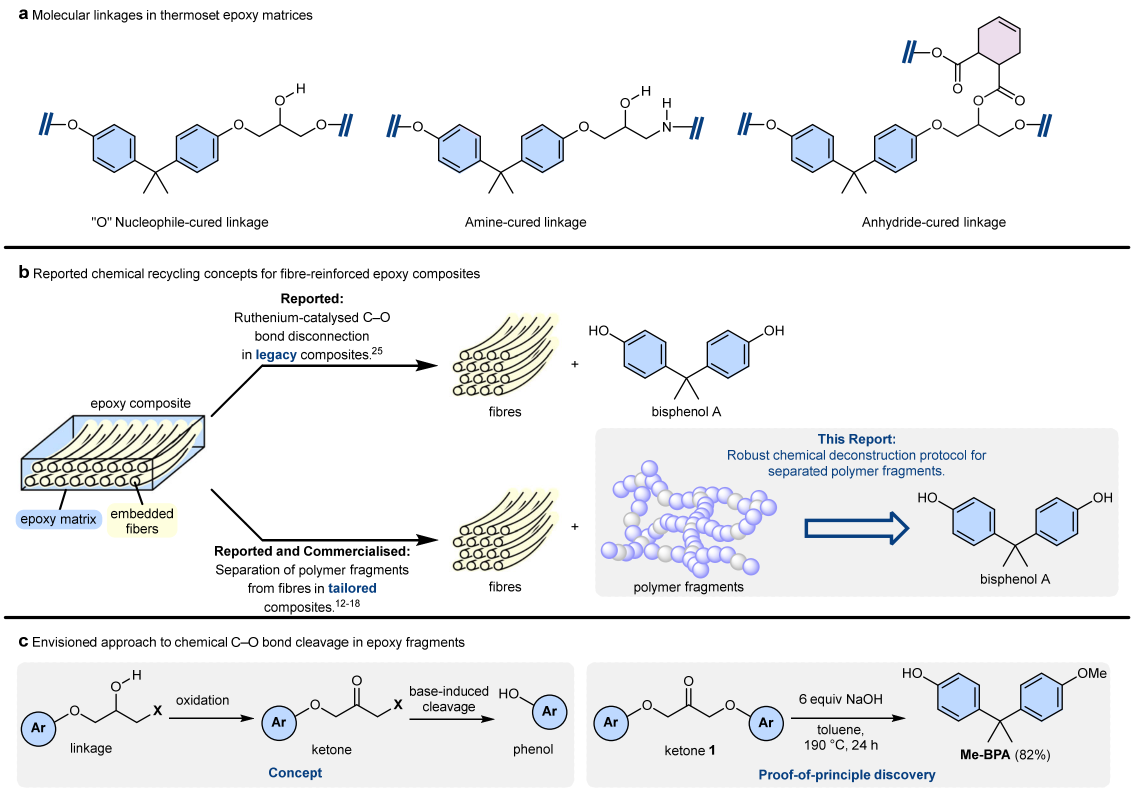


Fig. 1 Concept for deconstruction of epoxy resins recovered after separation from fibres. a. Schematic illustration of a fibre-reinforced epoxy composite and its reported separation into a fibre and epoxy fraction and the herein reported deconstruction of epoxy resins into BPA. b. Linear linkage motifs in amine-cured epoxy resins. c. Project idea for a chemical deconstruction of epoxy resins. Ar = Me-BPA.

amounts of base with respect to the epoxy resins (Fig. 1c). We also recognised that processes requiring base would neutralise the residual acid in an epoxy fraction recovered from composite materials, thus avoiding extensive drying processes after separation from the fibres.

