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Non-toxic degradation of anhydride-cured epoxy thermoset using a deep eutectic solvent

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Anhydride-cured epoxy thermosets, unparalleled in electrical insulation, mechanical strength and chemical stability, serve as indispensable materials in electrical engineering. Efficient degradation of epoxy resins is a critical key to sustainable resource utilization from decommissioned electrical equipment and to the green transformation of the electrical industry. However, existing degradation approaches often involve toxic catalysts and result in environmental pollution. We propose a non-toxic and efficient transesterification strategy employing a deep eutectic solvent (DES) composed of potassium carbonate (K_2CO_3) and ethylene glycol (EG). Compared to traditional organic base catalysis, inorganic K_2CO_3 offers advantages of non-toxicity, cost-effectiveness and sustainability. By leveraging the formation of activated carbonates as intermediates, the DES system can specifically cleave ester bonds, allowing for efficient degradation. Anhydride-cured epoxy thermosets are nearly completely degraded in DES containing 0.12 g mL^{-1} K_2CO_3 after 3 hours at $180\text{ }^\circ\text{C}$ under ambient pressure. In addition, the method facilitates the non-destructive and complete extraction of carbon fibers from the cured epoxy thermoset matrix. Catalysts can be partially recovered through a straightforward separation technique. Our work presents a non-toxic and genuinely green strategy for sustainable degradation and recycling of anhydride-cured epoxy resins, establishing a versatile platform that can be extended to diverse classes of thermosetting epoxy composites.

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1. Introduction

Epoxy resins are versatile thermosetting polymers celebrated for their exceptional mechanical strength, superior electrical insulation, robust adhesion and outstanding chemical resistance.^{1,2} These remarkable characteristics have established epoxy resins as indispensable materials in the field of electrical engineering.^{3,4} In particular, more than 90% of commercial epoxy formulations used in electrical and electronic applications are based on diglycidyl ether of bisphenol-A (DGEBA).^{5,6} However, once cured, these resins form a densely crosslinked, insoluble, and infusible network structure. This rigid structural network obstructs mechanical reprocessing, chemical degradation and efficient recycling at the end of their service life, imposing significant constraints on the sustainable utilization of epoxy resins. With growing concerns over the environmental impacts of conventional disposal strat-

egies for waste epoxy composites, approaches such as crushing for filler use, incineration and landfilling have come under increasingly strict scrutiny.⁷ Therefore, it remains a formidable challenge to find a sustainable degradation methodology with green recycling capabilities that can meet both environmental requirements and the urgent demand for efficient degradation.

Degradation strategies for epoxy resins have attracted much attention, including chemical degradation using strong acids,⁸ alkalis,⁹ heavy metals,¹⁰ and organic solvents,¹¹ as well as pyrolysis under elevated temperatures and pressures.¹² Although encouraging progress has been achieved using novel degradation mechanisms,^{13–15} most still rely on toxic catalysts or additives to promote the degradation process (*e.g.* the most commonly used organic catalysts 1,5,7-triazabicyclo[4.4.0]dec-5-ene and 1,8-diazabicyclo[5.4.0]undec-7-ene).¹⁶ The high toxicity of these catalysts persists in the products or waste liquids, significantly exacerbating post-treatment challenges and elevating processing costs. Moreover, they may pose substantial potential hazards to the environment upon disposal.

In this work, we propose a truly non-toxic and green deep eutectic solvent (DES) system to degrade anhydride-cured epoxy thermosets. DESs are eutectic mixtures of two or more components in defined stoichiometries, with melting points

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far below those of ideal liquid mixtures. Sustained by hydrogen bond donor and acceptor interactions, they exist as liquids.^{17–20} We selected the inorganic salt potassium carbonate (K_2CO_3) as the hydrogen bond acceptor (HBA) and ethylene glycol (EG) as the hydrogen bond donor (HBD). Although previous studies have shown that K_2CO_3 and EG can generate reactive alkoxy carbonates capable of effectively cleaving ester bonds in polyethylene terephthalate (PET),^{21,22} they have yet to be applied to epoxy resins because of their structural complexity and high crosslinking degree. Given the presence of similar functional groups in epoxy thermosets, this mechanism is likely applicable to transesterification degradation as well. Compared with other inorganic catalysts, K_2CO_3 is a low-cost, non-toxic substance widely used in water treatment and the food industry.^{23,24} EG can also be sourced from biomass sources, such as corn and lignocellulose, serving as a sustainable alternative to petrochemical feedstocks.^{25,26}

