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Review Article

Nanocomposites based on epoxy vitrimer

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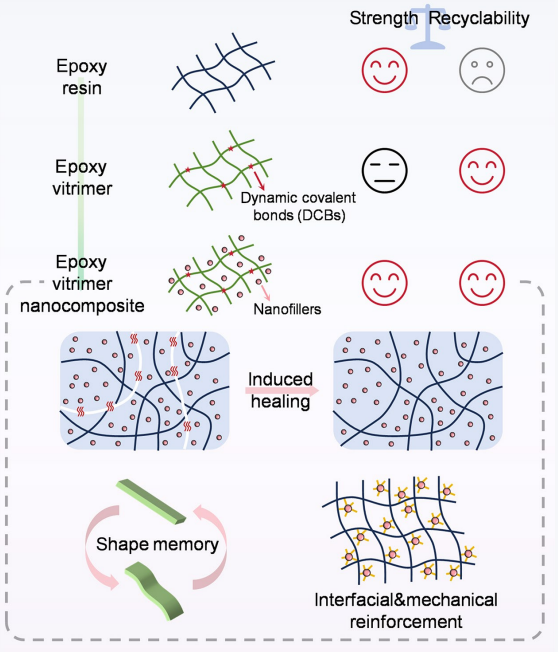
ABSTRACT: Epoxy vitrimer nanocomposites represent a class of advanced materials merging high performance with sustainability. The incorporation of nanofillers into a polymer matrix to enhance material properties is a well-established strategy. However, the permanently cross-linked networks of conventional epoxy thermosets render the resulting composites non-recyclable, contributing to significant economic and environmental concerns. In contrast, epoxy vitrimers incorporate dynamic covalent bonds, which enable reprocessability and self-healing while retaining the mechanical robustness of thermosets, thereby offering promising routes toward recyclable nanocomposites. This review outlines recent progress in epoxy vitrimer nanocomposites, highlighting the pivotal role of various nanofillers—such as carbon nanotubes, graphene, silica, and magnetic Fe_3O_4 nanoparticles—in not only reinforcing mechanical properties but also introducing functionalities including electrical and thermal conductivity, self-healing, and stimulus responsiveness. These materials have demonstrated considerable potential for applications in electromagnetic interference shielding, smart coatings, and flexible sensors. Through a comprehensive analysis of matrix materials, fillers, processing techniques, and emerging applications, this review aims to provide valuable insights for the future development of sustainable and multifunctional nanomaterials.



KEYWORDS: epoxy, vitrimer, nanocomposite, filler, dynamic covalent bond

1 Introduction

Epoxy resin, a thermosetting polymer, is indispensable in a wide range of applications—from everyday products to advanced industries such as aerospace and energy equipment. This broad utility stems from its exceptional mechanical strength, dimensional stability, and chemical resistance, which are derived from its permanently covalently cross-linked network [1–5]. However, this same permanent network, although responsible for its superior performance, leads to considerable environmental drawbacks: once cured, conventional epoxy thermosets cannot be melted or dissolved, rendering them extremely difficult to recycle. The global epoxy resin market is projected to reach USD 37.3 billion by 2025, yet less than 10% of them are currently recycled [6]. Currently prevalent disposal methods, such as landfilling or incineration, entail significant resource waste and further exacerbate environmental issues [7–12]. Consequently, the development of advanced materials that retain the superior performance of traditional thermosets while incorporating the reprocessability of thermoplastics represents a critical challenge in polymer science. Developing inherently recyclable materials can reduce waste generation from the source compared to end-of-life recycling.

To meet this challenge, researchers have turned to dynamic chemistry, aiming to incorporate reversible bonds within cross-linked networks to create dynamic or smart materials [13–18].

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Many noncovalent interactions—such as hydrogen bonds, ionic interactions, and host–guest recognition—are inherently reversible structures. Many non-covalent interactions—such as hydrogen bonds, ionic bonds, and host–guest recognition—are inherently reversible. Introducing these as reversible sites into polymer networks has achieved a certain degree of reprocessability. However, their lower bond energies often result in materials with low macroscopic modulus and poor creep resistance, limiting their suitability for load-bearing or harsh environments. The incorporation of robust dynamic covalent bonds (DCBs) into cross-linking networks combines the strength of covalent linkages with the adaptability of dynamic covalent chemistry, thereby enabling the construction of covalent adaptable networks (CANs) [19–24].

