Progress in Structural Design and Multifunction of Bio-based Epoxy Resin Composites Containing Dynamic Bonds

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Abstract: Epoxy thermosets are widely used in coatings, adhesives, and composites because of their excellent mechanical strength, chemical resistance, and dimensional stability. However, their petroleum-based origin and permanent crosslinking raise concerns regarding environmental impact and human health. Bio-based epoxy systems and dynamic covalent chemistry (DCC) have therefore emerged as promising strategies to achieve sustainability and recyclability. Although many studies have explored dynamic epoxy networks, most reviews emphasize resin-level properties rather than their behavior in composite systems. This review aims to provide a focused and comprehensive overview of bio-based epoxy composites containing dynamic bonds, clarifying how dynamic exchange mechanisms influence molecular design and multifunctional performance. Key findings highlight the synergistic roles of different dynamic chemistries in enabling reprocessability, self-healing, recyclability, and enhanced structural robustness. By identifying structure–property relationships across various composite strategies, this review fills a critical gap in the literature and offers guidance for the rational design of next-generation biodegradable epoxy composites.

Keywords: Bio-based epoxy, Dynamic covalent chemistry, Dynamic bond, Composite, Recyclability, Self-healing capability, Multifunction.

1. INTRODUCTION

Epoxy resins are three-dimensional network polymers formed by the crosslinking reaction between organic compounds containing epoxy groups and curing agents bearing active hydrogen. The development of epoxy resins dates back to the 1930s, with large-scale industrial production established by the late 1940s [1]. Conventional epoxy resins exhibit outstanding dimensional and thermal stability, high mechanical strength, excellent creep resistance, superior electrical insulation, and strong chemical resistance, establishing them as indispensable materials in protective coatings, adhesives, construction, advanced composites, electrical engineering, and electronic packaging [2–7]. In 2023, the global epoxy resin market was valued at USD 11.25 billion and is expected to reach USD 16.87 billion by 2030, with a compound annual growth rate (CAGR) of 6.3% from 2024 to 2030 [8]. The global epoxy resin market is showing steady growth, with continuous optimization of production capacity and expansion into emerging application areas.

At present, the majority of commercial epoxy resins are synthesized from non-renewable petroleum-based feedstocks, primarily bisphenol A (BPA) and epichlorohydrin (ECH). BPA is widely recognized as a metabolic disruptor detrimental to human health, and its inherent toxicity and sustainability concerns have attracted increasing global attention [9, 10]. To overcome these limitations, researchers have turned their attention to bio-based epoxy resins derived from renewable resources. Bio-based polymeric materials, derived from renewable feedstocks including lignin, eugenol, vanillin, cardanol, and tannins, offer distinct advantages such as environmental protection and reduced fossil fuel dependence. They represent promising alternatives to petroleum-based polymers in line with global sustainability goals [11–15]. Despite this progress, thermoplastic materials dominate the bio-based polymer market, whereas bio-based thermosets remain a relatively small segment [16]. This is because traditional thermosets form crosslinked networks that are insoluble and infusible, making thermal processing and recycling extremely difficult or even impossible. Thus, the development of bio-based epoxy resins faces substantial environmental burdens and end-of-life management challenges.

In recent years, the emergence of dynamic covalent chemistry (DCC) has offered a new approach to addressing this issue. By introducing reversible covalent bonds—such as transesterification, imine, disulfide, boronic ester, and phosphate ester linkages—the network structure can rearrange under external stimuli like heat, humidity, or chemical environments, thereby imparting innovative recyclability to the materials. The incorporation of such dynamic linkages enables thermosetting epoxy networks to decompose into monomers or oligomers under mild conditions. These depolymerized

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intermediates can be reprocessed through techniques such as extrusion or injection molding, allowing multiple reuse cycles [17-21]. Therefore, integrating dynamic bonds with bio-based resources represents an ideal strategy for sustainable material design. The development of bio-based epoxy resins containing dynamic covalent bonds not only prolongs material lifespan and reduces plastic waste but also decreases reliance on non-renewable feedstocks, thereby mitigating carbon emissions.

Building upon this foundation, researchers have progressively expanded dynamic network design into composite material systems. Introducing dynamic bonds within the resin matrix, interfacial region, or reinforcing phase markedly enhances filler dispersion and interfacial bonding strength, endowing materials with self-healing, reprocessable, and even recyclable capabilities. For instance, imine and boronic ester exchange systems achieve interfacial self-healing and reuse through chemical reconfiguration [22]. Meanwhile, the nanofillers (*e.g.*, graphene, MXene, nanocellulose) not only enhances mechanical and thermal performance but also enables multifunctionality such as conductivity, flame retardancy, and antibacterial activity [23]. Therefore, epoxy resin composites containing dynamic bonds have emerged as a key research direction integrating high performance with sustainability, where molecular designability and network reversibility provide strong support for future smart materials and circular economy.

Existing reviews on dynamic bond–containing epoxy resins have predominantly centered on the reprocessability, recyclability, and degradability endowed by dynamic covalent chemistry. However, comprehensive and systematic analyses of their applications in composite systems remain scarce. Most previous reviews discuss dynamic epoxy networks at the monomer or resin level, but do not integrate how dynamic exchange mechanisms influence composite architecture, interfacial interactions, filler integration, and multifunctional behavior. Moreover, no existing review provides an integrated perspective linking molecular design, network topology, dynamic mechanisms, and composite reinforcement strategies. This lack of composite-focused understanding limits the development of recyclable and high-performance bio-based epoxy materials.

Addressing this gap, the present review provides the first focused overview of bio-based epoxy composites incorporating dynamic covalent bonds. This review summarizes recent advances in epoxy resin composites containing dynamic bonds, systematically discussing molecular design strategies, structure–property relationships, and multifunctional developments. It also highlights the roles of various dynamic chemical mechanisms in improving the reconfigurability and sustainability of composites. Finally, the review outlines the key challenges and future perspectives of this emerging field, aiming to offer theoretical insights and technological guidance for the rational design of high-performance, recyclable, and intelligent epoxy-based composite materials.

2. LITERATURE COLLECTION AND REVIEW METHODOLOGY

To ensure the accuracy and reproducibility of the analysis presented in this review, a systematic literature collection strategy was adopted. Relevant publications were retrieved from Web of Science, Scopus, ScienceDirect, and Google Scholar using combinations of the following keywords: bio-based epoxy, dynamic covalent chemistry, vitrimer, dynamic bond, epoxy composite, recyclable composites, renewable monomers, and self-healing epoxy. The literature search covered the period from 2010 to 2025, with a focus on recent advancements reported after 2018.

Studies were included if they met the following criteria:

1. They involved bio-based epoxy systems or renewable curing agents;
2. They incorporated dynamic covalent bonds such as transesterification, imine, disulfide, boronic ester, or acetal linkages;
3. They investigated composite systems involving fibers, nanoparticles, or multifunctional fillers;
4. They provided quantitative or qualitative evaluations of recyclability, reprocessability, mechanical performance, or multifunctionality.

Duplicate records, non-peer-reviewed materials, and studies lacking sufficient detail on dynamic mechanisms or composite behavior were excluded. This structured methodology ensures that the review reflects a comprehensive, balanced, and reproducible analysis of current research on bio-based epoxy composites featuring dynamic covalent bonds.

3. MOLECULAR DESIGN STRATEGIES FOR DYNAMIC EPOXY NETWORKS

In recent years, dynamic covalent interactions have garnered growing scientific interest. Their bond strengths fall between those of strong covalent bonds and weak noncovalent interactions, allowing reversible breaking and reforming under equilibrium conditions

[24]. Dynamic covalent bonds are a typical type of dynamic interactions. The concept of dynamic covalent chemistry, first introduced by Lehn, centers on the reversible formation and cleavage of covalent linkages under mild conditions [25]. Later, Bowman *et al*. developed crosslinked polymer networks containing reversible bonds and coined the term covalent adaptive network (CAN) to describe such systems [26]. Dynamic covalent bonds can undergo reversible dissociation and reformation in response to external stimuli such as temperature, light, pH, or chemical agents. This reversible behavior endows materials with self-healing, adaptive, and reprocessable properties while retaining the structural integrity and strength of covalent networks [27].

3.1. Dynamic-Bond Placement and Network Architecture

The introduction of dynamic bonds into epoxy systems mainly follows two molecular design strategies: epoxy monomers containing dynamic bonds (Figure **1**(**a**)) or curing agents containing dynamic bonds (Figure **1**(**b**)). For example, Ding *et al*. [28] synthesized a novel tetrafunctional Schiff-base epoxy monomer (PTMSi) from natural aromatic protocatechualdehyde and synthetic siloxane amine. The dynamic covalent epoxy thermoset obtained by curing with DDM showed excellent thermal stability, mechanical strength, and modulus. Jiang *et al*. [29] prepared a bio-based monomer (VAN-AC-EP) via a Schiff-base condensation between vanillin and 4-aminocyclohexanol, which was subsequently cured using DDM. The resulting thermoset displayed favorable thermomechanical performance, and the presence of dynamic imine linkages imparted degradability, recyclability, and excellent reprocessability to the resin.