Experimental results demonstrate that DESs can effectively and rapidly degrade bulk anhydride-cured epoxy thermosets. Degradation can be completed within 3 hours at 180 °C at ordinary pressure when the molar ratio of potassium carbonate (K_2CO_3) to ethylene glycol (EG) is 1 : 20 (with a concentration of 0.12 g mL⁻¹). Furthermore, it enables 100% non-destructive recovery of carbon fibers from the cured epoxy thermoset matrix. The degradation solution can be recycled by adding ethanol to recover K_2CO_3 and by separating degradation products through water sedimentation, so as to achieve the recycling of the catalyst. This strategy demonstrates significant advantages in terms of simplicity of synthesis, cost-effectiveness, and non-toxicity. Moreover, it holds promise for extending to other types of cured epoxy resins, offering an efficient and feasible solution for the widespread problems in degradation and recycling.

2. Experimental

2.1. Materials

All reagents, including ethylene glycol (EG, Energy Chemical, 99%), potassium carbonate (K_2CO_3 , Macklin, AR, 99%), potassium phosphate tribasic (K_3PO_4 , Konosience, AR), bisphenol A diglycidyl ether (D.E.R. 331, Dow Chemical Company, epoxy value = 0.535 mol per 100 g), methyl tetrahydrophthalic anhydride (MeTHPA, Macklin, 95%), and *N,N*-dimethylbenzylamine (BDMA, Energy Chemical, 99%), were obtained commercially and used as received without any further purification. Degradation studies were conducted using systems based on conventional electrical-grade epoxy resin formulations. The carbon fibers were purchased from Jinhong Adhesive Co., Ltd.

2.2. General characterization

Proton nuclear magnetic resonance (¹H NMR) characterization was performed by dissolving the sample in deuterated DMSO-*d*₆ and analyzing it with a JEOL JNM-ECA400 (400 MHz) spectrometer. Fourier-transform infrared (FT-IR) analysis was conducted using a Spectrum 100 FT-IR spectrometer, with a

scanning range of 4000–450 cm⁻¹ for individual monomers or mixtures. Differential scanning calorimetry (DSC) analysis was carried out using TA-Q2000 equipment with a test temperature range of -50 to 180 °C. The temperature was initially raised to 180 °C to eliminate any thermal history, followed by qualitative testing at a heating rate of 10 °C min⁻¹. The morphologies of the carbon fibers were examined by scanning electron microscopy (SEM, JSM-5900 LV) at an accelerating voltage of 15 kV. All samples were sputter-coated with gold.

2.3. Preparations of DESs

Solid potassium carbonate (HBA) and ethylene glycol (HBD) solvent were mixed in a molar ratio of 1 : 20 at a concentration of 0.12 g mL⁻¹ and added to a 250 mL round-bottom flask. The mixture was then stirred and heated in an oil bath at 100 °C for 10 minutes, until the solid was completely dissolved, yielding a uniform and transparent deep eutectic solvent (DES) degradation solution. Experiments show that at a DES molar ratio of 1 : 8 (0.31 g mL⁻¹), K_2CO_3 fails to fully dissolve within a short time at 100 °C. As the temperature increases, the solution gradually turns yellow, ultimately compromising the degradation efficiency. In contrast, the 1 : 10 ratio (0.25 g mL⁻¹) retains long-term clarity and stability at the same temperature and is therefore designated as the upper concentration limit. Alternative synthesis methods varied only in the molar ratio of EG to K_2CO_3 , resulting in variations in the concentrations of the corresponding degradation solutions. A similar DES system of potassium phosphate tribasic (K_3PO_4) and ethylene glycol (EG) was prepared using the same method, with a concentration of 0.12 g mL⁻¹ as a control.

2.4. Degradation and recycling of epoxy resin

Initially, degradation test samples were prepared according to a conventional electrical resin formulation. Specifically, 15 g (40.12 mmol) of D.E.R. 331, 13.4 g (80.24 mmol) of MeTHPA, and 0.27 g of BDMA were thoroughly mixed under stirring. The resulting mixture was poured into a polytetrafluoroethylene (PTFE) mold and subjected to a two-stage curing process: 4 h at 80 °C followed by 12 h at 140 °C, yielding solid epoxy resin specimens. In addition, carbon fiber-reinforced epoxy composites were fabricated to facilitate subsequent comparative studies on the nondestructive recovery of carbon fibers. The self-prepared epoxy resin samples were cut into cubes measuring 10 × 10 × 3.9 mm³. A degradation solution with a 5 wt% solid content was added to the resin samples, which were then placed in a 30 mL glass bottle. Degradation was performed in an oil bath at various temperatures. Every 30 min, the resin samples were removed, the surface solution was wiped off, and the remaining mass was weighed. The color of the degradation solution deepened with increasing temperature and time, and after 3 hours, the degradation was nearly complete.

