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## 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 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 the loss of complex molecular structures during 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 process/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.

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## 1. Introduction

Thermoset epoxy resins are a class of polymers that are cured on-site by blending short-chain epoxy resins with a curing agent, resulting in crosslinked molecular networks dominated by  $\sigma$  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.<sup>1</sup> Alternatively, carboxylic acid anhydrides can be used to cure epoxides, forming carboxylate ester networks (Fig. 1a).<sup>1</sup> Besides coatings and laminates, the major application of these resins is fibre-reinforced epoxy composites.<sup>2</sup> To produce such

composites, glass or carbon fibres are embedded in a thermoset polymer matrix, resulting in favourable mechanical properties, which allow appliances to use them as high-performance materials. These composites are crucial for the production of wind turbine blades and for uses in the automotive, aeronautical and maritime industries.<sup>2,3</sup>

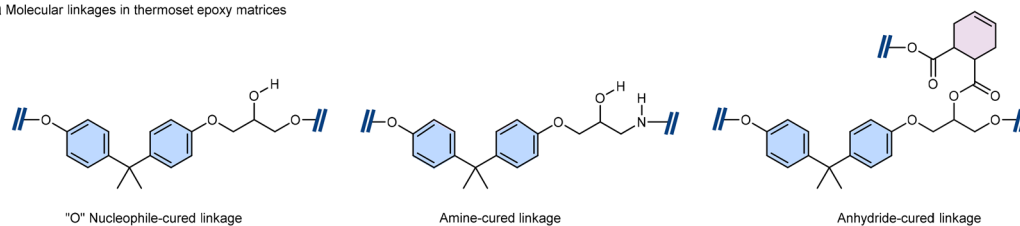
It is estimated that wind energy contributed to about 6% of the global energy supply in 2020.<sup>4</sup> Due to the environmental ramification of burning fossil fuels, as well as geopolitical concerns obstructing the procurement of such fuels, wind energy is widely regarded as instrumental in securing future energy supplies.<sup>4</sup> Nevertheless, recycling solutions for end-of-use composite materials are critically underdeveloped,<sup>2,5</sup> 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,<sup>6</sup> with each one containing 80 to 90% of composite material by weight.<sup>4</sup> 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.<sup>4</sup> 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;<sup>4,7,8</sup> however, processing these highly durable thermoset composites is notoriously challenging.<sup>2</sup>

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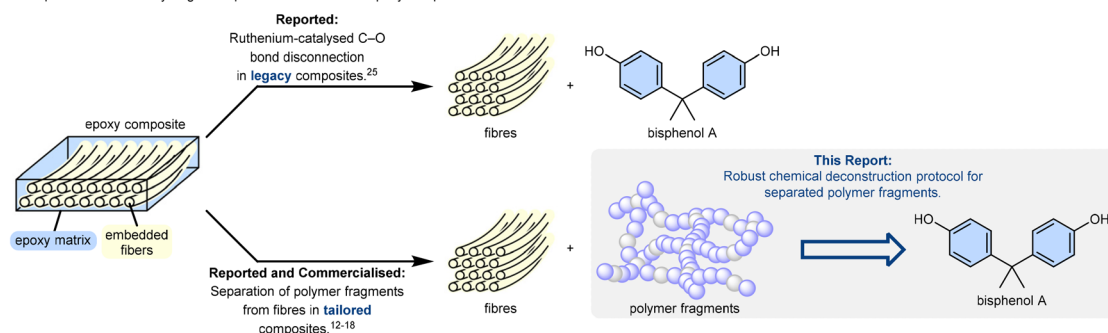
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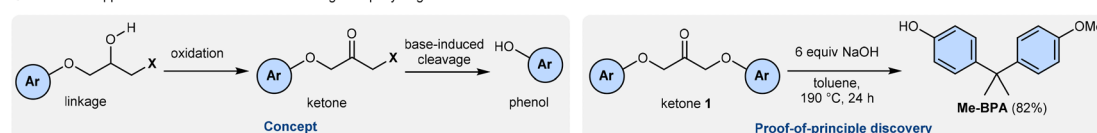
## a Molecular linkages in thermoset epoxy matrices



## b Reported chemical recycling concepts for fibre-reinforced epoxy composites



## c Envisioned approach to chemical C-O bond cleavage in epoxy fragments



**Fig. 1** Concept for the deconstruction of epoxy resins recovered after separation from fibres. (a) Linear linkage motifs in amine-cured epoxy resins. (b) 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. (c) Project idea for a chemical deconstruction of epoxy resins. Ar = Me-BPA.