At the same time, researchers have been actively investigating and designing curing agents incorporating reversible dynamic linkages. Toendepi *et al*. [30] synthesized three amine compounds containing different acetal-type dynamic covalent bonds (acetal, formal, and ketal linkages), and employed them as curing agents for epoxy resins. Owing to the reversible nature of these acetal linkages, all cured epoxy networks displayed significant stress relaxation at high temperatures and could be readily reprocessed through hot pressing. Kubota *et al*. [31] prepared an imine-containing phenolic curing agent by condensing vanillin with primary diamines. After three successive healing cycles conducted at 100 °C under 5 MPa for 2 h each, the repaired epoxy networks achieved a healing efficiency (η_σ) as high as 97%. Moreover, the reduction in η_σ after multiple healing cycles was minimal, indicating high structural retention. Such epoxy networks hold significant promise for sustainable coating materials and small-scale components that demand exceptional self-healing capabilities.

In the two molecular design routes described above, dynamic bonds are either intrinsically incorporated into the epoxy monomer or integrated within the backbone of the curing agent. It is well established that the ratio and spatial distribution of dynamic linkages within a

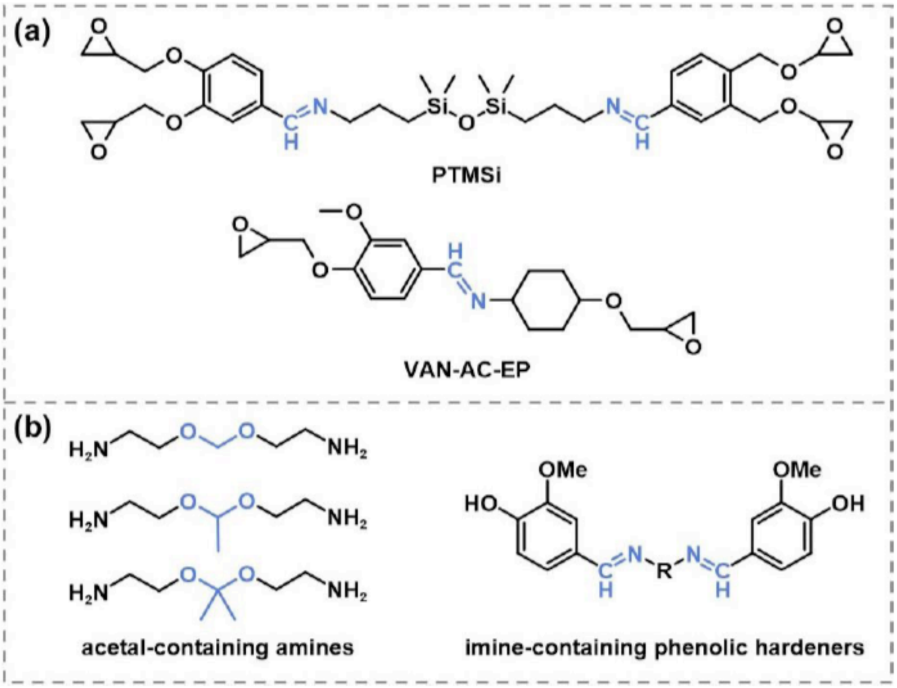

Figure 1: Chemical structure of epoxy monomers containing imine bonds (**a**) and curing agents containing acetal or imine bonds (**b**).

polymer network play a decisive role in governing its mechanical performance and overall dynamic behavior. Fanlo *et al*. [32] explored this concept by designing three systems in which the position of aromatic disulfides—located in the epoxy resin, the amine, or both—was systematically varied, while precisely tuning the proportion of dynamic bonds. The results indicated that conventional epoxy vitrimer formulations using aromatic amines with disulfide linkages failed to achieve full network relaxation in the absence of excess amine, owing to homopolymerization leading to non-dynamic crosslink formation. In contrast, incorporating aromatic disulfide moieties into the epoxy monomer facilitated full network relaxation, thereby maintaining recyclability and reprocessability. Nevertheless, an excessive content of aromatic disulfide linkages was found to markedly deteriorate the mechanical strength of the materials. Ultimately, the optimal formulation was achieved by partially substituting the disulfide-containing diepoxies (BGPDS) with a non-dynamic epoxy such as DGEBA—comprising 32 mol% DGEBA, 68 mol% BGPDS, and 1.0 equivalent of 4,4′-methylenedianiline (MDA). This optimized formulation simultaneously enhanced mechanical performance and reduced production cost, while preserving key dynamic functionalities including self-healing, reprocessability, and recyclability.

3.2. Multi-Dynamic Synergy

Recent advances in bio-based epoxy vitrimer have highlighted the power of multi-dynamic covalent bond design in achieving simultaneous improvements in mechanical strength, reprocessability, and recyclability. By incorporating more than one type of reversible bond within a single network, researchers have enabled cooperative exchange reactions that accelerate topological rearrangement while maintaining the intrinsic rigidity of thermoset structures.

For instance, Zhou *et al*. [33] reported a fully bio-based dynamic covalent–supramolecular epoxy resin derived from furfural and levulinic acid platform chemicals. The molecular design integrated dynamic ester bonds and hydrogen-bonding interactions within a single cross-linked network, as shown in Figure **2**(**a**). The tetrafunctional epoxy monomer (FDEP), obtained via condensation of levulinic acid and furfurylamine, was cured with 2,5-furandicarboxylic acid (FDCA) to form an ester-based network strengthened by abundant supramolecular hydrogen bonding (FDAA). The dynamic transesterification endowed the network with topological reconfigurability and chemical recyclability, whereas hydrogen bonding reinforced chain packing and energy dissipation, yielding a high glass transition temperature (T_g) (164 °C) and impressive tensile strength (111 MPa). This catalyst-free design achieved closed-loop recycling under alkaline conditions, highlighting the green molecular engineering potential of ester–hydrogen-bond synergistic networks.

Dong *et al*. [34] developed a vanillin-derived dual-dynamic epoxy network integrating imine (C═N) and disulfide (S–S) linkages, as shown in Figure **2**(**b**). The dual-dynamic crosslinked network was formed via a Schiff-base condensation between vanillin and aromatic diamines coupled with a thiol–epoxy click reaction. The imine bonds enabled rapid topological rearrangement and self-healing, while the disulfide bonds further facilitated bond exchange and closed-loop recycling under thermal activation. The synergistic interplay of both linkages markedly enhanced healing efficiency, mechanical strength, and solvent resistance, achieving rapid reconfigurability and recyclability without any catalyst assistance.

Collectively, these studies underscore that multi-dynamic synergistic mechanisms are emerging as a central molecular design strategy in the development of bio-based epoxy systems. Through

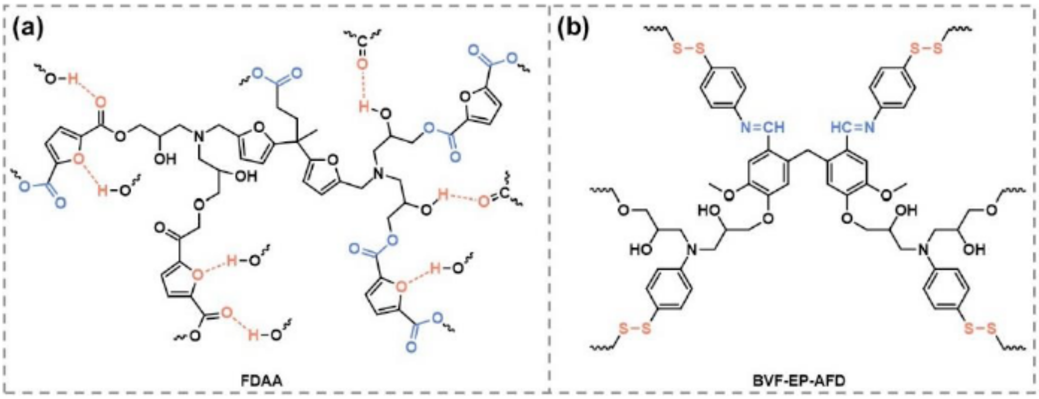

Figure 2: Chemical structure of multi-dynamic covalent bond in epoxy resin network: dynamic ester bonds and hydrogen-bonding interactions (**a**); imine and disulfide linkages (**b**).

coordinated control of crosslink topology and dynamic behavior, next-generation recyclable epoxy thermosets can be engineered to simultaneously achieve structural robustness, multifunctionality, and environmental sustainability.

4. TYPICAL STRUCTURE OF DYNAMIC BONDS

Dynamic bio-based epoxy networks exhibit adaptive behavior governed by various associative exchange mechanisms, where network topology can rearrange without loss of cross-link density. These dynamic covalent mechanisms directly determine thermal stability, mechanical robustness, and reprocessability. The following sections summarize how different exchange chemistries operate within epoxy frameworks and how they translate into macroscopic functionality.

4.1. Transesterification Networks

Among various dynamic covalent chemistries, transesterification (TE) has emerged as the most representative associative mechanism in epoxy vitrimer. In 2011, Leibler and coworkers [35] opened a window to vitrimer chemistry by using zinc acetate-catalyzed TE in epoxy thermosets through the reaction of conventional DGEBA and anhydride curing agents. It enables topological rearrangement through reversible ester–hydroxyl exchange, allowing network reconfiguration without reducing cross-link density. This mechanism is particularly compatible with phenolic architectures, which are the primary structure in natural biogenic materials, as hydroxyl groups readily participate in exchange with ester linkages under mild catalytic conditions (typically Zn^{2+} or DMAP) [36-38]. In addition to enabling topological rearrangement, ester-containing networks also possess inherent hydrolytic and alcoholysis-sensitive motifs. Under acidic or alkaline conditions, the ester linkages can undergo controlled cleavage, providing a potential degradation pathway and facilitating chemical recycling. Early investigations validated its high efficiency in petroleum-based epoxy matrices and subsequently expanded the approach to bio-based systems derived from eugenol, guaiacol, phloroglucinol, and furan derivatives. Through the synergy of aromatic rigidity and aliphatic ester-bridge flexibility, these systems achieved a unified enhancement of thermal stability, processability, and recyclability.