Upon completion of degradation, 20 mL of the resulting mixture was transferred to a round-bottom flask containing 2.4783 g of K_2CO_3 . Subsequently, five volumes of ethanol were added, causing the mixture to turn light brown. The suspension was then centrifuged, precipitated, and dried to yield

1.352 g of white K_2CO_3 precipitate, corresponding to a yield of approximately 54.55%. The supernatant was then concentrated by rotary evaporation to remove the solvents, followed by the addition of five volumes of water. Upon stirring and standing, the degradation products precipitated as a flocculent solid. The degradation experiment was conducted on 2.6524 g of epoxy resin, yielding 1.5628 g of product after purification. The purification process involved water precipitation, washing three times with acetone, and drying overnight under vacuum at 60 °C. Quantitative analysis of the isolated degradation derivatives revealed a yield of 0.5892 g per gram of epoxy resin, corresponding to 59%. The mechanism of the recycling of the epoxy thermoset *via* DES-assisted degradation is depicted in Fig. 1. The remaining solution was retained and reused as the DES medium for subsequent degradation cycles. The lower monomer precipitates were dried in a vacuum oven at 60 °C, and the product structure was characterized by 1H NMR.

3. Results and discussion

3.1. Characterization of the DESs

To systematically characterize the chemical structure and phase transition behavior of the samples, Fourier-transform infrared (FTIR) spectroscopy was first employed to analyze the functional groups, thereby confirming the successful incorporation of target moieties and monitoring structural changes. As shown in Fig. 2(a), the FTIR spectrum reveals that the $-OH$ absorption peak of the DES broadens and shifts to a lower wavenumber, indicating the formation of hydrogen bonds between K_2CO_3 and EG in the eutectic solvent. This phenomenon is widely regarded as the primary driving force behind the formation of the eutectic structure within the system.^{21,27} The formation of hydrogen bonds reduces the vibrational frequency of chemical bonds, causing the absorption peak to shift towards lower wavenumbers. In the K_2CO_3 -EG DES system, a new band appears at 1645 cm^{-1} , which is attributed to the asymmetric stretching mode of the $C=O$ in the $-OCOO-$ group, indicating the formation of ethylene glycol carbonate. This provides further evidence for the successful preparation of the DES. The increased number and diversity of hydrogen bonding interactions within the DES system decrease the

selectivity of molecular recognition. In addition, the charge delocalization resulting from hydrogen bond interactions also reduces the lattice energy of the system, thus lowering the freezing point of the mixture.

Differential scanning calorimetry (DSC) analysis reveals the thermal transitions of the material during the heating process, providing insights into its glass transition temperature (T_g), melting behavior, and crystallization characteristics. As illustrated in Fig. 2(b), the DSC curve of the transparent and homogeneous DES solution indicates no discernible melting transitions between -50 and 180 °C, suggesting the absence of a well-defined crystalline phase. However, a glass transition is observed at -34.6 °C, well below room temperature. The melting points of potassium carbonate ($T_m = 891$ °C) and ethylene glycol ($T_m = -15$ °C) are significantly lower than those of the two components involved in forming the DES,²⁸ further confirming the successful preparation of the DES.

3.2. Degradation performance analysis

The degradation process of a thermosetting epoxy resin is complex, involving multiple chemical reaction pathways and mechanisms. During heating, the epoxy resin may degrade through several routes, including the ring-opening of epoxy groups, polymer chain cleavage, and the pyrolysis of side groups. Such reactions can lead to a decrease in molecular weight, modifications in the crosslinked structure, and the generation of volatile low-molecular-weight compounds, all of which adversely impact the thermal stability and mechanical performance of the material.^{29,30}