In CANs, bond exchange of DCBs proceeds through two stages: cleavage of existing bonds and formation of new ones. According to the sequence of these stages and the crosslinking state of the intermediate network, CANs are typically classified into dissociative and associative types [25, 26] (Fig. 1). In dissociative CANs [27–30] (e.g., systems based on the Diels–Alder reaction), the exchange reaction begins with the cleavage of the original DCBs. This leads to a pronounced temporary decrease in the network crosslinking density during the intermediate state, causing the network to lose its three-dimensional integrity and chemical stability temporarily. Such a reduction may induce a substantial "flow" state, which can significantly compromise material stability. Vitrimers, commonly categorized as associative CANs, represent a third class of materials distinct from conventional thermosets and thermoplastics [25, 26, 31–34]. Their associative exchange mechanism enables vitrimers to maintain a constant network crosslinking density even under processing conditions, thereby preserving material integrity effectively. Therefore, vitrimers exhibiting thermoplastic-like reprocessability while maintaining the crosslinking stability. Vitrimers exhibit insolubility in non-reactive solvents while retaining melt-processability and reshapeability at elevated temperatures. First synthesized and defined by Leibler and coworkers in 2011 [31], the bond exchange reaction within a vitrimer network begins with the formation of an adduct at the dynamic crosslink (where new and old bonds connect at the same site), followed by cleavage of the original linkage. The characteristic temperature at which the crosslinked network undergoes topological rearrangement under thermal stimulus is defined as the topology freezing transition temperature (T_v). When the temperature exceeds T_v, rapid reversible bond exchange occurs, granting vitrimers macroscopic fluidity and enabling properties such as self-healing, welding, and reshaping [31, 32, 35–39].

Since this pioneering work, transesterification-catalyzed epoxy vitrimer networks and their catalysts have been extensively investigated [32, 33, 40–46]. Anhydride/dicarboxylic acid-cured epoxy resins constitute a major subclass, characterized by abundant ester bonds and hydroxyl groups within their network structures, which provide a suitable chemical environment for vitrimer formation. Transesterification-based epoxy vitrimers can be prepared by incorporating catalysts that promote bond exchange during the curing process. For other widely used curing systems, such as amine-cured epoxies, structural design is required to impart vitrimer behavior. This involves integrating dynamic covalent chemistries—including disulfide exchange [47–53], imine exchange [54–57], borate ester exchange [58–61], and siloxane exchange [62–65]—into the polymer network, yielding structurally diverse and reprocessable epoxy materials. Such integration typically demands careful monomer selection and design. Strategies for incorporating DCBs into epoxy networks can be categorized into two main approaches. The first employs epoxy monomers or curing agents bearing DCB units, introducing exchangeable linkages at non-crosslinking sites without altering the original curing mechanism [47, 49, 57, 64, 66, 67]. The second strategy involves developing novel epoxy curing reactions where DCB formation serves as the dynamic crosslinking sites [68, 69]. For example, prepolymers or linear chains can be engineered to undergo final crosslinking via reactions between boronic acid and hydroxyl groups, creating dynamic boronic ester linkages and resulting in epoxy vitrimers based on boronic ester exchange.

In traditional epoxy resins, nanofillers are widely introduced to prepare nanocomposites, to enhance material properties and expand potential functional capabilities [70–77]. This also applies to epoxy vitrimers. Unlike traditional epoxy resins, not only the polymer matrix is enhanced by nanofillers, but the properties of fillers are enriched by vitrimer matrix in vitrimer-based nanocomposites. Specifically, the incorporation of nanofillers into epoxy vitrimers modulates the macroscopic properties of polymer

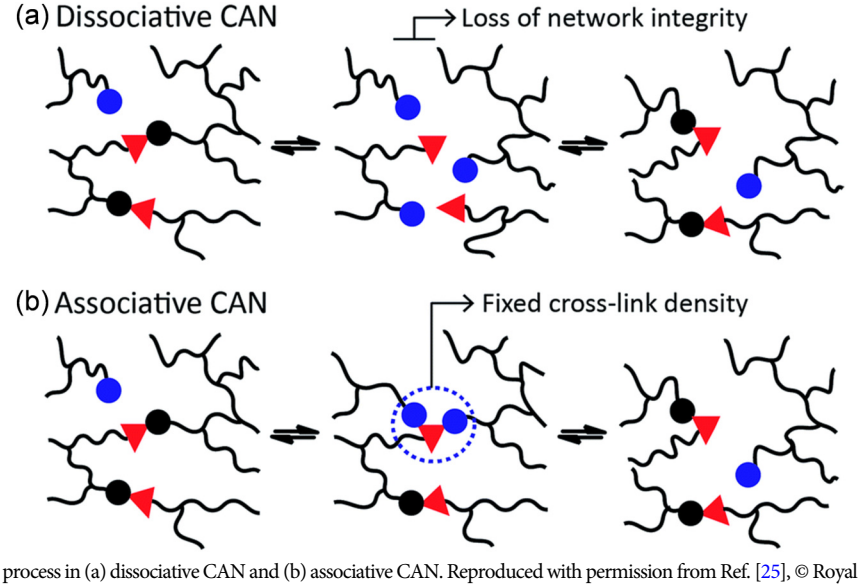

Figure 1 The bond exchange process in (a) dissociative CAN and (b) associative CAN. Reproduced with permission from Ref. [25], © Royal Society of Chemistry 2015.