Liu and co-workers [39] designed one of the earliest bio-based epoxy monomer using eugenol via Williamson etherification and *meta*-chloroperoxybenzoic acid (*m*-CPBA) epoxidation, followed by curing with succinic anhydride (SA) in the presence of $Zn(acac)_2$. Figure **3** illustrates the resulting network contained abundant ester and hydroxyl groups capable of Zn^{2+}-catalyzed transesterification at elevated temperatures (>150 °C). The system with an epoxy/anhydride ratio of 1:0.5 exhibited optimal dynamic behavior, enabling shape memory, crack healing, and chemical recyclability. Importantly, the polymer could be decomposed in ethanol and reformed upon heating, realizing mild closed-loop recycling. The design integrates bio-based epoxy chemistry with vitrimer transesterification dynamics, demonstrating the potential of phenolic-derived epoxies for sustainable material cycles.

Xia *et al*. [40] extended the transesterification to a fully bio-based epoxy system derived from furfuryl glycidyl ether and succinic acid. The furan ring provided rigidity and π-interaction, while the ester–hydroxyl exchange enabled rapid transesterification-driven topology rearrangement under Zn^{2+} catalysis. The system achieved self-healing and reprocessing at 180 °C, maintaining high modulus and strength due to balanced ester density and cross-link distribution. The study emphasized molecular architecture tuning—introducing flexible aliphatic linkers alongside rigid furan moieties—to

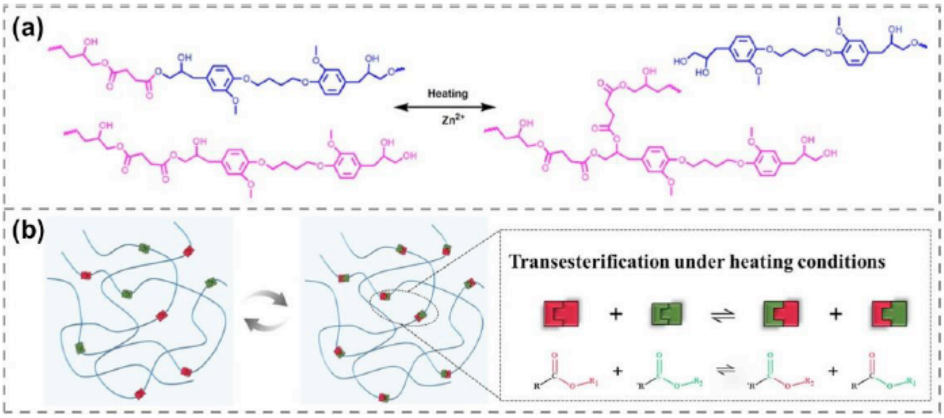

Figure 3: Illustration of heat-induced transesterification reaction in the presence of Zinc catalyst [39] (**a**); schematic of mechanism for transesterification reactions [41] (**b**).

modulate the activation energy and optimize both dynamics and mechanical stability.

4.2. Imine (Schiff-Base) Exchange

Schiff base is a compound generated through the condensation of an aldehyde or ketone with a primary amine, characterized by the general structure –RC=N–. Since the late 2010s, researchers have begun integrating Schiff-base linkages into epoxy and other thermosetting polymer systems to endow them with exchangeable and reconfigurable characteristics [42]. Early investigations revealed that imine linkages generated through aldehyde–amine condensation can serve as dynamic crosslinking centers, imparting reversible topology rearrangement and chemical responsiveness within rigid epoxy networks [43, 44]. Imine bonds are known to reversibly dissociate in aqueous or mildly acidic environments. This reversible hydrolysis enables not only efficient chemical recycling but also contributes to potential degradability under environmental conditions. This conceptual breakthrough laid the foundation for subsequent developments in bio-based epoxy vitrimers, where imine exchange has emerged as a key dynamic mechanism for achieving self-healing and recyclability in sustainable thermoset materials.

Recent advances have targeted bio-based architectures, integrating lignin-derived aldehydes and amino acid–based hardeners to enhance sustainability and performance. Imine bond is active at lower temperatures, and the transamination reaction is catalyst-free, as illustrated in Figure **4**, providing superior reprocessing characteristics. Chen *et al*. [45] designed a series of solvent-free vanillin-derived epoxy vitrimers, synthesized via the polycondensation of aliphatic and aromatic dialdehydes or a trifunctional vanillin derivative with bisphenol F diglycidyl ether (BFDGE) and 4,4′-diaminodiphenyl sulfone (DDS). The resulting network exhibited high rigidity coupled with efficient stress relaxation and chemical degradability, delivering a tensile strength of 109 MPa, modulus of 6256 MPa, and a maximum glass transition temperature of 158 °C. Remarkably, after thermal reprocessing at 200 °C, the material retained over 70% of its original strength. These results confirmed that imine exchange alone can endow aromatic epoxy systems with strong dynamic adaptability and recyclability.

Zhang *et al*. [13] advanced the field by synthesizing fully bio-based Schiff-base curing agents derived from vanillin and amino acids such as lysine and L-DOPA. The cured resins exhibit high T_g (>140 °C), excellent mechanical robustness, and acid-degradable recyclability. The system maintained over 90% of its mechanical integrity even after repeated hot-press reprocessing. Moreover, the networks were completely degradable in 1 M HCl at 50 °C, reflecting a fully closed-loop chemical recyclability pathway without catalyst assistance. Therefore, the unique reversible reactions of Schiff bases—association (imine-amine exchange) and dissociation (imine hydrolysis/recombination)—open up new pathways for researching the degradation of bio-based epoxy resins.

4.3. Disulfide Metathesis

Since the early 2010s, disulfide (–S–S–) and diselenide (–Se–Se–) linkages have been increasingly incorporated into thermosetting polymers as effective

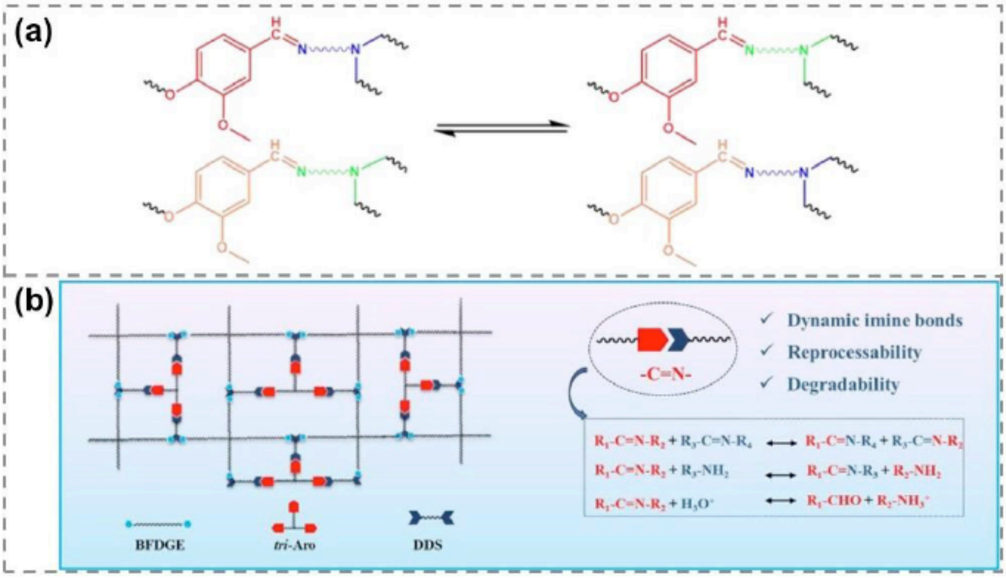

Figure 4: Illustration of imine metathesis reaction (**a**); dynamic crosslink of imine epoxy vitrimers and exchange reactions of imine bonds (**b**) [45].

dynamic covalent motifs for constructing reconfigurable and self-healing networks [46, 47]. These bonds can undergo reversible metathesis reactions upon thermal or photo-stimuli, enabling chain rearrangement and crack repair without disrupting overall network integrity. Early studies demonstrated that introducing aromatic disulfide linkages into polyurethane and epoxy systems allows radical- or redox-driven exchange at room temperature, affording both high toughness and rapid healing [48]. Diselenide analogues, owing to their lower bond energy and faster exchange kinetics, further enabled visible-light-induced repair and low-temperature reprocessability [49]. Nevertheless, disulfide linkages remain more widely adopted, as they provide higher cross-link density and superior thermal–mechanical stability [50].

Following the establishment of disulfide metathesis, recent research has focused on integrating –S–S– linkages into epoxy vitrimers to realize self-healable and reprocessable bio-based thermosets. In these systems, cystamine (CTA) serves as a key bio-based diamine, introducing exchangeable disulfide bonds that undergo reversible cleavage and recombination under mild thermal or photochemical conditions, enabling network rearrangement without catalyst assistance.