To investigate the degradation performance of bulk epoxy resins in specific DES solutions, a dynamic insulation epoxy thermoset was prepared using bisphenol A-type diglycidyl ether resin (D.E.R. 331) as the epoxy precursor, methyl tetrahydrophthalic anhydride (MeTHPA) as the curing agent, and *N,N*-dimethylbenzylamine (BDMA) as the accelerator. The molecular structures are illustrated in Fig. 3(a). This system served as a model to evaluate the degradation efficiency and behavior of the resin in green DES solutions. It aims to provide a scientific theoretical basis for the recycling of epoxy resins used in insulating transformers and wind turbine blades. Initially, the anhydride-cured epoxy polymers were immersed

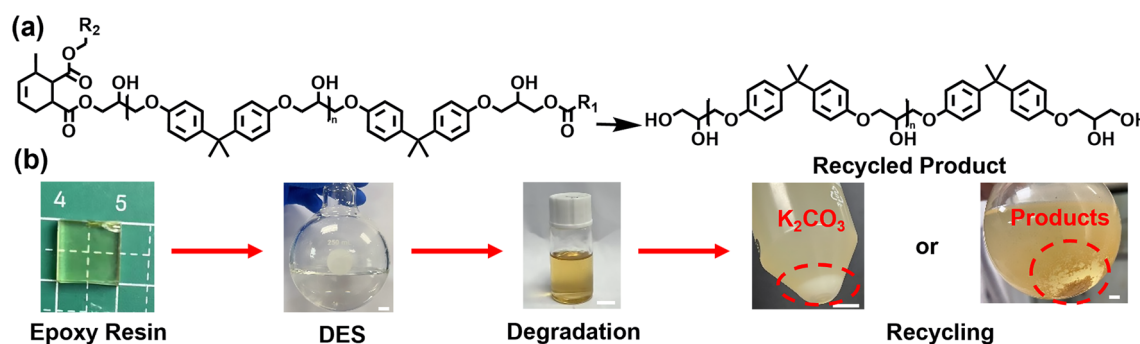


Fig. 1 (a) Molecular structure during the degradation process; (b) the physical images corresponding to the degradation process. Scale bar: 1 cm.

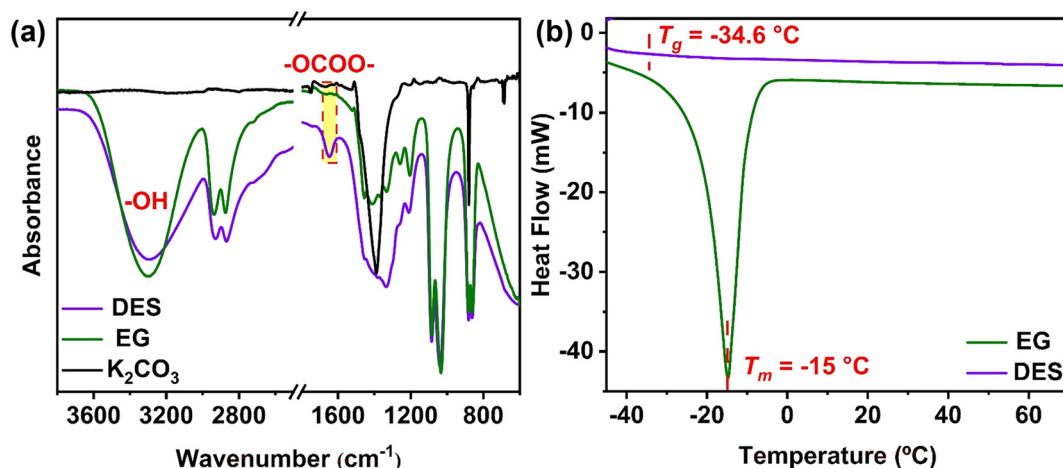


Fig. 2 FI-TR spectra (a) and DSC thermograms (b) of the DESs.