and participates in bond exchange reactions and the network formation [5, 35, 78–83]. Topological rearrangement is no longer confined to the polymer interior, with diversified stimulus modes to trigger it. The enhancement in mechanical properties originates from the high specific surface area of nanofillers uniformly dispersed within the polymer matrix. This dispersion strengthens interfacial interactions and promotes efficient stress transfer, thereby improving the overall strength and toughness of the material. In addition, owing to the inherent properties of the nanomaterials—such as high photothermal conversion efficiency [84–87] and paramagnetism [88]—vitrimers gain enriched dynamic behavior and multifunctionality. The resulting composites exhibit added capabilities including electrical conductivity, thermal conductivity, and enhanced barrier performance. Consequently, vitrimer nanocomposites extend beyond conventional thermally activated reversible properties; their network rearrangement can be precisely and efficiently controlled via external stimuli such as light or magnetic fields, depending on the functional characteristics of the nanofillers [35, 89–94]. Furthermore, the reversible nature of vitrimers enables convenient recovery and reuse of valuable or difficult-to-separate nanofillers from the polymer matrix. This synergistic interplay between vitrimer matrices and nanofillers has led to the emergence of novel sustainable multifunctional nanocomposites.

This review aims to present recent advances in epoxy-based vitrimer nanocomposites. We begin by introducing the diverse dynamic covalent chemistries utilized in epoxy vitrimers and outlining strategies for constructing their matrix materials. We then categorize the various nanoparticle systems, composite preparation approaches, and characterization methods employed in this field. Finally, we discuss the multifunctional properties of these nanocomposites—including enhanced mechanical performance, stimulus responsiveness, and electrical/thermal conductivity—and highlight their emerging applications in areas such as soft robotics, electromagnetic interference shielding, and smart sensors.

2 The design principle of epoxy vitrimer and different dynamic systems in nanocomposites

The two principal subclasses of epoxy resins are anhydride-cured and amine-cured systems [6] (Fig. 2). Traditional anhydride-cured epoxies inherently possess the potential to form vitrimers, as their crosslinked networks contain abundant hydroxyl and ester groups—a chemical environment conducive to dynamic transesterification. Thus, by introducing suitable catalysts and optimizing monomer ratios, anhydride-cured epoxy resins can be transformed into vitrimers with covalent adaptable networks capable of topological rearrangement. In contrast, amine-cured epoxy resins, which represent the most widely used category beyond anhydride systems [95, 96], typically form permanently crosslinked networks. The C–N bonds generated during curing are not dynamically reversible, and the networks lack inherent dynamic covalent bonds (DCBs). Consequently, converting conventional amine-cured epoxies into vitrimers requires deliberate molecular design. This can be achieved either by functionalizing the curing agents or epoxy monomers to introduce DCBs at non-crosslinking points within the final network, or by designing the curing chemistry itself so that DCB formation serves as the crosslinking event. In the latter strategy, the reaction that creates DCBs—such as boronic ester or imine formation—is utilized to build the epoxy network, yielding a dynamically crosslinked vitrimer. This section reviews the bond-exchange reactions currently employed in preparing epoxy vitrimers for nanocomposites. It outlines the underlying chemical principles and network-construction strategies, offering practical guidance for selecting suitable vitrimer matrices in nanocomposite design.

2.1 Transesterification-based epoxy vitrimer substance

Transesterification is a reversible reaction in which hydroxyl and ester groups exchange to form new hydroxyl and ester bonds under acid or base catalysis [31, 32, 46]. This process proceeds through an addition-elimination mechanism, with each step being reversible. As the earliest studied and one of the most extensively investigated dynamic bond exchanges in epoxy vitrimers (Fig. 3), transesterification benefits from the inherent abundance of hydroxyl and ester bonds in traditionally synthesized anhydride-cured epoxies. During preparation, epoxy monomers and anhydrides are polymerized in the molten state, with transesterification catalysts uniformly premixed into the melt. Consequently, compared to conventional anhydride-cured epoxy resins, the production of epoxy vitrimers primarily differs in the raw material formulation and the inclusion of a transesterification catalyst. The resulting epoxy vitrimer retains mechanical strength comparable to that of traditional resins at service temperatures,