Guggari *et al*. established a coherent research route on vanillin-based disulfide epoxy vitrimers, progressively advancing from single disulfide dynamics to dual-bond systems. In their 2023 study [51], a vanillin-derived diglycidyl ether containing imine moieties was cured with CTA, as a possible alternative to the widely used petro-sourced curing agent 4-aminophenyl disulfide (4AFD). Therein, with only 20 mol% of CTA in 4AFD-based formulations, similar relaxation times to cystamine-based analogues can be obtained with a competitive T_g of 80 °C. The resulting vitrimer demonstrated thermally activated reprocessability at 170 °C and self-healing efficiency above 80%, attributed to rapid disulfide metathesis under mild heating. Moreover, the resins can be easily dissolved in the presence of free thiols owing to the thiol−disulfide exchange reactions.

In their 2024 follow-up study [52], a dual-dynamic epoxy was developed, where the vanillin-based epoxy served simultaneously as an imine donor and a disulfide exchange accelerator. This dual mechanism, as shown in Figure **5**(**b**), promoted accelerated stress relaxation, enhanced mechanical recovery, effectively lowering the topology-freezing temperature (T_v) and improving recyclability compared with the single dynamic model. Importantly, this acceleration did not significantly impair the performances of the materials with still competitive T_g values in the range of 120 °C. Together, these two studies illustrate a clear progression from single disulfide-based adaptability to cooperative imine–disulfide synergy, revealing the molecular design strategies necessary for high-performance, reprocessable vanillin-derived epoxy vitrimers.

4.4. Boronic-Ester Exchange

Since the late 1950s, the ability of boric acid to form stable cyclic esters with diols has garnered significant scientific interest [54]. Boronic esters are characterized by their rapid transesterification kinetics and are widely employed to construct dynamic crosslinked networks in bulk materials, solution systems, and aqueous hydrogels. Hydrogels incorporating boric acid

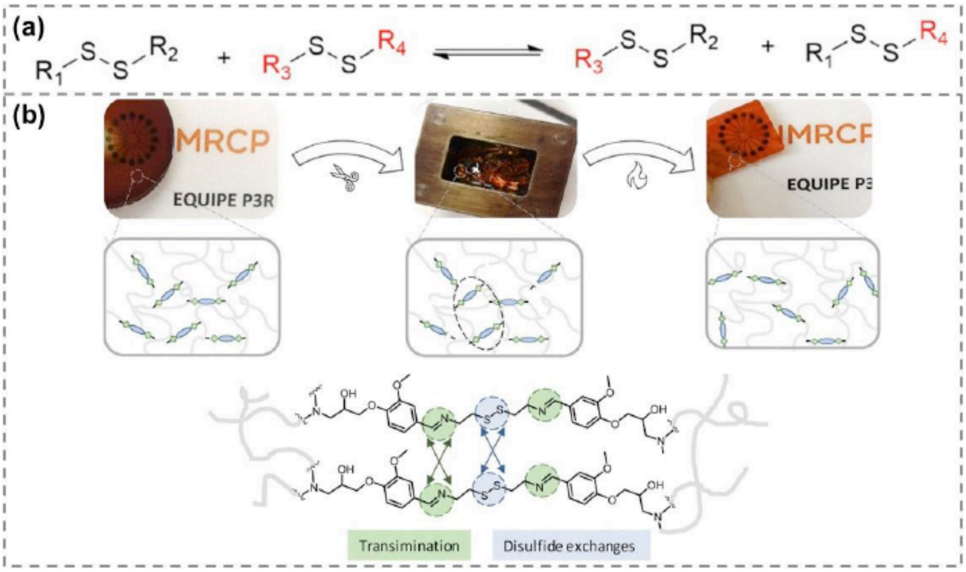

Figure 5: Illustration of exchange reactions of disulfide bonds [53] (**a**); example of reprocessing experiment via compression moulding in a hot press at 170 °C [52] (**b**).

chemistry have been extensively explored in biomedical applications owing to their self-healing and stimuli-responsive properties [55].

Boronic esters undergo reversible exchange between boronic acids and diols through both associative transesterification and dissociative hydrolysis–reformation pathways. As shown by Cash *et al*. [56], the dominant route depends strongly on humidity and temperature: dry environments favor direct diol–ester exchange (Figure **6**(**a**)), while humid conditions promote hydrolysis followed by re-esterification (Figure **6**(**b**)). This dual pathway enables a tunable balance between network stability and adaptability, allowing efficient self-healing and reprocessing, as shown in Figure **6**(**c**). Kang and Kalow [57] further revealed that internal and external catalysis significantly accelerate bond exchange. Amide-adjacent diols act as internal catalysts by facilitating proton transfer, while external species (*e.g*., phosphate or acetate ions) enhance reactivity via anion-assisted catalysis. Such catalytic control enables precise modulation of stress relaxation and viscoelastic behavior, making boronic ester chemistry a versatile platform for designing stimuli-responsive and reprocessable polymer networks.

Ke *et al*. [58] synthesized a bio-based epoxy monomer (EHCPP) with a rigid core and multi-arm architecture by reacting cardanol with hexachlorocyclotriphosphazene followed by *m*-CPBA epoxidation. The resulting EHCPP monomer was subsequently cured with 2,2′-(1,4-phenylene)-bis[4-mercapto-1,3,2-dioxaborolane] (BDB), thereby incorporating dynamic boronic ester linkages to construct the EHCPP–BDB polymer network. The EHCPP–BDB polymer displayed remarkable mechanical performance (tensile strength of 34.9 MPa), exceptional thermal stability (up to 308 °C with a char yield of 19.43%), and strong solvent resistance. Due to the presence of dynamic reversible boronic ester bonds illustrated, the EHCPP-BDB polymer network exhibited good recyclability.

In summary, boronic ester chemistry offers a multifunctional dynamic platform for constructing recyclable, self-healing, and thermo-responsive bio-based thermosetting materials. However, the direct integration of boronic ester linkages into bio-based

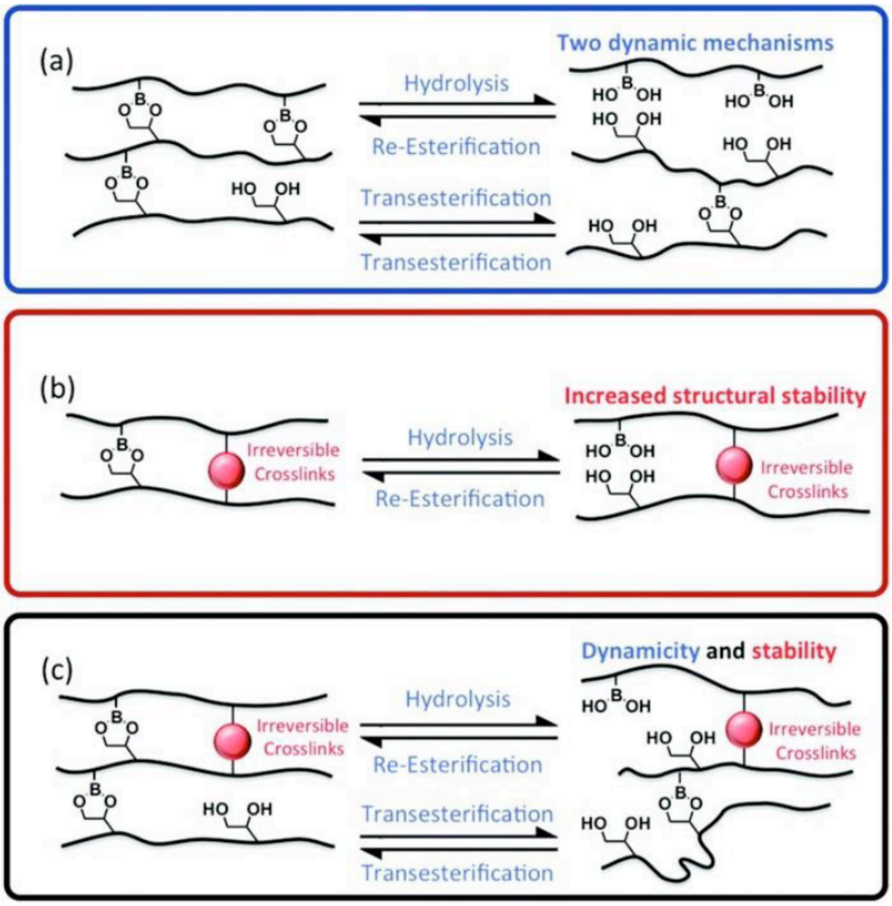

Figure 6: Schematic representation of boronic ester networks with excess diol (**a**); boronic ester networks that incorporate irreversible crosslinks (**b**); boronic ester networks that include excess diol and irreversible crosslinks (**c**), illustrating reversible exchange between boronic acids and diols through both associative transesterification and dissociative hydrolysis–reformation pathways [56].

epoxy networks remains scarcely explored, presenting a highly promising but still underdeveloped research avenue.

4.5. Acetal Structure

Aldehydes or ketones react with polyols in acidic environments to yield acetal structures. Thermosetting materials containing acetal linkages can undergo reversible cleavage and recombination under acid catalysis or heating, exhibiting reconfigurability, self-healing ability, and chemical recyclability [59–61]. The dynamic behavior of acetal bonds proceeds through two main routes. (1) In the proton-catalyzed nucleophilic addition–substitution cycle, protonation of the ether oxygen renders it vulnerable to nucleophilic attack by water or hydroxyl groups, forming hemiacetal or aldehyde intermediates that subsequently condense with neighboring hydroxyls to regenerate new acetal linkages (Figure **7**(**a**)). (2) A non-catalytic, thermally driven pathway also exists (Figure **7**(**b**)). At elevated temperatures (150–200 °C), the acetal oxygen bridge can self-catalytically exchange with nearby hydroxyl or ether oxygens, displaying reconfigurable dynamics akin to transesterification or imine exchange.