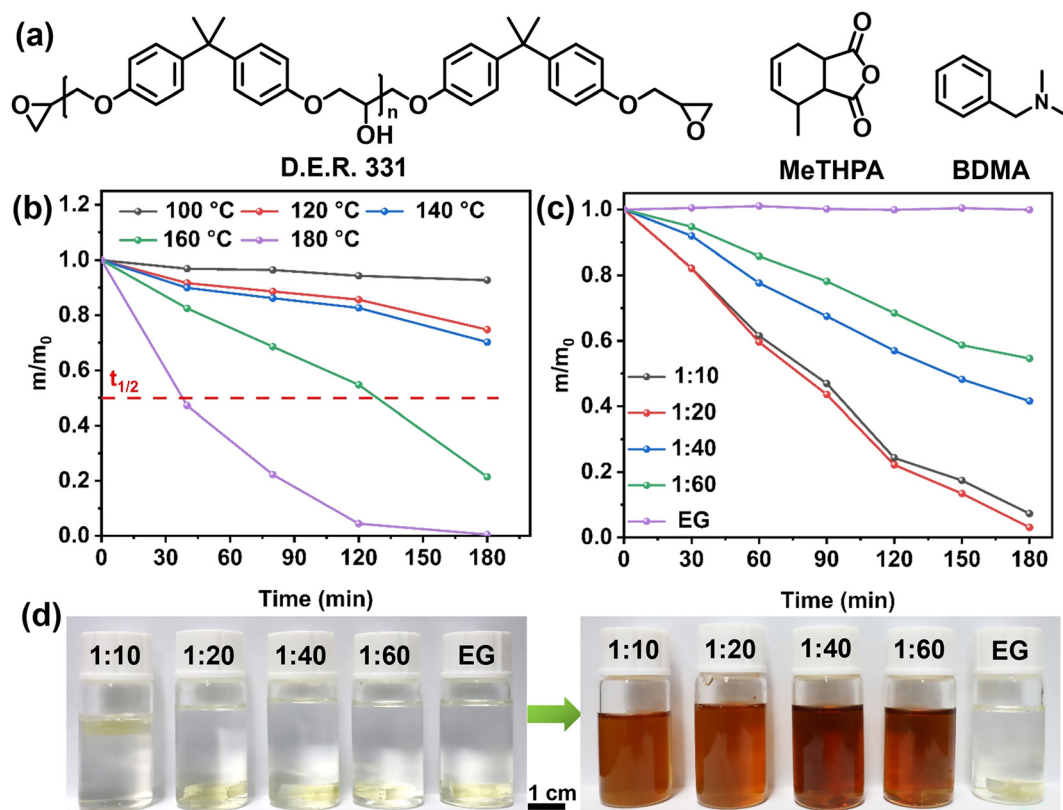


Fig. 3 (a) Molecular formulae for epoxy degradation formulations; normalized mass vs. time for epoxy degradation at different temperatures (b) and varying DES concentrations (c); and (d) photographs of the solutions before and after degradation.

in a K_2CO_3 -EG based DES system and subjected to degradation at set gradient temperatures of 100 $^{\circ}\text{C}$, 120 $^{\circ}\text{C}$, 140 $^{\circ}\text{C}$, 160 $^{\circ}\text{C}$, and 180 $^{\circ}\text{C}$. The mass loss of the residual samples was recorded at fixed time intervals until complete degradation was achieved. Fig. 3(b) shows the variation in normalized mass of the epoxy sample over time. The degradation curves exhibit distinct rates of mass loss at different temperatures, with

higher temperatures leading to more rapid degradation. This trend indicates that the reaction rate accelerates as the temperature increases. Fig. 3(b) also highlights $t_{1/2}$, the half-life, which represents the time required for the sample mass to decrease to half of its initial value. As the temperature rises, the half-life is markedly shortened. Among the tested conditions, 180 $^{\circ}\text{C}$ was identified as the optimal degradation

temperature, under which nearly complete degradation occurred within 3 h. Furthermore, a comparative analysis of degradation rates was conducted at 180 °C using DES with varying molar ratios (1 : 10, 1 : 20, 1 : 40, and 1 : 60), as well as pure EG as a control. Fig. 3(c) reveals that the most effective degradation was obtained at a molar ratio of 1 : 20, corresponding to a DES concentration of 0.12 g mL⁻¹. This finding indicates that both temperature and DES composition play critical roles in facilitating the efficient cleavage of the epoxy network. In contrast, under pure EG conditions, the sample exhibited virtually no mass change, indicating that EG alone is incapable of degrading the epoxy resin. Fig. 3(d) presents the state of the samples before and after the reaction. Owing to the high density of the 1 : 10 DES formulation, the bulk epoxy resin remained suspended at the surface of the degradation solution. The pre-reaction samples were all transparent liquids, while after the reaction, all except the EG sample turned into brown liquids. As degradation proceeded, the bulk epoxy resin gradually swelled and softened, eventually breaking down completely.