We commenced our investigations on this two-step process by examining the feasibility of a catalytic acceptorless dehydrogenation[32,33] for ketone formation. Several protocols rely on the use of stochiometric amounts of bases, which also have the potential for promoting the C–O bond cleavage event in ketone **I**.[34–37] Beller *et al*. reported a highly efficient dehydrogenation of isopropanol using an *in situ* generated Ru-MACHO complex in the presence of stoichiometric NaO*i*Pr in regards to the alcohol.[37] The dehydrogenation in tandem with the base-induced C–O bond cleavage was optimised on **model 1**. The conditions reported by Beller *et al.*,[37] 3 mol% of $(Ph_3P)_3Ru(CO)H_2$ together with 3 mol% of bis[(2-diphenylphosphino)ethyl]ammonium chloride were chosen as the catalytic system (Fig. 2a, see SI for optimisation). By combining the ruthenium catalyst with 6 equiv of powdered NaOH at a reaction temperature of 190 °C, it was possible to deconstruct the model compound over the course of 24 h, affording **Me-BPA** in a yield of 68% (Fig. 2a). While using stochiometric reagents in excess generally can be considered a flaw regarding atom economy[38] and the concept of green chemistry,[39] caustic soda (NaOH) is a commodity chemical exploited in several industrial processes in large quantities and importantly is formed as a by-product during the production of chlorine.[40] Much to our surprise, when the appropriate control experiments were conducted, we discovered that the transformation of **model 1** in the absence of the catalyst system still afforded a 34% yield of **Me-BPA**.

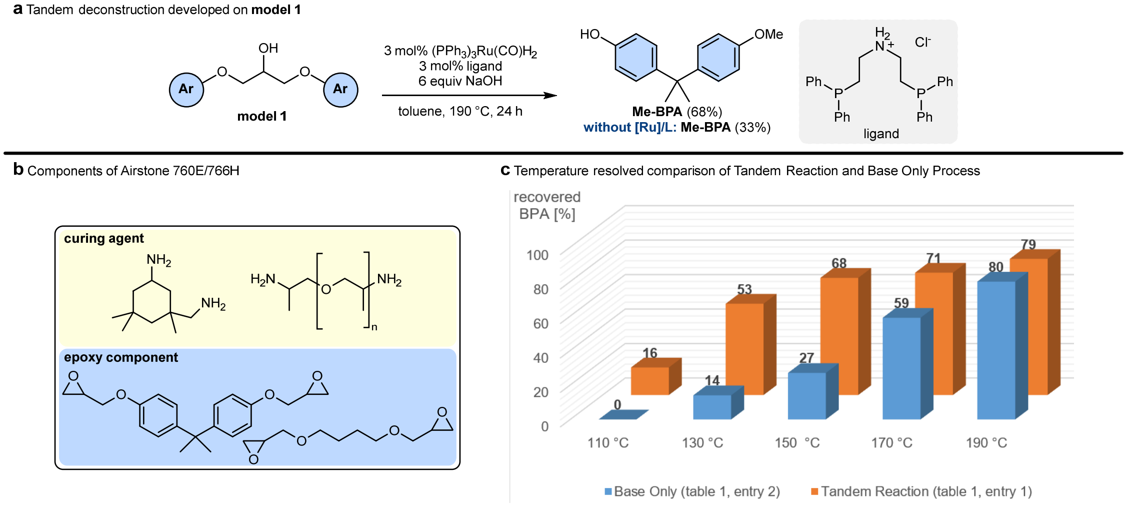


Fig. 2 Method development and application to thermoset epoxy resin. a. Method for C–O bond cleavage developed on **model 1**. b. Formulation of the epoxy resin used as benchmark system. c. Comparison between the tandem reaction using a ruthenium catalyst and base and the base only process in the deconstruction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

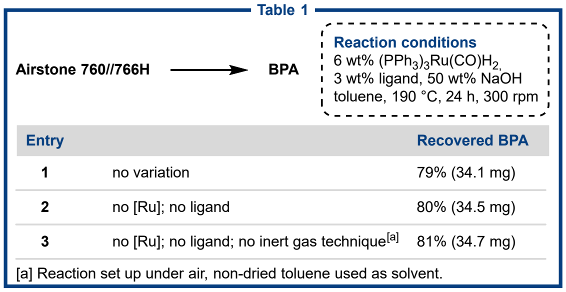


Table 1

Airstone 760//766H $\longrightarrow$ BPA

Reaction conditions
6 wt% $(PPh_3)_3Ru(CO)H_2$,
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 1 Tandem reaction and background reaction on polymeric sample.