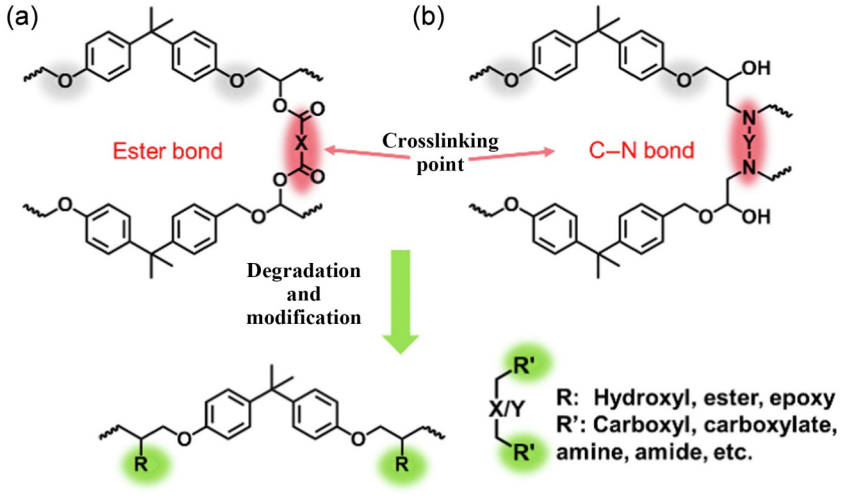

Figure 2 The general structures of (a) anhydride-cured epoxies and (b) amine-cured epoxies. Reproduced with permission from Ref. [6], © Elsevier Ltd. 2023.

while exhibiting rapid topological network rearrangement at elevated temperatures, thereby endowing the material with thermally reconfigurable properties.

In epoxy vitrimer nanocomposites, nanofillers are commonly introduced by premixing with the monomer precursors prior to curing. Our group was among the first to incorporate carbon nanotubes (CNTs)—known for their high photothermal conversion efficiency—into an epoxy vitrimer matrix (CNT-vitrimer) [41]. This integration allows the dynamic covalent network of the vitrimer to be remotely triggered and manipulated via photothermal effects, thereby achieving unprecedented photo-welding and self-healing capabilities and overcoming the limitation of thermal responsiveness alone. The vitrimer matrix was synthesized via a classical epoxy–dicarboxylic acid reaction system, using bisphenol A diglycidyl ether (DGEBA) as the epoxy monomer, adipic acid as the dicarboxylic acid crosslinker, and triazabicyclodecene (TBD) as both the transesterification reaction catalyst and curing reaction accelerator (Fig. 4(a)). The resulting

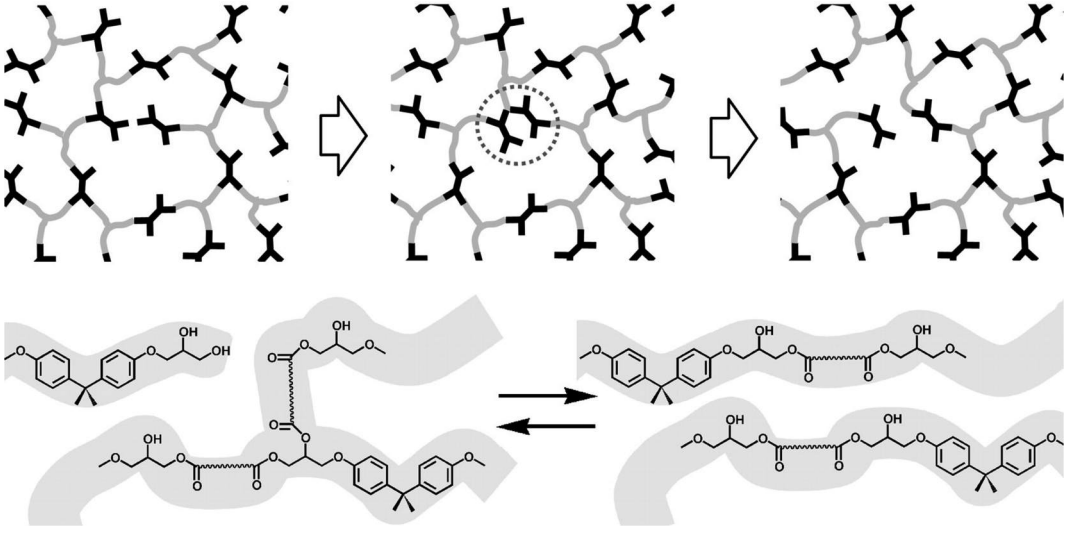

Figure 3 The topological rearrangement of epoxy vitrimer based on transesterification without loss of network integrity. Reproduced with permission from Ref. [31], © The American Association for the Advancement of Science 2011.