To assess the universality of DESs in epoxy resin degradation, we explored an alternative inorganic salt system, guided by mechanistic insights into hydrogen-bonding interactions. The primary role of potassium carbonate (K₂CO₃) in the DES system is to provide an alkaline environment that facilitates deprotonation or nucleophilic attack on the epoxy network, thereby accelerating transesterification degradation. Among inorganic salts, potassium phosphate tribasic (K₃PO₄) exhibits stronger basicity than K₂CO₃. Upon hydrolysis, K₃PO₄ generates phosphate ions (PO₄³⁻), which subsequently produce hydroxide ions (OH⁻), further making the solution alkaline.³¹ Based on this rationale, a K₃PO₄-EG DES system was prepared at the same concentration (0.12 g mL⁻¹), and degradation experiments were conducted on the same electrical-grade epoxy resin system at the previously determined optimal temperature of 180 °C. To ensure the accuracy and reproducibility of the experimental outcomes, all reaction parameters were carefully regulated under identical conditions. By comparing the degradation efficiencies of epoxy resins under

different DES systems, as shown in Fig. 4(a), it was observed that the K₃PO₄-EG system degraded less than 50% of the resin within 2 h, indicating limited degradation capability under these conditions. The core degradation mechanism involves the synergistic effect of selective ester bond cleavage and solvation. Strong hydrogen-bonding interactions lead to high viscosity in the K₃PO₄-EG system, thereby impeding molecular mobility. In contrast, the lower viscosity of the K₂CO₃-EG system not only favors the reaction but also more effectively promotes epoxy resin swelling, thereby enhancing degradation efficiency.³²

Subsequently, the precipitates obtained from the stepwise water treatment were analyzed by ¹H NMR spectroscopy, as shown in Fig. 4(b). The main products were identified as D.E.R. 331 derivatives with terminal hydroxyl groups, consistent with ester bond cleavage during degradation. The ¹H NMR spectral data of the D.E.R. 331 tetra-alcohol derivative (400 MHz, DMSO-*d*₆) are as follows: δ ppm: 7.06–6.76 (–Ar, 4H), 3.91–3.88 (–CH, 1H), 3.78–3.70 (–CH₂, 2H), 3.39–3.35 (–CH₂, 2H), 1.56–1.47 (–CH₃, 3H). Relevant reports have indicated that this tetra-alcohol is insoluble in water, while the anhydride-derived diester and the accelerator BDMA are highly water soluble.^{33,34} The efficient degradation by the DES arises from three synergistic factors: structural characteristics, active intermediate generation, and epoxy network penetration. Specifically, the DES's alkaline environment provides fundamental degradation thermodynamics. Reaction-generated activated carbonates enable specific ester bond cleavage to boost degradation efficiency. Higher temperature and DES concentration accelerate transesterification and enhance degradation kinetics. To validate the precision of ester bond cleavage, the FTIR spectra of the anhydride-cured thermoset, the raw D.E.R. 331, and its degradation product were compared. The recycled derivatives showed no characteristic anhydride ester peak (–COO–) at 1735 cm⁻¹ (Fig. S1). The recycled degradation products also exhibited a structure similar to the raw D.E.R. 331, along with more prominent –OH (3318 cm⁻¹) and C–O–C (1232 cm⁻¹) stretching vibrations.³⁵ These results also support the proposed mechanism.

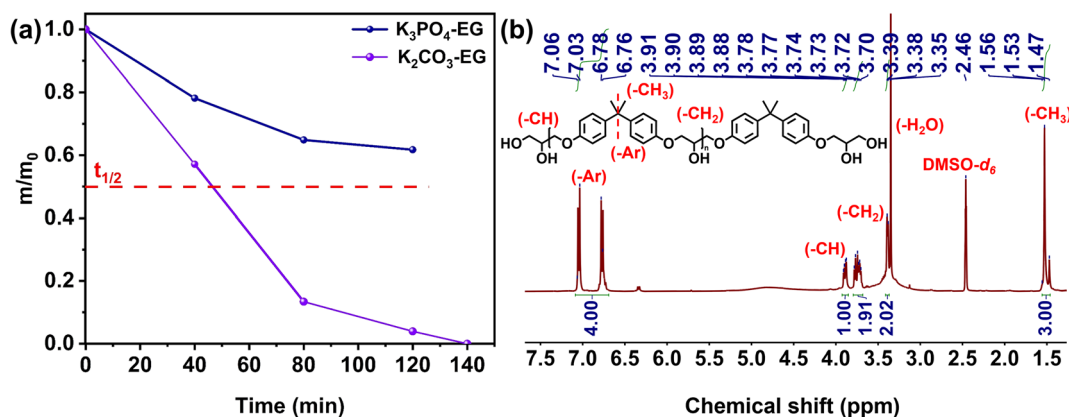


Fig. 4 (a) Comparison of the degradation rates of the two DESs and (b) ¹H NMR spectrum of the degradation products.