In the last decades, attempts to valorise end-of-use composites have focused on recovering fibres *via* harsh treatments that destroy the polymer matrices. For example, energy intense pyrolysis or solvolysis using molten potassium hydroxide has been shown to be efficient at removing epoxy resins. However, such processes are energy intensive and result in the recovery of lower quality fibres.<sup>2,5,9</sup> 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.<sup>2,10,11</sup> As an example, the incorporation of ester moieties allows selective cleavage *via* thermal<sup>12</sup> or basic<sup>13</sup> treatment. Alternatively, epoxy resins can be designed to effectively fracture through physical processes upon exposure to certain triggers, such as acetic acid.<sup>14,15</sup> 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,<sup>16</sup> due to the inclusion of ketal containing diamines (recyclamine) in the hardener.<sup>17,18</sup> 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,<sup>19-24</sup> 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 the 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.<sup>25</sup> While performing the chemical deconstruction directly on the composite is desirable, the current catalytic system on the solid-liquid interface of the composite samples requires reaction times on the order of days. Therefore, we became 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)<sup>26,27</sup> 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 the recovery of both fibres and polymer building blocks through a combinatory disassembly process/chemical deconstruction strategy, a circular economy<sup>28,29</sup> for composite-based structures such as wind turbine blades can potentially be achievable (Fig. 1b).

## 2. Results and discussion

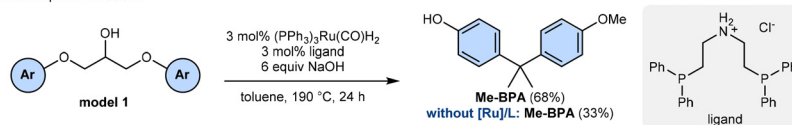
Reported protocols for separating fibres from the resins rely on acidic or basic treatments of the composites, and therefore identifying a methodology for the disassembly of the epoxy polymer tolerating potential residual acid or base in the epoxy fraction from the fibre separation process is a critical success criterion. Recently, a photocatalytic process for disconnecting C–C bonds in epoxy polymers has been reported.<sup>30,31</sup> Although mild and elegant in nature, a subsequent ether cleavage using  $\text{BBr}_3$  is necessary to release bisphenol A. As an alternative approach, we decided to target the C(alkyl)–O bonds formed during the reaction of BPA with epichlorohydrin, as their cleavage would directly liberate BPA (Fig. 1b), analogously to the Ru-catalysed process for composites.<sup>25</sup> 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 **1**, 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<sup>−1</sup> upon oxidation of  $\alpha$ -phenoxyalcohols to their corresponding ketones.<sup>25</sup> Based on this finding, we envisioned combining a robust alcohol oxidation process with stoichiometric amounts of bases with respect to the epoxy resins (Fig. 1c). We also recognised that processes requiring a 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<sup>32,33</sup> for ketone formation. Several protocols

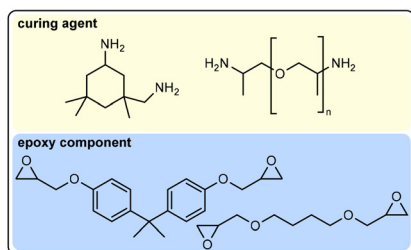
rely on the use of stoichiometric amounts of bases, which also have the potential to promote the C–O bond cleavage event in ketone **1**.<sup>34–37</sup> Beller *et al.* reported a highly efficient dehydrogenation of isopropanol using an *in situ* generated Ru-MACHO complex in the presence of stoichiometric NaOiPr in regard to the alcohol.<sup>37</sup> The dehydrogenation in tandem with the base-induced C–O bond cleavage was optimised on **model 1**. The conditions reported by Beller *et al.*,<sup>37</sup> 3 mol% of  $(\text{Ph}_3\text{P})_3\text{Ru}(\text{CO})\text{H}_2$  together with 3 mol% of bis[(2-diphenylphosphino)ethyl] ammonium chloride, were chosen as the catalytic system (Fig. 2a, see the ESI† 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 stoichiometric reagents in excess is generally considered a flaw regarding atom economy<sup>38</sup> and the concept of green chemistry,<sup>39</sup> 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.<sup>40</sup> 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**.