As an example, Ma *et al*. [62] developed a vanillin-derived spiro-diacetal compound, which was converted to an epoxy monomer via reaction with epichlorohydrin and then cured with isophorone diamine to form a thermoset. The labile vanillin-based spiro-diacetal structure endows the thermoset with excellent degradability under mildly acidic conditions, while maintaining stability in neutral or basic environments, along with a high T_g and modulus.

Wang *et al*. [22] designed a bio-derived structural unit combining rigidity, flexibility, and chemical degradability through the reaction of vanillin with epichlorohydrin. The epoxy resin cured with DDM exhibited mechanical properties (higher strength and toughness) far superior to those of commercial high-performance BPA-based plastics, while maintaining comparable thermal stability. Moreover, the acetal linkage introduced dynamic depolymerization capability, enabling acid-triggered breakdown and recovery of the original monomers and polyols.

5. ADVANCES IN BIO-BASED EPOXY COMPOSITES FEATURING DYNAMIC BONDS

Epoxy resin–based composites have long dominated high-performance structural materials, prized for their exceptional strength, thermal stability, and interfacial adhesion in fields such as aerospace, automotive engineering, and wind energy. With the advancement of sustainable material concepts, researchers have gradually incorporated bio-based epoxy systems into fiber-reinforced and filler-reinforced composite structures [64, 65]. These systems maintain the high strength and stiffness of conventional composites while embracing the circular-design philosophy through the use of renewable matrices. Incorporating dynamic, exchangeable bond networks into epoxy matrices transforms conventional composites into multifunctional systems—capable of self-healing, reprocessing, fiber recovery, and structural remanufacturing—propelling bio-based epoxy composites toward a new generation of high-performance, sustainable, and adaptive materials [66, 67].

5.1. BIO-Based Epoxy Blends with Dynamic Bonds

According to IUPAC, polymer blending is a macroscopic mixture of two or more distinct polymers. Combining multiple polymers allows tailoring the final product's properties and is typically more economical than creating entirely new polymers. In bio-based epoxy systems, blending has emerged as a promising strategy to simultaneously enhance performance and sustainability. Researchers have achieved significant advances in processing, toughness, viscosity

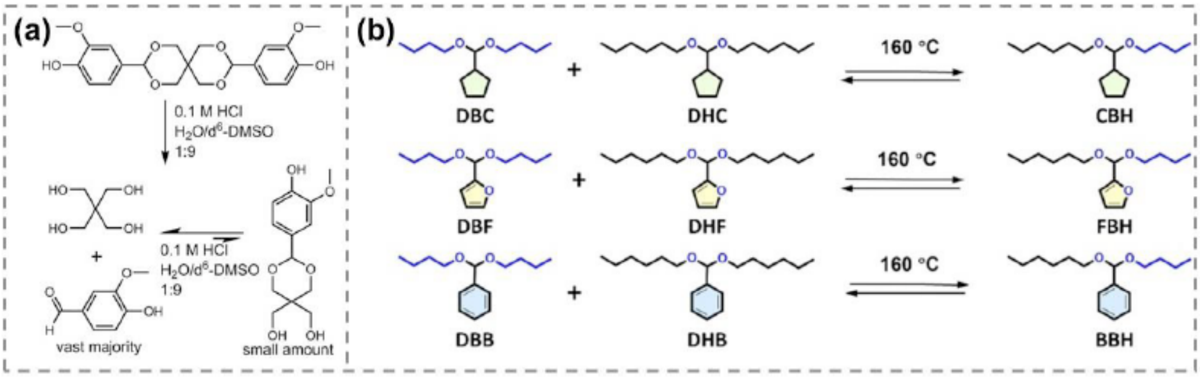

Figure 7: Supposed degradation mechanism of the acetal structure in the proton-catalyzed nucleophilic addition–substitution cycle [62] (**a**); acetal metathesis model reactions [63] (**b**).

regulation, and fracture resistance by physically mixing bio-based epoxy prepolymers or matrices with thermoplastics, elastomers, renewable oils, or rubber—avoiding covalent copolymerization—and thus addressing the traditional brittleness of bio-based epoxies [68-70].

Zamani *et al*. [71] developed a fully aromatic imine/Schiff-base (VAP) dynamic bisphenol curing agent using vanillin and 4-aminophenol. DGEBA was subsequently cured with VAP, with or without poly(ethylene glycol) (PEG), to produce a glassy network. In the DGEBA-VAP system, imine linkages incorporated within the vanillin-based hardener endow the epoxy network with reprocessability, self-healing, and recyclability. When PEG is added, the DGEBA-VAP-PEG system evolves from a purely reversible imine network into one featuring both reversible imine bonds and supramolecular forces. Studies show that the presence of PEG, induced by supramolecular interactions—especially hydrogen bonding—enhances mechanical strength (fracture toughness and fracture energy increased by 21% and 48%, respectively), thermal stability, self-healing ability (Figure **8**(**a**)), storage modulus, and stress relaxation rate.

Polycaprolactone (PCL) possesses remarkable shape-memory behavior, returning to its original shape after temporary deformation once the temperature exceeds its T_g. Raman *et al*. [72] investigated how PCL modulates bio-epoxy properties by creating ternary composites with varying PCL ratios incorporated into tannic-acid-cured bio-based epoxy systems. The resulting composites, with strong crosslinking and dynamic networks, display outstanding shape-memory performance under thermal and chemical triggers. Additionally, the PCL effect combined with dynamic bond exchange mechanisms enables these composites to achieve high self-healing efficiency and recover from physical damage. It was found that incorporating 5 wt.% PCL enhances the composite's flexural strength by over fivefold (520.47%). This result demonstrates the excellent synergy between PCL and the epoxy matrix. Composites with 5 wt.% PCL achieve the best balance of mechanical and self-healing performance. whereas 20 wt.% PCL demonstrates enhanced chemical responsiveness. As shown in Figure **8**(**b**), the observed chemical-responsive shape-memory of PCL-incorporated bio-epoxy composites in methanol and acetone arises primarily from solvent–network interactions, especially the dynamic ester bonds generated during curing.

Shadmand *et al*. [73] employed chitosan as a green reinforcing agent in the glassy polymerization of thermosetting epoxy resins. Combining chitosan addition with mechanochemical ball milling yielded epoxy–chitosan composites with superior performance. The resulting glassy composites display enhanced mechanical performance, featuring increased modulus and tensile strength, excellent solvent resistance, and

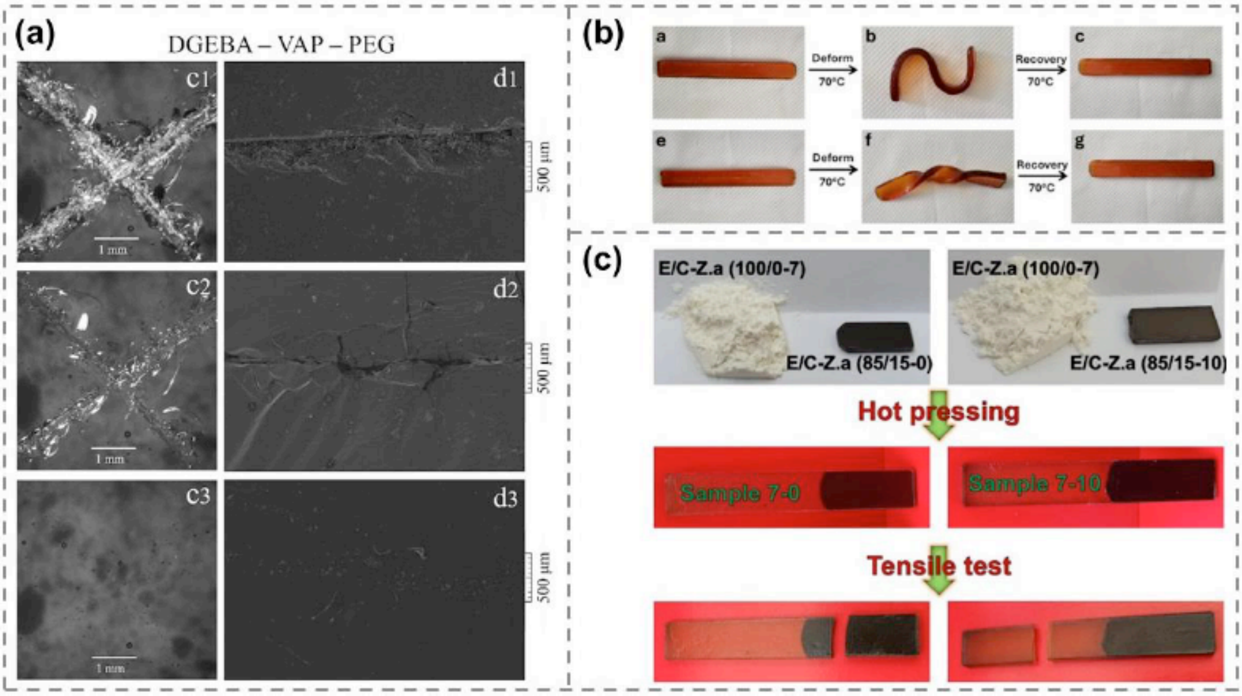

Figure 8: Multifunction of composites containing dynamic bonds through blending: self-healing ability of the DGEBA-VAP-PEG system containing reversible imine bonds and supramolecular forces [71] (**a**); shape-memory of PCL-incorporated bio-epoxy composites containing dynamic ester bonds [72] (**b**); reprocessing ability of epoxy–chitosan composites through ester–amide exchange reactions [73] (**c**).

facile stress relaxation. The free hydroxyl groups in chitosan promote ester exchange reactions, which are key to network reconfiguration. Additionally, primary amine groups on the chitosan backbone may act as internal catalysts, further accelerating exchange reactions. These amines can also engage in ester–amide exchanges, enabling the formation of multiple dynamic bonds within the network. Furthermore, the introduction of zinc acetate as an external catalyst further modifies the characteristics of the glassy composites. It facilitates metal–ligand complexation and provides additional crosslinking sites, resulting in a stiffer and more robust material. The presence of zinc acetate also promotes ester–amide exchange reactions, forming a network with multiple dynamic exchange mechanisms and imparting reprocessability (Figure **8**(**c**)) and recyclability.