3.3. Investigation of the degradation mechanism

Polymer composites inevitably face disposal at the end of their service life in practical applications. A comprehensive understanding of the degradation mechanisms is crucial for optimizing degradation conditions, enhancing efficiency, and mitigating the negative environmental impact. In the crosslinked network of epoxy resins, the primary functional groups include ester bonds, ether bonds, and aromatic rings.^{36–38} Due to the inherent stability of the aromatic ring structure, the cleavage of ether bonds typically requires elevated temperatures or highly alkaline environments. Consequently, during the degradation process, ester bonds are preferentially cleaved, with the C–O bonds undergoing transformation into carboxylate and hydroxyl groups.³⁹

In our degradation system, the polar molecules of the deep eutectic solvent (DES) initially infiltrate the crosslinked network, inducing swelling and thereby facilitating the cleavage of crosslinking points. Concurrently, the basic components of the DES trigger hydrolysis or transesterification of reversible bonds within the crosslinked structure, as illustrated in Fig. 5(a), leading to the scission of the epoxy resin backbone. This process results in the relaxation and decomposition of the overall network, yielding recoverable monomers or oligomers. As summarized in Table 1, the DES system demonstrates clear advantages over traditional catalysts across all major criteria: catalytic activity, environmental benignity, cost-effectiveness, recyclability, and process safety. To evaluate the recyclability of this green degradation medium, we applied the system to epoxy-based carbon fiber reinforced composites (CFRPs) fabricated using an electrical-grade epoxy formu-

Table 1 Multidimensional comparison of catalysts for epoxy thermoset degradation

Catalyst	Cost (¥ per kg)	Toxicity/safety	Recyclability
TBD ⁴⁰	High (28 000)	Corrosive, irritant	Poor
DBU ¹⁶	Moderate (294)	Irritant	Moderate
Zinc acetate ⁴¹	Moderate (75–250)	Moderate, metal salt	Moderate
PPh ₃ ⁴²	High (200–450)	Toxic, air-sensitive	Poor
K ₂ CO ₃ ^a	Low (8–15)	Non-toxic	Good

Superscript numbers indicate literature references. ^a This work, (¥ per kg): median price per kg (¥); the data are derived from ChemicalBook.

lation. The process enabled 100% non-destructive recovery of carbon fibers, along with successful reclamation of both the catalyst and solvent, as outlined in Fig. 5(b). This study highlights the DES system's efficient penetration and bond cleavage in complex matrices and its potential for sustainable material recycling. Meanwhile, scanning electron microscopy (SEM) observations were made on pristine and recycled carbon fibers (CFs) (Fig. S2). The surfaces of both pristine and recycled fibers were generally smooth, continuous, and free from adherent impurities. Recycled carbon fibers showed slight roughness, yet no etching pits, delamination, or cracks were detected. Nevertheless, the minuscule dimensions of carbon fibers make direct mechanical testing challenging. Mechanical tests were conducted on fifty pristine and fifty recycled carbon fibers (diameter: 7 µm). From the stress–strain curves measured at room temperature, the tensile stress of the pristine fibers was 1280 MPa, decreasing to 812 MPa after re-