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.<sup>25</sup> 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

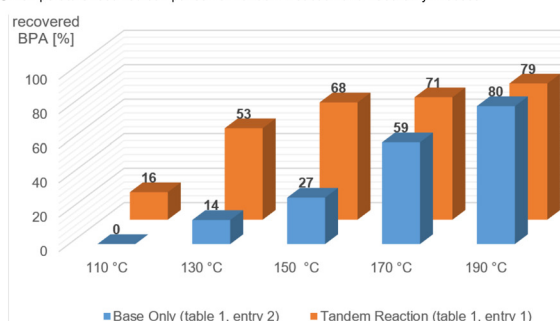
a Tandem deconstruction developed on **model 1**



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 resins. (a) Method for C–O bond cleavage developed on **model 1**. (b) Formulation of the epoxy resin used as a 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 given for isolated BPA. Reaction conditions are based on those described in Table 1, entries 1 and 2, respectively. Ar = Me-BPA.

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 ground 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 and 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 the described conditions, an isolated yield of 79% of BPA from Airstone was achieved (entry 1). Unexpectedly, this yield is significantly higher than that 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 a 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 simultaneously cleave 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 a 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. In contrast, in the presence of a ruthenium catalyst (orange), the quantity of liberated BPA only decreases slightly as the temperature is reduced, as exemplified by 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 a focus on the hydroxide-promoted process.

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. However, operating with a solvent significantly above its boiling point and the resulting buildup in pressure in the reaction vessel could be problematic on a larger scale. Therefore, solvents similar to toluene, but with higher boiling points, were 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) while circumventing the safety issues of pressurised reactors. Additionally, *p*-cymene has the advantage of low toxicity and can be sourced from biomass.<sup>41,42</sup>

In industrial production, BPA is purified *via* washing and/or crystallisation rather than column chromatography.<sup>26</sup> The exact purification processes used by respective companies are commonly not public knowledge. Nonetheless, we were interested to see if a simple aquatic workup using hydrochloric acid to protonate the amines and extraction with ethyl acetate could serve to yield BPA in acceptable purity. To our delight, running the reaction on a 500 mg scale (entry 7) and applying this simplified workup yielded 221 mg of BPA with a purity of 91.3% as determined by GC-MS (see the ESI†). As around 20% of BPA was not recovered from the resin under the standard reaction conditions, an extended reaction time of 2 days was tested (entry 8). Here, no improved yield was observable. As the previously reported ruthenium catalysed C–O bond scission on the same epoxy polymer also did not provide a BPA recovery above 80%,<sup>25</sup> we speculate that certain (cross)linkages are not accessible to these deconstruction methodologies.

Next, we studied the relationship between the polarity of the reaction medium and the efficiency of deconstruction. Initially, conditions were tested that should increase the solu-

**Table 1** Tandem reaction and background reaction on polymeric samples

<div style="display: flex; align-items: center; justify-content: center;"> <div style="text-align: center; margin-right: 10px;"> Airstone 760//766H </div> <div style="margin-right: 10px;"> → </div> <div style="text-align: center; margin-right: 10px;"> BPA </div> <div style="border: 1px dashed black; padding: 5px; text-align: center; font-size: 0.8em;"> <b>Reaction conditions</b>  6 wt% (PPh<sub>3</sub>)<sub>3</sub>Ru(CO)H<sub>2</sub>,  3 wt% ligand, 50 wt% NaOH  toluene, 190 °C, 24 h, 300 rpm </div> </div>		
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 <sup>a</sup>	81% (34.7 mg)

<sup>a</sup> Reaction set up under air, and non-dried toluene used as a solvent.