The improved performance of these bio-based epoxy blends can be interpreted through their structure–property relationships governed by dynamic bond chemistry. In blended systems, reversible linkages such as transesterification, imine exchange, and disulfide metathesis play a critical role in regulating phase compatibility and chain mobility at the molecular level. These exchangeable bonds enable topological rearrangement under external stimuli, allowing stress relaxation and adaptive interfacial interactions between different resin components. As a result, the blended networks exhibit enhanced toughness, accelerated healing kinetics, and improved recyclability compared with non-dynamic counterparts. Furthermore, the cooperative interactions between rigid bio-based aromatic units and flexible dynamic moieties create a hierarchical balance between stiffness and reconfiguration ability. This mechanistic understanding aligns with trends reported for other dynamic epoxy systems and highlights the effectiveness of dynamic covalent design in optimizing blend performance for sustainable epoxy materials.

5.2. Applications of Multifunctional Fillers in Bio-Based Epoxy Vitrimers

Recent research has shown that adding fillers—ranging from micro-scale particles to nanomaterials—into dynamic-bond epoxy systems is a crucial strategy for improving mechanical properties, functional responsiveness (such as thermal, electrical, and photothermal performance), and recyclability. However, filler aggregation, poor interfacial compatibility, hindrance to network exchange kinetics, and filler-induced limitations on crosslink density or chain mobility may weaken the self-healing or reprocessing ability of dynamic-bond systems [74, 75]. Accordingly, contemporary research has emphasized two strategies: low filler content with high dispersion and functionalized interfaces, and fillers that simultaneously enhance mechanical strength and dynamic response [76].

Sun *et al*. [77] created a recyclable, processable epoxy composite with high thermal conductivity using bio-based epoxy soybean oil (ESO). The epoxy achieves recyclability and processability through disulfide bond exchange, while the addition of boron nitride (BN) fillers enhances the thermal conductivity of the matrix. At 35% filler loading, the EPV/BN35 composite achieves a thermal conductivity of 2.27 $W\cdot m^{-1}\cdot K^{-1}$. Results show that EPV/BN35 maintains excellent self-healing and reprocessing capabilities. BN fillers were oriented in films through calendaring, and bulk composites were produced by hot-pressing these stacked films. The stacked EPV/BN35-F sample exhibits a thermal conductivity of 3.01 $W\cdot m^{-1}\cdot K^{-1}$ along the stacking direction, 32.6% higher than EPV/BN35. Moreover, due to disulfide bond exchange, the matrix can decompose under chemical conditions, allowing the fillers to be fully separated via physical filtration and reused, achieving closed-loop filler recycling, as illustrated in Figure **9**(**a**).

Luo *et al*. [78] created a bio-based zinc-chelated casein phosphopeptide (CPP-Zn) specifically for use as a flame retardant and smoke suppressant in epoxy glass networks. Simultaneously, a reactive DOPO derivative (DDP) was synthesized via grafting with bio-based itaconic acid ester. It was subsequently employed together with CPP-Zn as a synergistic flame retardant, with varying ratios of CPP-Zn and DDP used to fabricate epoxy vitrimer composites. Research shows that CPP-Zn and DDP together provide epoxy resins with superior flame retardancy through synergistic gas-phase and condensed-phase mechanisms, as shown in Figure **9**(**b**). In the flame-retardant epoxy composite 50D/EST-25Cz, compared to EST without fillers, the peak heat release rate (pHRR) decreased by 73.7%, total heat release (THR) by 41.4%, UL-94 achieved V-0 rating, and LOI reached 32.3%. Ester exchange in these flame-retardant composites effectively maintained the self-healing and recyclability of the glassy monomer. The remolded 50D/EST-25Cz exhibits a tensile strength of 11.4 MPa, retaining 72.2% of its original mechanical strength.

Song *et al*. [79] prepared a multifunctional polymer (TAESO) from epoxy soybean oil and vanillin, creating a dual-dynamic 3D network with both hydrogen bonds and dynamic imine bonds. This polymer exhibits high flexural strength (25.51 ± 0.27 MPa) and rigidity (fracture strain ~2%) at room temperature. The dual-dynamic cross-linking network provides the polymer with high self-healing efficiency, recyclability,

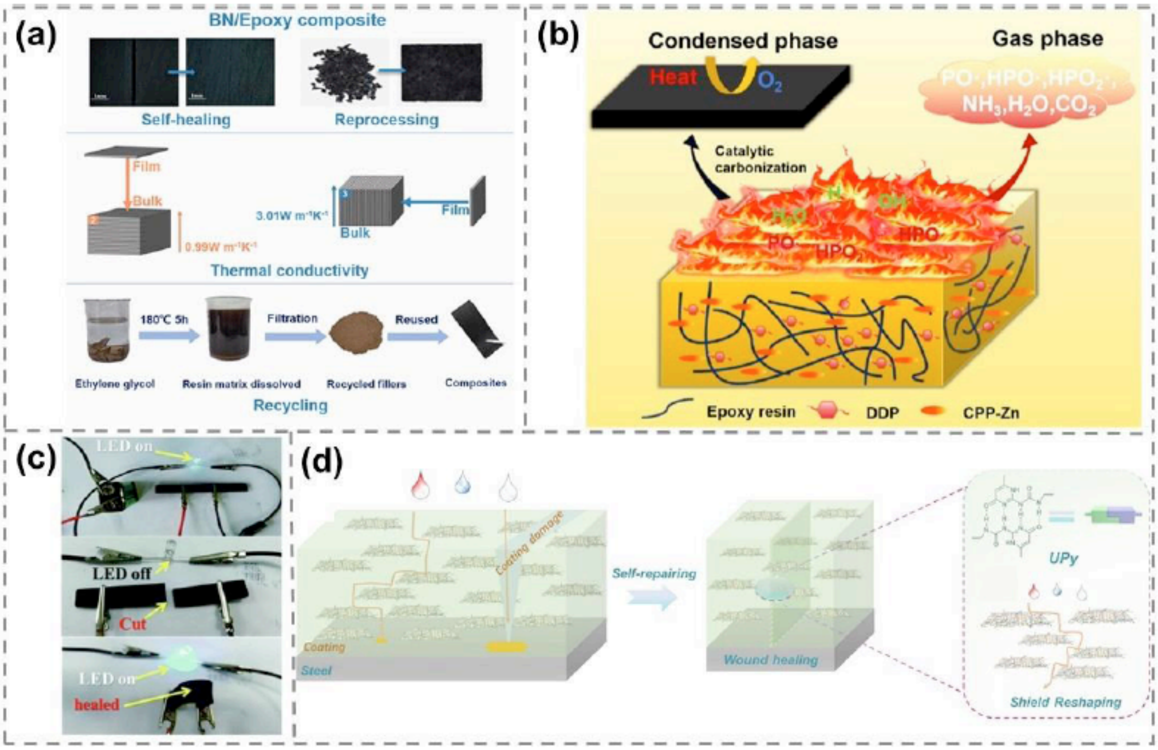

Figure 9: Multifunction of composites filled with BN [77] (**a**); flame-retardant epoxy composite filled with CPP-Zn and DDP [78] (**b**); the conductive composite by adding MWCNTs with self-healing ability [79] (**c**); the corrosion protection principle of composites with UPy-modified GO [81] (**d**).

reversible and permanent shape adaptability, and reusable adhesive properties. By adding multi-walled carbon nanotubes (MWCNTs), a conductive composite was produced that preserved the polymer's inherent properties, improved adhesion, and exhibited both plasticity and self-healing, forming an adhesive conductive material, as shown in Figure **9**(**c**).

Hu *et al*. [80] designed a doubly reversible cross-linked epoxy vitrimer EDCS/DPTA using cashew phenol-based epoxy monomer and 3,3'-dithiodipropionic acid as curing agent. Catalyzed by 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD), the combined effect of disulfide and dynamic ester bonds allows the EDCS/DPTA network to rearrange readily, exhibiting superior recyclability, multi-stimuli responsiveness, degradability, and reprocessing capabilities. Graphene was then incorporated to obtain the EDCS/DPTA/G conductive composite, enhancing both thermal stability and electrical properties relative to the original resin. Relative to EDCS/DPTA, the EDCS/DPTA/G composite doubled its mechanical strength and Young's modulus, with toughness markedly enhanced to 81.1 $J{\cdot}m^{-3}$. Strong π-π interactions exist between the phenyl rings of the cashew phenol epoxy monomer and graphene, resulting in strong interfacial interactions between EDCS/DPTA and graphene. Therefore, during stretching, graphene bridges across fracture surfaces, similar to the toughening mechanism of carbon nanotube-reinforced polymers. This interfacial bridging contributes to more efficient stress transfer and crack deflection, fundamentally explaining the observed enhancement in modulus and toughness. In addition to *π–π* interactions, the presence of dynamic covalent bonds within the vitrimer matrix allows reversible bond exchange near the graphene–resin interface. This dynamic adaptability reduces interfacial residual stresses and enables local topological rearrangement under load, thereby facilitating more effective load transfer and mitigating interfacial debonding. Comparable interfacial reinforcement effects have also been reported in other graphene-modified vitrimer systems, indicating that the combination of *π–π* interactions and bond-exchange-driven interfacial reconfiguration serves as a universal mechanism for strengthening bio-based epoxy composites.