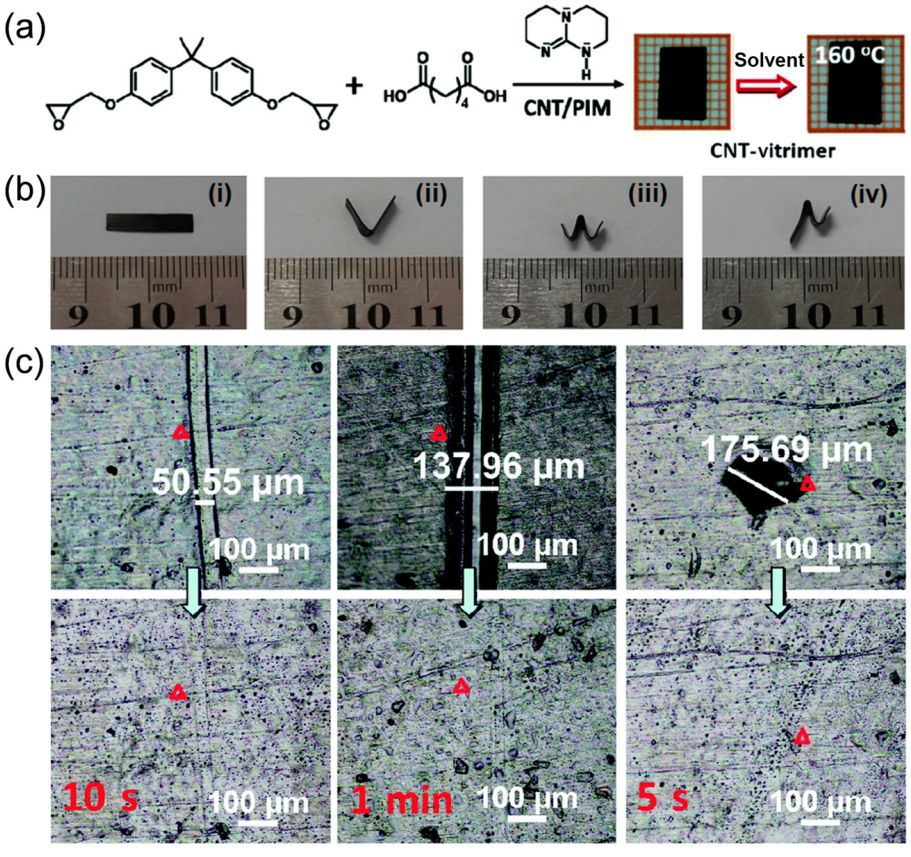

Figure 4 (a) Synthesis of CNT-vitrimer. (b) Light-induced partial shape memory and (c) light triggered healing of CNT-vitrimer with different damage. Reproduced with permission from Ref. [41], © Royal Society of Chemistry 2014.

epoxy vitrimer exhibits a glass transition temperature (T_g) of approximately 39–45 °C and a topology freezing transition temperature (T_v) around 160 °C. Leveraging the high photothermal conversion efficiency of CNTs enables localized heating above T_v under light irradiation (particularly in the infrared range), facilitating remote and spatially precise photo-controlled shape memory and repair (Figs. 4(b) and 4(c)). The distinctive properties of nanofillers thus significantly broaden the functional scope of nanocomposites, enabling responsive and actuation behaviors in complex environments under diverse stimulus modes.

In transesterification-based epoxy vitrimer nanocomposites, the curing reaction typically involves commercial epoxy monomers, most commonly bisphenol A-type epoxies [42, 45, 64, 89, 97, 98] and long-chain dicarboxylic acids [35, 41, 91, 99, 100]. The long hydrocarbon chains of these diacids reduce the crosslinking density of the network by increasing the chain segment length and spacing between crosslinks. This structural adjustment enhances the flexibility and ductility of the material, effectively mitigating the inherent brittleness of conventional epoxy resins and expanding their potential applications. Excessive brittleness and hardness would otherwise hinder vitrimer deformation and severely limit the macroscopic manifestation of dynamic chemistry. Beyond adipic acid, other diacids such as sebacic acid and dodecanedioic acid have also been utilized as curing agents for vitrimer matrices in nanocomposites (Fig. 5) [99, 100]. The epoxy monomer and dicarboxylic acid curing agent are generally employed in a 1:1