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

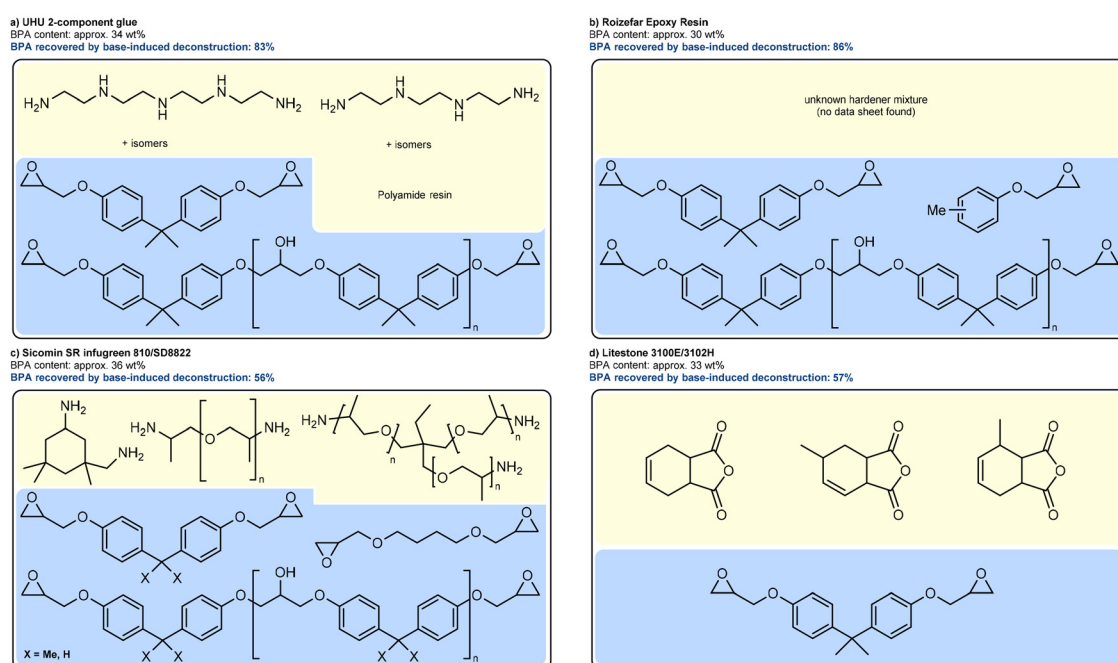
<div style="display: flex; align-items: center; justify-content: center;"> <div style="text-align: center; margin-right: 10px;"> Airstone 760//766H </div> <div style="margin-right: 10px;"> → </div> <div style="text-align: center; margin-right: 10px;"> BPA </div> <div style="border: 1px dashed black; padding: 5px; text-align: center; font-size: 0.8em;"> <b>Reaction conditions</b>  50 wt% NaOH  toluene, 190 °C, 24 h, 300 rpm </div> </div>		
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	Aquatic workup only, 500 mg scale	221 mg (91.3% pure)
8	2 d reaction time	80%
9	DMF instead of toluene	Traces
10	Water instead of toluene	0%
11	5 vol% water	0%
12	12 vol% MeOH, 170 °C	45% (19.1 mg)
13	10 mol% TBAC, 170 °C	45% (19.3 mg)
14	10 mol% 15-crown-5, 170 °C	55% (23.5 mg)

bility of NaOH. Exchanging toluene for representatives of aprotic-polar (DMF) or protic-polar (water) equivalents proved unrewarding (entries 9 and 10). Likewise, adding 5 vol% of water to toluene completely inhibited the chemical deconstruction (entry 11). To follow up on the role of the polarity of the reaction medium, other variations using toluene as the solvent were tested at 170 °C (entries 12–14), compared to the 59% yield of BPA that had previously been isolated from the disassembly process (Fig. 2c). The addition of methanol or tetrabutylammonium chloride (TBAC), which promotes the solvation of hydroxide in toluene, resulted in reduced yields in both cases. These observations indicate that the key aspect of 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 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,<sup>43</sup> thereby supporting the formation of poorly shielded hydroxide ions. We tested the addition of catalytic amounts of a crown ether (entry 14) at 170 °C. However, no significant change was observed.