With a proof-of-principle of the tandem disconnection process at hand, we moved on to test 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.[25] It consists of bisphenol A diglycidyl ether and ethylene glycol diglycidyl ether as the liquid epoxy resin with a combination of isophorone diamine and poly(oxypropylene) diamine as the curing agents (Fig. 2b). The cured resin contains approximately 43 wt% of BPA, from which isolated yields were calculated. As the crosslinked resins are not soluble, and chemical reactions are limited to the surface of the material, 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% Ru-precatalyst and ligand each were added. The 50 wt% of NaOH added roughly equates to 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). Using these conditions, an isolated yield BPA from Airstone of 79% was achieved (entry 1). Unexpectedly, this yield is significantly higher than when employing **model 1** as a substrate. This discrepancy in yield led us to investigate the base-mediated background reaction observed with the polymeric sample in the absence of catalyst. Interestingly, omitting the ruthenium complex and ligand, an 80% isolated yield of BPA was recovered from the resin (entry 2). A consistent yield could be obtained (entry 3) when the catalyst-free reaction was repeated under air using benchtop-grade solvent (309 ppm water content).

At this point we assumed that two reaction pathways are simultaneously cleaving the Airstone epoxy resin: a base only process, and a Ru-catalysed tandem reaction as originally envisioned. To test this hypothesis, we compared the influence of the catalyst across different temperatures. The results described in Table 1, entries 1 and 2 were used as the benchmark (Fig. 2c). In the absence of catalyst (blue), the reaction is highly efficient at 190 °C with 80% recovery of the BPA. However, already at 170 °C, the yield plummets to 59%. The trend continues almost linearly, with no detection of liberated BPA at 110 °C. On the contrary, in the presence of a ruthenium catalyst (orange), the quantity of liberated BPA only decreases slightly as the temperature is reduced, as exemplified with the isolation of a 68% yield of BPA at 150 °C compared to 27% yield in the absence of the catalyst. Below 130 °C, the yields drop significantly with 16% of isolated BPA at 110 °C. These results suggest that both pathways for the liberation of BPA are viable, with the tandem route favouring the overall cleavage at a lower reaction temperature. However, in light of the simplicity of a catalyst free protocol for the deconstruction of epoxy resins, we decided to continue this study with focus on the hydroxide-promoted process.

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	*o*-xylene instead of toluene	75% (32.1 mg)
6	*p*-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)

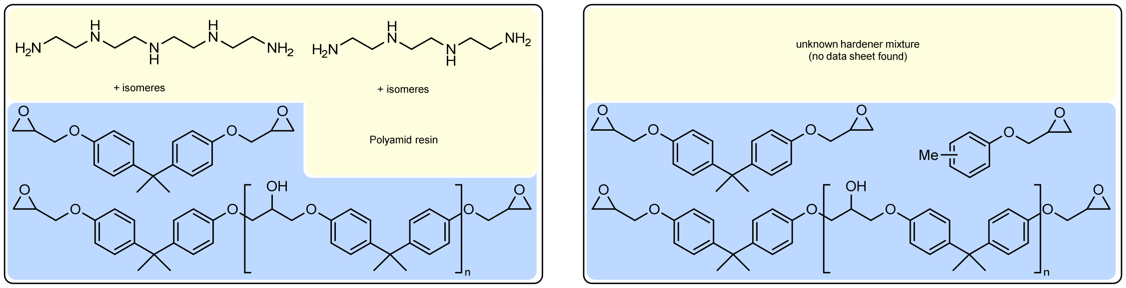
Table 2 Optimisation and control experiments for the base-induced C–O bond cleavage.