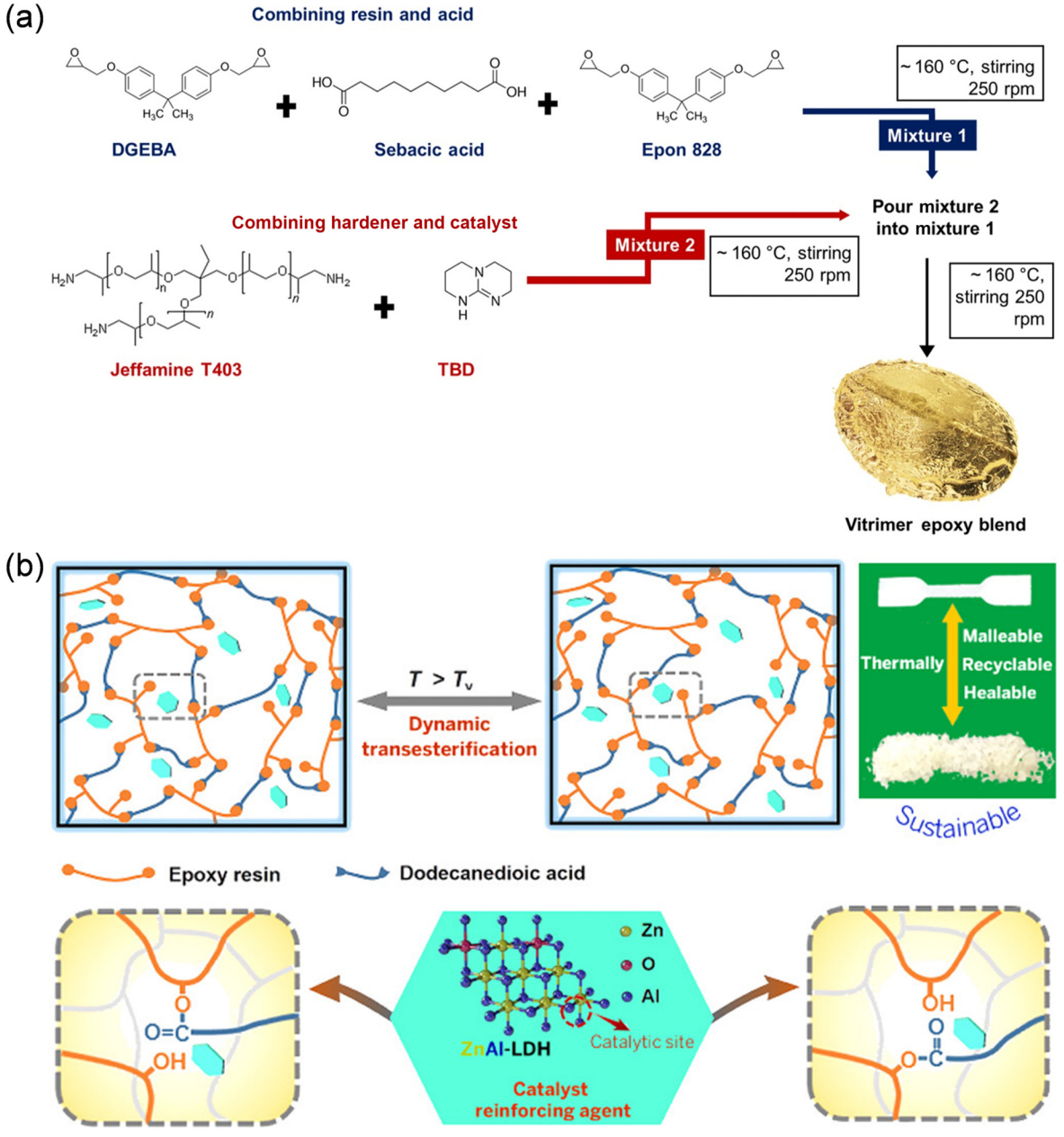

Figure 5 (a) Scheme for the synthesis of epoxy vitrimer substance cured with sebacic acid in transesterification-based vitrimer nanocomposites. Reproduced with permission from Ref. [99], © Elsevier Ltd. 2025. (b) Schematic illustration of transesterification-induced topological rearrangement of vitrimer substance cured with dodecanedioic acid and structure of ZnAl-layered double metal hydroxide (ZnAl-LDH) nanosheets. Reproduced with permission from Ref. [100], © American Chemical Society 2021.

equivalent ratio. This stoichiometry ensures complete reaction and full crosslinking while preserving the hydroxyl groups generated from epoxy ring-opening, allowing them to participate effectively in the vitrimer's transesterification exchange reactions.