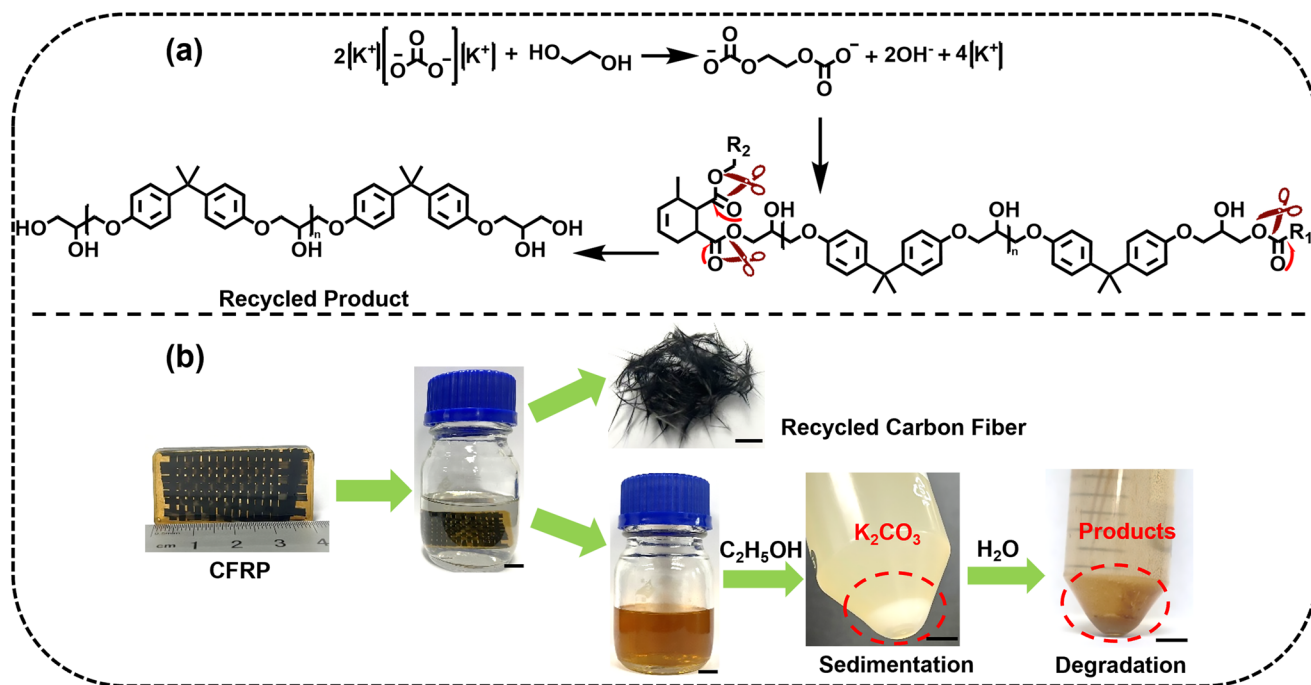


Fig. 5 (a) Schematic diagram of ester bond cleavage and (b) nondestructive recovery of carbon fibers from the composite materials. Scale bars: 1 cm.

cycling (Fig. S3). We propose that the 36.6% loss in the tensile strength of carbon fibers arises from damage induced by mechanical agitation during degradation, thereby affecting the fibers' overall strength. This issue could be avoided by isolating the material from the stir bar.

4. Conclusions and outlook

This study demonstrates that the K_2CO_3 -EG DES system offers a non-toxic, green, economically viable strategy for the degradation and recycling of waste electrical epoxy thermosets. By leveraging the strong nucleophilicity and hydration properties of the alkoxy organic carbonates formed within the DES system, we successfully recovered high-value monomers and catalysts, as well as achieved 100% non-destructive recycling of carbon fibers from the composites. This further highlights the significant potential of DESs as ideal alternatives to traditional chemical or solvent-based degradation processes in industrial applications. Moving forward, the degradation products can be utilized as fillers in the synthesis of polycarbonates, polyesters, and other materials, or developed into functional additives (such as crosslinking agents, flame retardants, and plasticizers) to be incorporated into resins, thereby achieving closed-loop recycling and material reuse. Notably, regenerated polyols can be engineered into high-performance materials through chemical functionalization or co-polymerization/blending with bio-based polyols, exhibiting enhanced flame retardancy, durability, and energy dissipation. Such valorization of recycled products not only establishes them as strategic resources but also substantially improves the economic and environmental sustainability of epoxy resin chemical recycling.

Author contributions

H. L. and W. T. conceived the core idea of this study. W. T. conducted the majority of the experiments, with E. H. providing valuable assistance in the experimental process. H. L. and W. T. wrote the manuscript draft. All authors engaged in in-depth discussions on the results. Y. W. reviewed the manuscript and made critical revisions to enhance its quality.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

All data analyzed in this study are included in this article and can be obtained from the corresponding author.

The Supporting information (SI) includes the FTIR spectra of the pristine D.E.R. 331 resin and its degradation products, as well as the comparative data graphs of the morphology and

mechanical properties of the pristine and recycled carbon fibers. See DOI: <https://doi.org/10.1039/d5py00829h>.

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