Despite the high temperature and the relatively high base loading, this catalyst-free transformation is simple, inexpensive and robust, and as such potentially attractive for large scale applications. Due to the potential of high pressure build up applying toluene as the solvent at 190 °C, similar media with higher boiling points were also considered (Table 2). Switching to either *o*-xylol (bp 144 °C) or *p*-cymene (bp 177 °C) gave comparable yields of recovered BPA (entries 5 and 6). *p*-Cymene has the advantage of low toxicity and that it can be sourced from biomass.[41,42] Next, we studied the relationship between polarity of the reaction medium and efficiency of deconstruction. Initially, conditions were tested that should increase the solubility of NaOH. Exchanging toluene for representatives of aprotic-polar (DMF) or protic-polar (water) equivalents, proved unrewarding (entries 7 and 8). Likewise, adding 5 vol% of water to toluene completely inhibited the chemical deconstruction (entry 9). To follow up on the role of the polarity of the reaction medium, other variations using toluene as solvent were tested at 170 °C (entries 11 and 12), comparing to the 59% yield of BPA that had previously been isolated from the disassembly (Fig. 2c). The addition of methanol or tetrabutylammonium chloride (TBAC), which promotes the solvation of hydroxide in toluene, both resulted in reduced yields. These results indicate that the key aspect of the base induced bond cleavage is the mismatch between sodium hydroxide and toluene. The poorly solvated hydroxide ions would undoubtedly lead to high reactivity. Likewise, ionic or polar intermediates formed during the transformation would be equally affected by the polar medium, thus enabling challenging bond cleavages. The C–O bond scission proceeds more efficiently on polymeric samples then 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 led to no improvement over the reaction time of 24 h (entry 10). Based on the findings discussed above, we conclude that 81% yield is the maximal amount of BPA that can be recovered from Airstone using this methodology. The inability to release the remaining BPA may potentially be linked to its presence adjacent to specific crosslinking motifs that remain more challenging to cleave. This is in accordance with the reported highest BPA yield from the same resin applying the ruthenium catalysed hydrogenolysis.[25]

With this robust and convenient method at hand, testing this solvent-base mismatch disassembly protocol on a selection of other commercial thermoset resins was undertaken in order to scrutinise the generality (Fig. 5). A universal two-component epoxy adhesive from the company UHU was cured, grinded into a powder and tested under optimised conditions. As the curing agents for this glue are composed of a mixture of alkyl-based tetra- and pentaamines, the cured resin should be characterised by a higher degree of crosslinking than Airstone. Nonetheless, to our delight an 84% yield of BPA was recovered. Next, a clear cast epoxy resin marketed for arts and crafts by Roizefar was subjected to the basic treatment. Here too, the deconstruction performed 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 an adequate yield of 58% BPA was recovered. Finally, a molecularly different epoxy represented by Lightstone 3100E/3102H and developed for pultrusion applications was tested. In contrast to the other epoxy resins, the alkyl amine curing agents are replaced by carboxylic anhydrides. As the central alcohol groups in the polymer are functionalised as esters (Fig. 1a), the chemical properties and behaviour of this resin are significantly different in comparison to the amine-cured resins. To our delight, here too BPA was recovered from the resin in a yield of 57%. Mechanistically, we propose the cleavage of the ester functionalities by sodium hydroxide takes place prior to the key C–O bond cleavage.

a) UHU 2-component glue
BPA content: approx. 34 wt%
BPA recovered by base-induced deconstruction: 83%

b) Roizefar Epoxy Resin
BPA content: approx. 30 wt%
BPA recovered by base-induced deconstruction: 86%



c) Sicomin SR infugreen 810/SD8822
BPA content: approx. 36 wt%
BPA recovered by base-induced deconstruction: 56%

d) Litestone 3100E/3102H
BPA content: approx. 33 wt%
BPA recovered by base-induced deconstruction: 57%