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 of the process (Fig. 3). A universal two-component epoxy adhesive from the company UHU was cured, ground 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 cross-linking 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 Litestone 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 with the amine-cured resins. To our delight, here too BPA was recovered from the resin in a yield of 57%. Mechanistically, we propose that the cleavage of the ester functionalities by sodium hydroxide takes place prior to the key C–O bond cleavage.

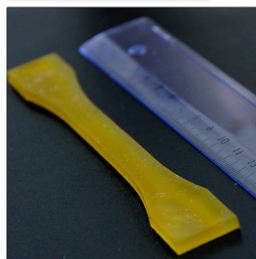
In order to have a perspective for application, the developed protocol must tolerate residual chemical contaminants from the initial separation processes of the fibres from the epoxy resins. To investigate this, tailored resins designed for facile separation from fibres at their end-of-use were tested. 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.*<sup>14,15</sup> The second is a composite sample applying the Aditya Birla recyclamine technology,<sup>18</sup> now commercialised for the con-



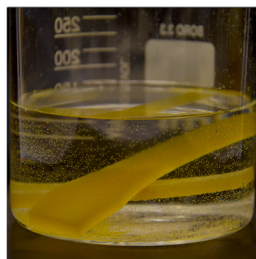
**Fig. 3** Base-mediated deconstruction of a small selection of commercial thermoset epoxy resins. All yields are given for isolated BPA. 100 mg of powdered resin were reacted with 50 wt% powdered NaOH in 4.2 ml toluene at 190 °C for 24 h.



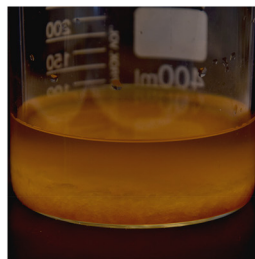
## a Acidic fragmentation of tailored epoxy resins

L-Cystine-containing Resin<sup>14,15</sup>

Clearcast "Dogbone" Sample



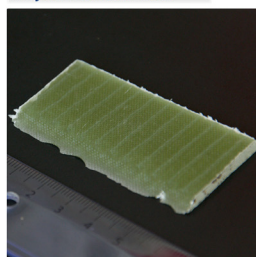
Submerged in 75% Acetic Acid



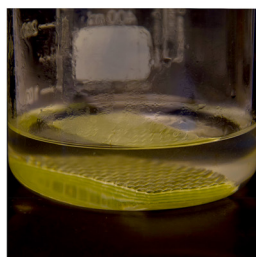
After Fragmentation



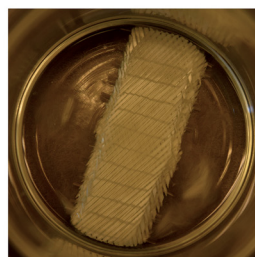
Polymer Fraction

Recyclamine-based Resin<sup>17,18</sup>

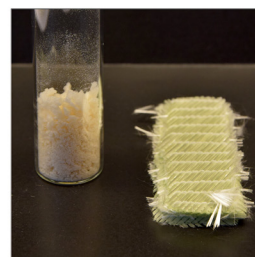
Composite Sample



Submerged in 75% Acetic Acid

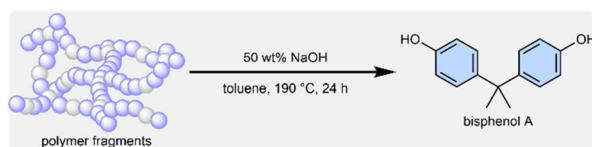


After Fragmentation



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 the separation process.