In transesterification-based epoxy vitrimers, catalyst selection is a critical consideration. For nanocomposite systems, TBD [33, 41, 89, 101, 102] is often the preferred catalyst owing to its high efficiency and better compatibility with organic matrices compared to highly active metal salt alternatives [31, 32]. To circumvent compatibility and processing challenges associated with externally added catalysts, subsequent efforts have been directed toward catalyst-free or intrinsically catalyzed epoxy vitrimer systems [32, 42, 103–106]. These systems typically rely on carefully designed chemical environments that exploit neighboring group effects or hyperbranched epoxy architectures. To date, few studies have incorporated nanofillers into catalyst-free transesterification-based vitrimer composites. While the addition of nanofillers may introduce novel functionalities, the feasibility of such systems—both in terms of chemical environment design and practical processing—requires thorough evaluation.

2.2 Disulfide exchange-based epoxy vitrimer substance

Disulfide bonds are a classic type of DCB that undergo bond exchange without requiring external catalysts, and have been widely studied over an extended period [67, 107–114]. Compared to ester bonds, disulfide exchange occurs more readily under mild conditions. The exchange mechanism is relatively complex, proceeding mainly through three pathways: thiol-disulfide exchange, disulfide-disulfide exchange, and radical-mediated exchange (Fig. 6) [113]. The dominant mechanism depends on the chemical environment surrounding the disulfide linkage. For example, aromatic disulfides exhibit more efficient exchange than aliphatic disulfides. While the former can induce topological rearrangement in epoxy vitrimers under mild conditions, the latter typically require elevated temperatures for reprocessing.

Disulfide-bond-containing epoxy vitrimers are widely employed in nanocomposite systems. 4,4′-Diaminodiphenyl disulfide (4-AFD) is a commonly used curing agent that is commercially available as a standard polymer synthesis material [115–119]. Its structure resembles that of conventional amine curing agents, while introducing dynamic disulfide bonds into the cured network. The aromatic moieties flanking the disulfide bond confer high exchange activity, enabling network formation without external catalysts. Consequently, epoxy vitrimers based on disulfide exchange can be prepared using processing methods identical to those of conventional bulk epoxy thermosets. Nanofillers can be incorporated prior to mixing the curing agent with epoxy monomers, allowing them to be embedded uniformly within the final cured network. In such disulfide-exchange-based epoxy vitrimer systems, the epoxy monomer is typically a commercially available epoxy oligomer, most often a bisphenol A-type epoxy resin.

Bohra et al. prepared an epoxy vitrimer by curing Epon 828 resin with the 4-AFD curing agent at elevated temperature, thereby incorporating the inherent dynamic disulfide bonds from 4-AFD into the network backbone (Fig. 7(a)) [120]. This yielded a dynamic vitrimer based on aromatic disulfide exchange and its corresponding functionalized graphene nanocomposites. The