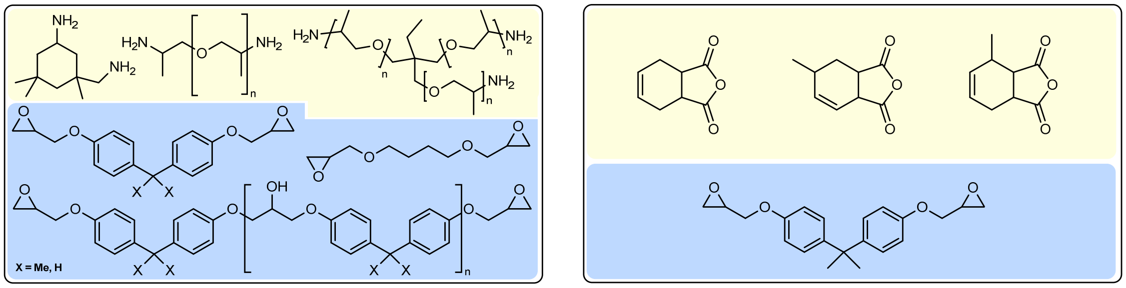
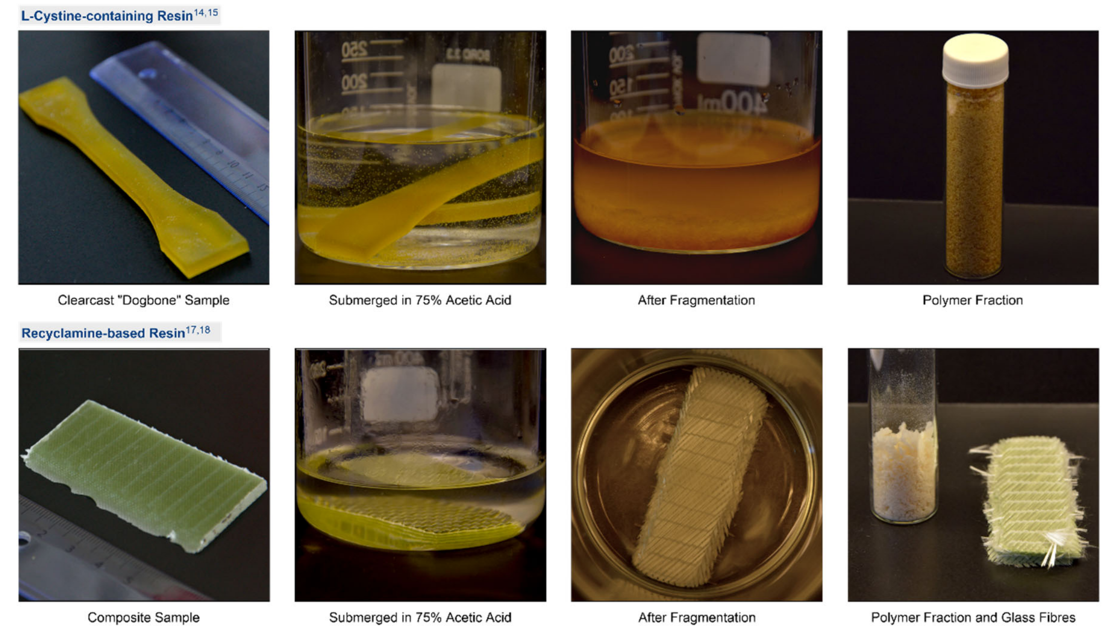


Fig. 3 Base-mediated deconstruction of a small selection of commercial thermoset epoxy resins. All yields are given for isolated BPA. 100 mg of powdered resin were reacted with 50 wt% powdered NaOH in 4.2 ml toluene at 190 °C for 24 h.

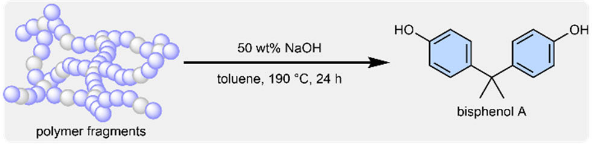
In order to have a perspective for application, the developed protocol must tolerate residual chemical contaminants from the initial separation processes of the fibres from the epoxy resins.