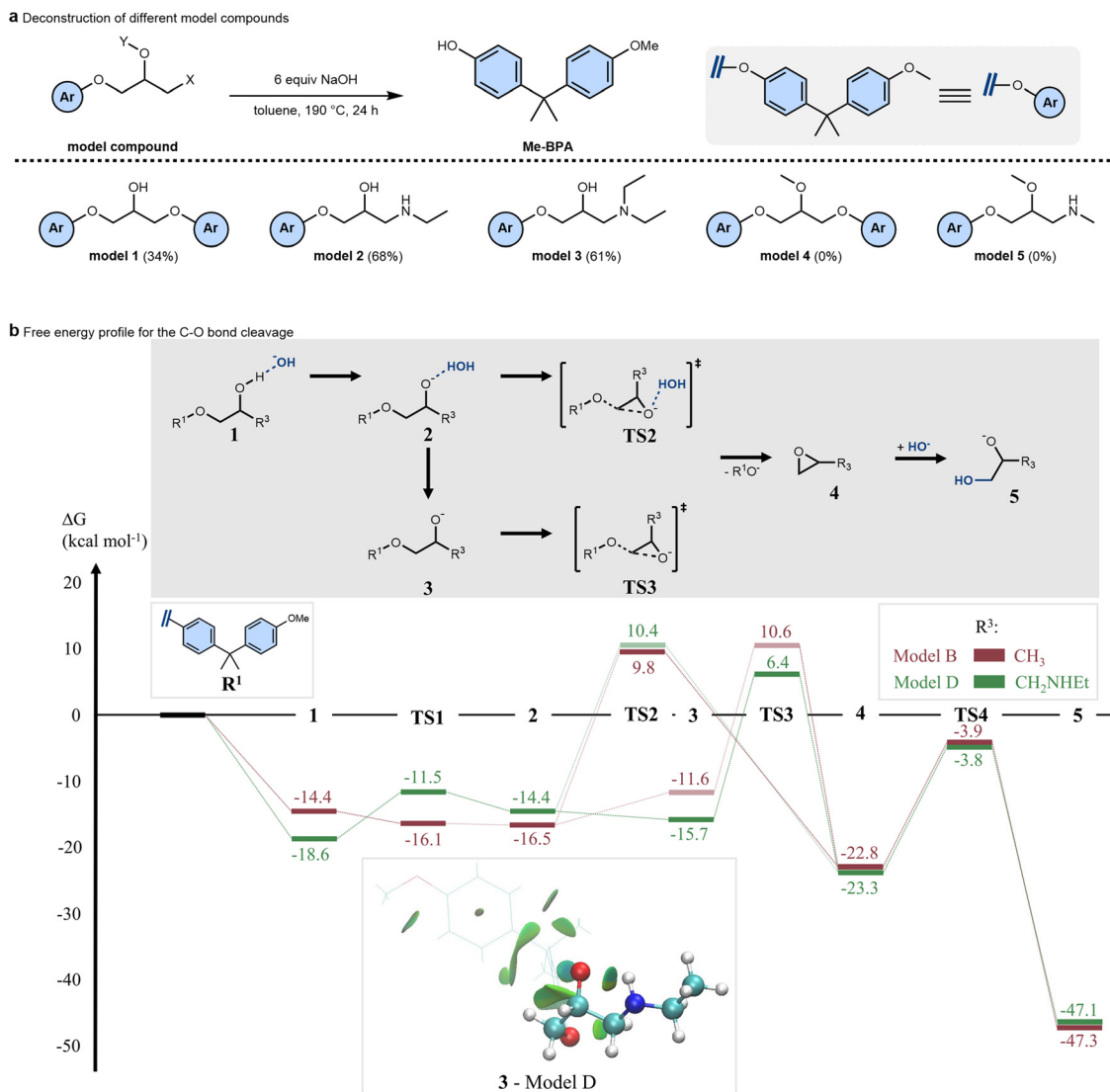
struction of wind turbine blades.<sup>16</sup> Each sample was submerged in 75% acetic acid at 70 °C overnight (Fig. 4a). Over the course of this time, the L-cystine-containing resin fragmented into a fine granule, while the recyclamine-based resin fragments dissolved, leaving behind the glass fibre mesh. The solution of the latter 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 recyclamine-based resin in 22 wt% and 15 wt%, respectively. Unfortunately, a yield in percentage cannot be reported, as the solvent absorbed within the thermoset matrix changes the mass to BPA ratio. For these resins designed for disassembly, experiments on a 10 times larger scale were conducted. 1 g of each material was reacted with NaOH in a steel autoclave fitted with a Teflon liner. Gratifyingly, BPA was recovered from both samples in comparable wt% to the small batch (Fig. 4b).