Wu *et al*. [81] reported an innovative strategy to construct self-healing, high-performance, nacre-mimetic bio-based nanocomposites. Flexible side chains with quadruple hydrogen bonds were incorporated into the cashew phenol epoxy network to form a bio-based self-healing elastomer. Subsequently, UPy-modified graphene oxide (GO) with isocyanate groups was integrated into the bio-based prepolymer, producing a nacre-mimetic nanocomposite exhibiting outstanding strain tolerance (617%) and ultimate tensile strength of 5.1 MPa. Benefiting from the densely packed quadruple dynamic reversible supramolecular hydrogen bonds, the coating exhibits excellent self-healing efficiency at room temperature within 10 min. Moreover, the coating retains effective self-healing performance even in artificial seawater. The high-barrier GO layers also provide the nacre-inspired coating with exceptional long-term resistance to corrosive media, as illustrated in Figure **9**(**d**). In addition to the brick-and-mortar architecture of GO platelets, the dynamic nature of the quadruple hydrogen-bond arrays enables reversible dissociation and reformation under mechanical loading. This reversible bond-exchange process promotes interfacial stress relaxation, redirects crack propagation pathways, and dissipates deformation energy through multiscale dynamic rearrangement. Such dynamic-bond-assisted toughening aligns with other nacre-inspired vitrimer and supramolecular epoxy systems, where reversible interactions synergize with hierarchical structures to enhance mechanical robustness and environmental durability. In conclusion, the nacre-mimetic coating demonstrates outstanding sustainability, reliability in use, and durable protective capability.

The diversity of fillers provides a versatile platform for the multifunctional modification of bio-based epoxy resins, enabling the integration of properties such as enhanced strength, flame retardancy, and self-healing. Furthermore, the strong interfacial interactions, often achieved through surface engineering, facilitate efficient stress transfer, leading to a simultaneous improvement in the toughness, strength, and modulus of the composite materials. This "structure-function" integrated design philosophy is pivotal for guiding the development of next-generation high-performance bio-based epoxy resins.

5.3. Carbon/Glass Fiber Reinforced Epoxy Composites

Carbon fiber reinforced composites (CFRCs) are advanced polymer-carbon fiber materials that combine high strength, lightweight properties, and fatigue resistance, making them widely used in aerospace and various high-performance applications [65,82]. However, a major limitation of CFRCs is their lack of recyclability, attributed to the permanent cross-linked networks of thermosetting resins. Incorporating degradable dynamic bonds into polymer matrices provides a new route to address the environmental and economic issues of traditional recycling, allowing carbon fibers to be recovered intact while preserving composite performance.

Dong *et al*. [83] synthesized a bio-based reinforcing agent (BVT) through condensation of vanillin and tyramine, integrating it as hydrogen-bonded physical entanglement sites within an epoxy vitrimer. The incorporation of BVT significantly enhanced the mechanical properties of the epoxy vitrimer, increasing tensile strength, flexural strength, and impact strength by 44%, 26%, and 225%, respectively. The synergistic action of dual dynamic covalent bonds (Schiff base and siloxane bonds) enhanced dynamic behavior, endowing the material with self-healing, efficient reprocessing, and closed-loop recyclability. Furthermore, CFRC laminates prepared by vacuum infusion molding showed excellent reprocessability during hot pressing, with both original and reprocessed composites maintaining high mechanical performance. In THF solvent, the composite network can dissolve, enabling separation of carbon fibers from the epoxy resin interface. Recovered carbon fibers (CFs) retained their original morphology, chemical structure, and mechanical properties, while the reclaimed resin preserved stable structure and thermal characteristics, as illustrated in Figure **10**(**a**). These improvements can be attributed to the ability of dynamic linkages to reorganize at the fiber–matrix interface, reducing residual stresses and improving stress transfer efficiency. Similar interfacial self-adjustment has been documented in other imine-based CFRPs, confirming that dynamic exchange at the interface is a key contributor to mechanical retention after recycling.

Xu *et al*. [84] produced a self-healing, chemically degradable vitrimer matrix without catalysts by curing tung oil-derived triglycidyl ester (TOTGE) with methylenediamine (MDA). Due to the introduction of terminal epoxy groups and rigid MDA, both TOTGE-MDA vitrimer matrix and CFRC exhibited excellent mechanical properties. Tertiary amines in the vitrimer network catalyze dynamic transesterification reactions (DTER), enabling the resin to exhibit excellent self-healing, physical recyclability, and shape memory without additional catalysts. Simultaneously, TOTGE-MDA-CF composites also showed excellent reprocessability, self-adhesion (Figure **10**(**b**)), and shape memory properties. The self-adhesive CFRC exhibited a shear strength of 4.1 ± 0.2 MPa. In addition, TOTGE-MDA-CF composites could be completely chemically degraded after 60 minutes of ethanolamine (EA) treatment at 90 °C. Importantly, the recycled carbon fibers retained their chemical structure and tensile performance after EA-induced degradation.

Yang and colleagues [85] developed two intrinsic flame-retardant epoxy monomers (VAD-EP and VDP-EP) from vanillin, DOPO, and DDM. By adjusting the ratio of the two epoxy vitrimers (A_xP_y–D230), the resin achieved a UL-94 V0 flame-retardant rating even at a low phosphorus content of 0.66%. In addition, the imine bonds served as dynamic covalent sites with remarkable reversibility, imparting outstanding

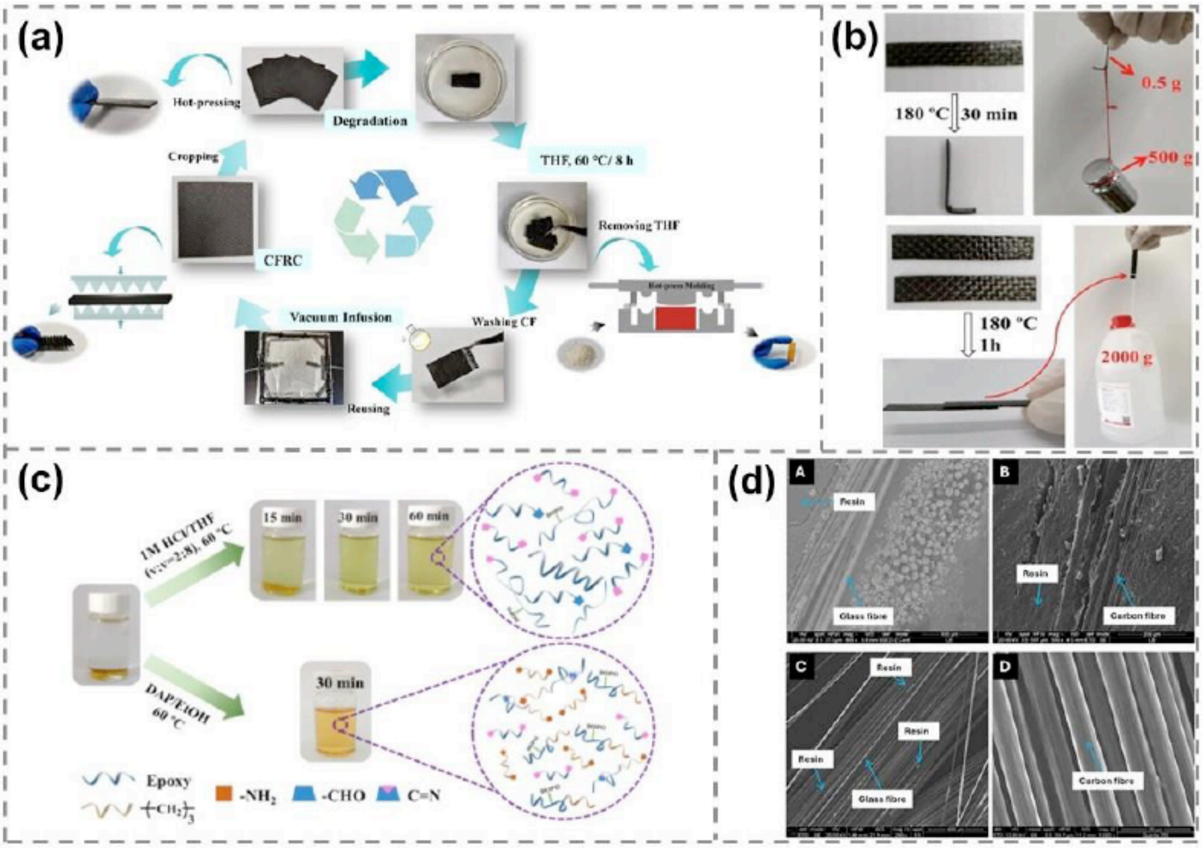

Figure 10: Schematic illustration of preparation and recycling of CFRC [83] (**a**); reprocessing and self-adhering ability of the TOTGE–MDA–CF composite [84] (**b**); degradation behavior of epoxy vitrimer [85] (**c**); SEM images of recycled fibers [87] (**d**).

recyclability and self-healing performance. The carbon fiber–reinforced composite (A_7P_3–D230/CF), fabricated via manual brush impregnation, demonstrated strong interfacial adhesion, showing a flexural strength of 187 MPa and a modulus of 15.2 GPa. Under mild acidic conditions, the vitrimeric epoxy network underwent full degradation, as illustrated in Figure **10**(**c**). As a result, carbon fibers were fully recovered from vitrimer composites while maintaining their original chemical composition, mechanical integrity, and morphology.