To investigate this, tailored resins designed for facile separation from fibres at their end-of-use were tested. The first example is a clear cast sample formulated with L-cystine as an additive to the polymer according to reports by Hinge *et al.*[14,15] The second is a composite sample applying the Aditya Birla recyclamine technology,[18] now commercialised for the construction of wind turbine blades.[16] Each sample was 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 recylamine-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. BPA was recovered for both the L-cystine and the recylamine-based resin in 22 wt% and 15 wt%, respectively. Unfortunately, a yield in percentage cannot be reported, as solvent absorbed within the thermoset matrix changes the mass to BPA ratio. For these resins designed for disassembly, preliminary upscaling experiments were conducted. 1 g of each

a Acidic fragmentation of tailored epoxy resins



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.

material was reacted with NaOH in a steel autoclave fitted with a Teflon liner. Gratifying, BPA was recovered from both samples in comparable wt% to the small batch.

Elucidation of observed reactivity

At last, investigations were conducted to elucidate the mechanistic intricacies of the deconstruction process 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.[25] For example, based-mediated pulping of the latter does not require solvent-base mismatch conditions.[44] We initiated the investigation by testing model compounds mimicking the amine-cured linkages (Fig. 5a). The reaction proceeded smoothly for both the secondary and tertiary amines, yielding roughly double the amount of **Me-BPA** compared to **model 1**. However, the latter contains two cleavable C–O bonds. Therefore, the bond scission efficiency for **model 1**, **2** and **3** is consistent under the applied conditions. To scrutinise whether the central alcohol moiety is involved in the deconstruction, the *O*-methylated **model 4** was tested. For this substrate, no conversion was observed, supporting a mechanism proceeding through the deprotonation of the alcohol followed by intramolecular nucleophilic attack to generate an epoxide with simultaneous release of the phenoxide. In linkages containing amines, the lone electron pair of the nitrogen 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.

In silico investigations were undertaken, probing the free energy profiles of various mechanisms (see Supporting Information). Simplified versions of **model 1** and **model 2**, denoted model B (green) and model D (red), respectively, will be discussed. Intermediates are indicated with arabic numerals. The most favoured reaction mode (Fig. 5b) was found to be initiated from an equilibrium between the alcohol and hydroxide adduct **1** and the alkoxide and water adduct **2**. Interestingly, for the amine containing model D, dissociation from water forming the free alkoxide **3** is favoured by 1.3 kcal/mol due to the intramolecular hydrogen bond stabilising the anion (O–H(N) distance of 2.2 Å). This attractive interaction is also observed in the Non-Covalent Interaction-plot analysis for intermediate **3**. No such interaction is observed for model B. The central C–O bond cleavage step can occur from both the free alkoxide **3** and its water adduct **2**. 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}) becomes favoured over TS2 ($\Delta G = 10.4$ kcal mol^{-1}) due to the intra-molecular hydrogen bond with the amine group, which is entropically preferred than the inter-molecular interaction with water. In either case, the nucleophilic attack forms epoxide **4**, while liberating a phenolate. Considering the energy barriers as well as the observations that the energy difference between the epoxide and the alcoholate **2** is only 6.7 kcal mol^{-1} (model B) and 11.8 kcal mol^{-1} (model D), respectively, a dynamic equilibrium between these species can be expected at 190 °C. However, the nucleophilic attack of hydroxide on the epoxide **4** to generate **5** acts as a thermodynamic driving force.