While this combinatory approach is specifically promising for tailored resins that allow easy separation of fibres and polymers, options for recovering values from the more widespread legacy materials can be considered. Recently, Ballout *et al.* reported a formic acid-based disassembly of aerospace grade composites using "legacy" epoxy resin.<sup>44</sup> Although the fibres

remain coated in the polymer, such a process could also be combined with the chemical deconstruction process demonstrated here. Nonetheless, the reuse perspective of the recovered fibres can be expected to strongly affect the economic and ecological feasibility of deconstructing composite materials. As such, tailored resins that allow the recovery of pure glass or carbon fibres should be easier to implement.

### 3. Elucidation of observed reactivity

Lastly, investigations were conducted to elucidate the mechanistic intricacies of the deconstruction process on a molecular level. We recognised that the base-induced bond scission could operate through epoxide intermediates analogously to alkaline-based lignin pulping processes.<sup>45</sup> However, there are considerable chemical and physical differences between thermoset epoxy resins and lignin.<sup>25</sup> For example, base-mediated pulping of the latter does not require solvent–base mismatch conditions.<sup>45</sup> 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**. While this hints at the amine



**Fig. 5** Mechanistic investigation of the deconstruction process. (a) Experimental investigation on different model compounds probing their scopes 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<sup>−1</sup>.

functionality easing the C–O bond cleavage, it has to be considered that **model 1** contains two cleavable C–O bonds. Therefore, the bond scission efficiency for **models 1, 2** and **3** is consistent with the molar amounts of released **Me-BPA** 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 an intramolecular nucleophilic attack to generate an epoxide with the simultaneous release of the phenoxide. In linkages containing amines, the lone pair on 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, implying that the formation of azetidines is probably not involved.

*In silico* investigations were undertaken, probing the free energy profiles of various mechanisms (see the ESI†). Simplified versions of **model 1** and **model 2**, denoted as 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<sup>−1</sup> 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 ( $\Delta G = 9.8 \text{ kcal mol}^{-1}$ ) is slightly preferred over TS3 ( $\Delta G = 10.6 \text{ kcal mol}^{-1}$ ) because of the hydrogen bonding with water. Instead, for model D, TS3 ( $\Delta G = 6.4 \text{ kcal mol}^{-1}$ ) becomes favoured over TS2 ( $\Delta G = 10.4 \text{ kcal mol}^{-1}$ ) due to the intra-molecular hydrogen bonding with the amine group, which is entropically preferred over 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 alcoholate **2** is only  $6.7 \text{ kcal mol}^{-1}$  (model B) and  $11.8 \text{ kcal mol}^{-1}$  (model D), respectively, a dynamic equilibrium between these species can be expected at  $190^\circ\text{C}$ . However, the nucleophilic attack of hydroxide on the epoxide **4** generates **5**, which is  $32.7 \text{ kcal mol}^{-1}$  (model B) and  $30.8 \text{ kcal mol}^{-1}$  (model D) more stable than **2**. Therefore, besides acting as a base, one equivalent of NaOH is consumed for every C–O bond cleaved. This in turn drives the thermodynamics of the reaction with the favourable formation of **5**, and explains the need for stoichiometric amounts of NaOH.

## 4. Conclusion

Disassembly processes for tailored epoxy composite materials, which enable circularity for their embedded fibres, have been reported and even commercialised.<sup>12–18</sup> 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 by 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 practices.

## Author contributions

Conceptualisation was carried out by HS, AA, AKR and TS. The experimental design was done 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 supervised by AN. Funding acquisition was carried out by TS. All authors reviewed the final manuscript.

## Methods and data availability

See the ESI† for further methods. The authors declare that all other data supporting the findings of this study are available within the paper and its ESI.†

## Conflicts of interest

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 the co-owner of SyTracks A/S, which commercialises the COTubes.

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