Comparable to CFRPs, glass fiber reinforced composites (GFRCs) offer a desirable balance of strength, rigidity, and affordability, making them a preferred option. Mora *et al*. [86] reported a novel sustainable self-healing polymer composites developed from glass fabric reinforced copolymers of eugenol/furfurylamine-derived benzoxazine resin (E-fa) and epoxidized castor oil (ECO) actuated by near infrared (NIR) light. Results revealed that glass fiber reinforcement markedly enhanced the composite's thermal performance, stability, and flexural mechanical strength. The authors suggested that the self-healing mechanism of the E-fa/ECO copolymer involved reversible ring-addition reactions at 90 °C, the temperature associated with dimensional change. The GF-reinforced E-fa/ECO composites showed excellent macroscopic thermal healing performance at fiber contents ranging from 64% to 86%. Additionally, the composite demonstrated a NIR–induced microscopic repair effect, achieving healing within approximately 152 s.

Billington and Shaver [87] prepared degradable epoxy resins from traditional epoxy monomers and a new bio-derived curing agent, bis(1,3-dioxolan-4-one) (bisDOX). The resulting resins served as matrices for fiber-reinforced polymer composites (FRPs) and displayed outstanding thermo-mechanical characteristics. The GFRP exhibited a tensile strength of 289 MPa and modulus of 18 MPa, whereas the CFRP reached 119 MPa and 28 GPa. These values render the composites comparable in performance to traditional FRP materials. Owing to the presence of ester bonds, bisDOX-based epoxy composites can be recycled under mild conditions using organic catalysts to break crosslinks and dissolve oligomers, enabling fiber recovery and reuse, as shown in Figure **10**(**d**). The recycled composites exhibited negligible loss of mechanical strength, establishing a circular-use system that offers a sustainable substitute for conventional FRPs.

Overall, the improved performance of carbon- and glass-fiber-reinforced bio-based epoxy composites arises from the synergistic effect of dynamic covalent bonds and fiber reinforcement. Reversible linkages such as imine, siloxane, and transesterification bonds enable interphase stress relaxation and adaptive topology rearrangement, leading to more efficient load transfer and preventing irreversible debonding. Dynamic covalent bonds allow the epoxy matrix to be selectively depolymerized during acid- or alcohol-assisted chemical recycling, enabling efficient recovery of carbon or glass fibers. This dynamic interfacial behavior, combined with the stiffness of bio-based aromatic networks and the hierarchical reinforcement of continuous fibers, results in enhanced mechanical strength, reprocessability, and recyclability. Thus, the structure–property relationship of these composites is governed by the coordinated interaction of dynamic bond chemistry and fiber–matrix interfacial architecture.

5.4. Other Routes for Reinforcement

Natural fibers, recognized as sustainable reinforcement materials, have garnered increasing attention for their potential in developing eco-friendly bio-based composites. Fei and colleagues [88] designed a high bio-content vitrimer matrix through a facile synthesis route employing hemp seed oil–derived epoxy resin, limonene diamine, and commercially available isophthalaldehyde. This study established a dual-dynamic crosslinked network composed of hydroxyl ester and imine linkages, and reinforced the resulting vitrimer with hemp fibers to obtain a natural fiber composite. The introduction of amino silane increased the crosslinking density and enhanced interfacial adhesion between the hemp fibers and the matrix. The results showed that both the vitrimer and composites exhibited higher glass transition temperatures (79.9–90.8 °C), comparable elastic modulus (>3400 MPa), and lower water absorption (49%–38%). Furthermore, because both hydroxyl ester and imine linkages are degradable in amine solutions, the resulting composites can be efficiently recycled through aminolysis under mild conditions (100 °C for 3 h).

Xu and co-workers [89] introduced a polarity-difference–induced in situ microphase separation strategy to enhance the toughness of vanillin-derived epoxy resins. By grafting alkyl glycidyl ether onto rigid amine curing agents, , chemically integrated micro/nanoscale domains were generated during the curing process. The modified vanillin-based epoxy resin exhibited a 6.1-fold increase in tensile toughness compared with the pristine resin, attributed to a toughening mechanism analogous to block copolymer systems and microphase-separation-induced reinforcement. Benefiting from the microphase-separation toughening and flexible chain structure, the glass fiber composites showed high toughness and excellent flexural performance. Meanwhile, the reversible Schiff base bonds endowed the material with degradability and processability, enabling resin and fiber separation and recycling, thereby reducing the environmental impact of glass fiber composites.

Overall, current research on bio-based epoxy resin composites containing dynamic bonds remains primarily focused on carbon fiber–reinforced systems, owing to the exceptional specific strength, specific modulus, and thermal/electrical conductivity of carbon fibers, which make them ideal model systems for verifying the reversibility and recyclability of dynamic network structures. Incorporating exchangeable dynamic covalent bonds—such as ester, imine, and disulfide linkages—into epoxy matrices enables self-healing, reprocessing, and closed-loop recycling in carbon fiber composites, without compromising, and often improving, mechanical and thermal performance. These efforts are advancing from conceptual models toward multifunctional materials—high-end structural composites combining flame retardancy, conductivity, self-healing, and shape-memory functions.

However, the application of other types of reinforcing materials in dynamic bond–based bio-epoxy systems remain relatively limited. For instance, basalt fibers—naturally inorganic and thermally stable—are used extensively in wind turbines, vehicles, and fire-resistant composites [90]. Aramid fibers such as Kevlar® combine light weight with outstanding impact resistance, ideal for tough and protective structures [91]. Natural reinforcements like bamboo, flax, and straw fibers excel in biodegradable composites [92]. Coupling them with dynamic epoxy systems could unlock new opportunities for lightweight, recyclable, and sustainable structures. Moreover, functional fillers—nanocellulose, MXenes, carbon quantum dots, and layered clays like montmorillonite—offer strong thermal, shielding, and flame-retardant performance [93], yet their integration with bio-based dynamic epoxies remains scarcely explored. Looking ahead, merging these reinforcements with reconfigurable dynamic networks could harmonize performance and sustainability, paving the way for dynamic epoxy composites in flexible electronics, healable coatings, and smart structural monitoring systems.

CONCLUSIONS AND OUTLOOKS

This review highlights the great potential of dynamic bond–containing epoxy resin composites to achieve a balance among high performance, recyclability, and sustainability. Biomass-derived building blocks such as lignin and cellulose provide renewable alternatives to

BPA-based epoxy systems, while dynamic covalent chemistry enables reversible bond exchange that improves mechanical integrity, environmental stability, and closed-loop recyclability. The chemistry and exchange mechanism of dynamic bonds dictate self-healing kinetics, topological rearrangement temperature, and mechanical behavior. Overall, dynamic covalent strategies fundamentally reshape the structure–property relationships of bio-based epoxy systems, allowing molecular-level dynamics to translate into macroscopic improvements across diverse composite designs.

Despite significant advances in molecular design and performance optimization, bio-based dynamic epoxy networks still face several challenges. Epoxy resins containing reversible covalent linkages exhibit certain drawbacks; for instance, imine-based systems are moisture-sensitive with poor hydrothermal stability, and disulfide-based networks often display inferior thermal resistance, all of which constrain their broader application potential. In addition, the recycling and reuse of bio-based epoxy resins remain in their infancy, with numerous technical and practical issues still to be addressed. Variations in reactivity and purity of bio-based raw materials cause poor batch reproducibility, limiting industrial scalability and standardization. Moreover, the cooperative and competitive interactions among multiple dynamic exchange reactions remain poorly understood, as systematic theoretical frameworks are still lacking, limiting precise prediction of the relationship between network architecture and macroscopic performance. Although some systems have achieved closed-loop recycling, optimization is still needed regarding recycling cycles, energy consumption, and byproduct control to meet genuine sustainability goals.

Looking forward, the evolution of this field will likely emphasize precision molecular design and the coordinated optimization of structure–function relationships. On one hand, advances in computational chemistry and machine learning will allow predictive modeling of dynamic reaction barriers and topological rearrangement behaviors. On the other hand, integrating renewable monomers (*e.g.*, magnolol, vanillin, tannins) with multi-responsive dynamic structures (*e.g.*, acetal–imine or ester–boronated hybrid bonds) will broaden functional dimensions and enable precise control over photo-, thermal-, and pH-responsive behaviors. Applying these dynamic bio-based epoxy systems to recyclable carbon fiber composites, degradable coatings, flexible electronic encapsulations, and high-temperature structural components is expected to catalyze the paradigm shift of thermosetting materials from “single-use” to “multi-cycle” sustainability. Practically, dynamic bio-based epoxy systems show strong potential in next-generation recyclable composites, environmentally friendly coatings, and smart structural applications. Future work should also focus on scalable synthesis, dynamic interface engineering, and standardized recycling protocols to facilitate industrial translation.

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CONFLICTS OF INTEREST

The author declared no conflicts of interest.

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