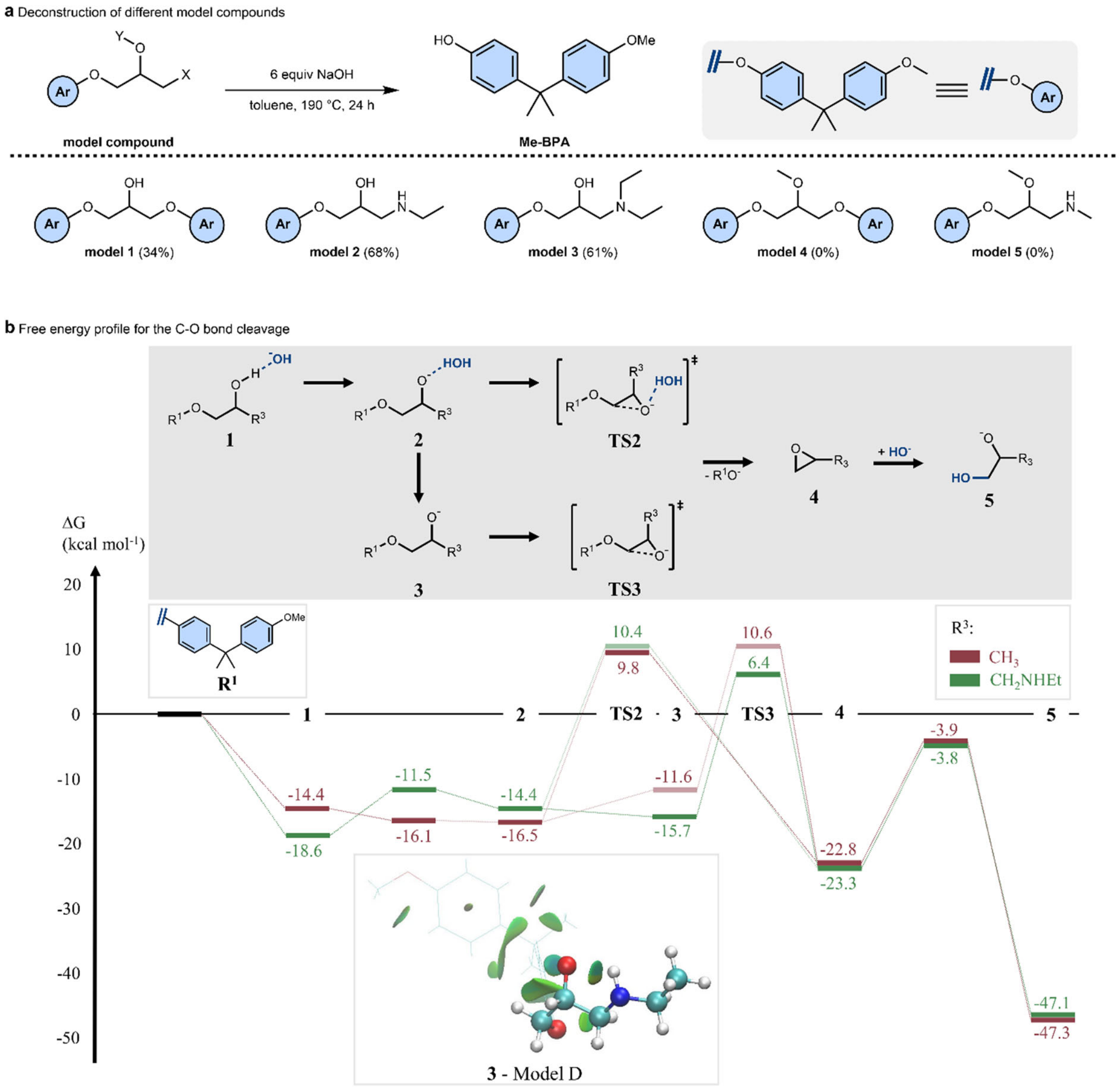


Fig. 5 Mechanistic investigation on deconstruction process. a. Experimental investigation on different model compounds probing scope and limitations. b. Free energy profile for the C–O bond cleavage reaction with models B (red) and D (green) with the NCI plot for intermediate D3 and free energies in kcal mol^{-1}.

Conclusion

Disassembly processes for tailored epoxy composite materials, which enable circularity for their embedded fibres, have been reported and even commercialised.[12–18] Our results reported here on a simple chemical deconstruction protocol of the epoxy matrix provide a powerful means for the recovery of the important polymer building block BPA from such resins after separation from the fibre fraction. Furthermore, both amine-cured and anhydride-cured resins can be deconstructed applying this chemical technology. Notably, the recovered BPA could potentially be reintroduced into existing production chains, leading to the fabrication of virgin polymers, and thereby enabling circularity for the polymer fraction. This work opens yet another door for chemical recycling-based approaches that hold the potential to offer solutions necessary to implement policies ending unsustainable practises.

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.

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Methods and Data availability

See Supplementary Information for further methods. 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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