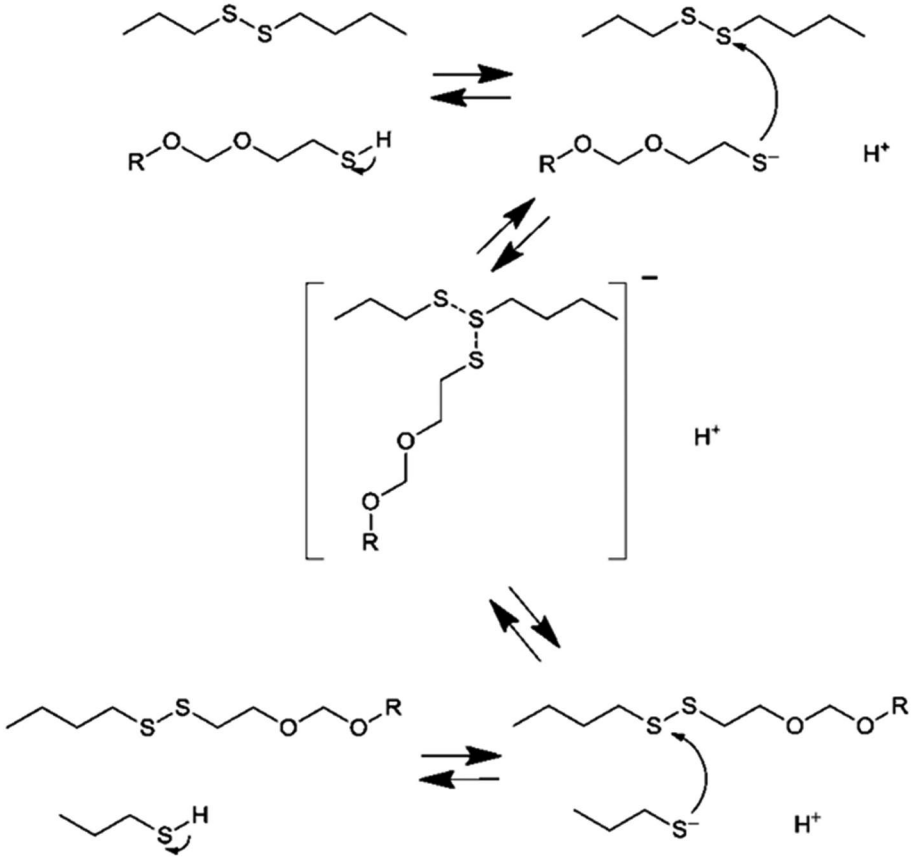

Figure 6 Illustration of the echanism of disulfide exchange: deprotonation of the thiol group, nucleophilic attack of the thiolate on one of the sulfur atoms of the disulfide followed by dissociation of the intermediate product Reproduced with permission from Ref. [113], © Royal Society of Chemistry 2013.

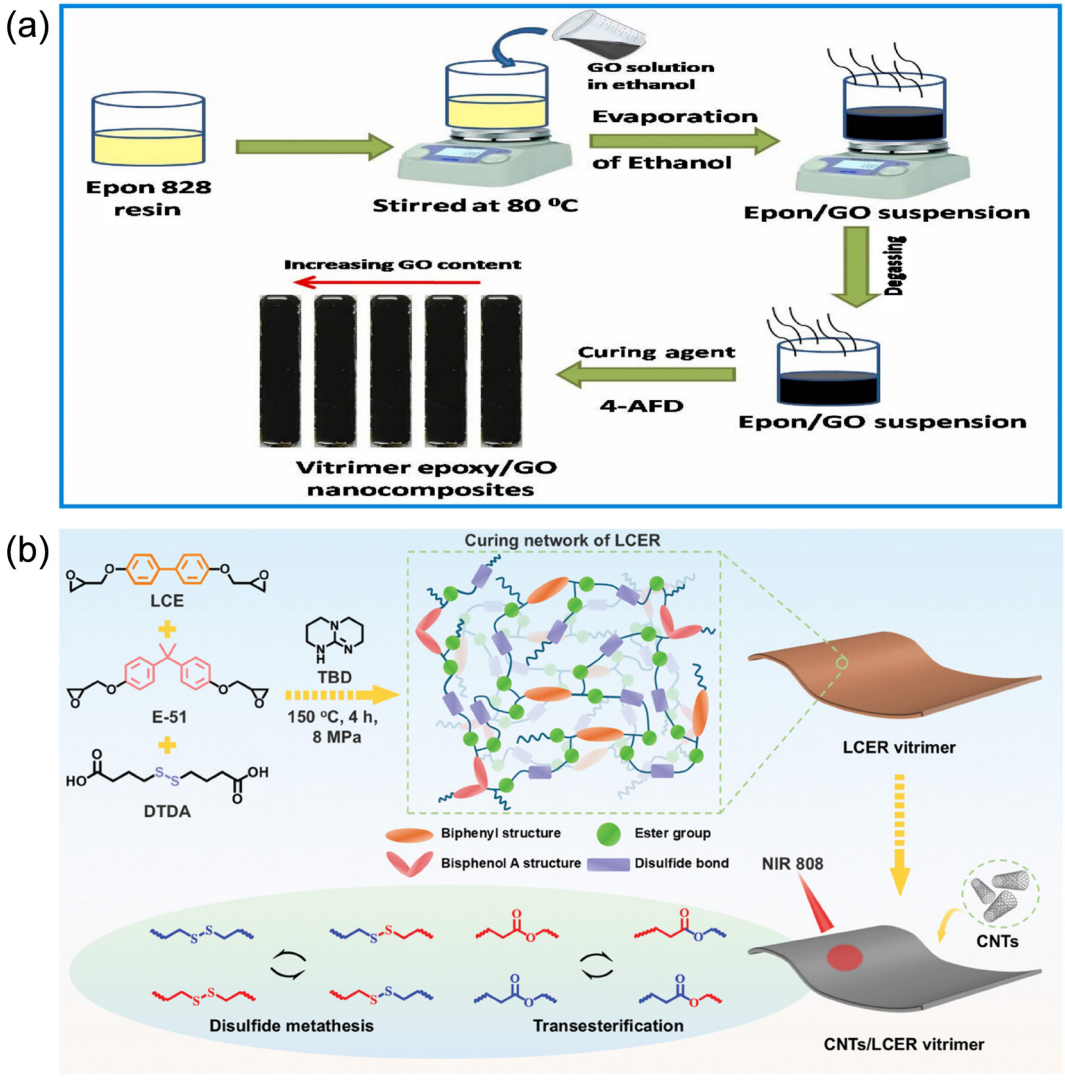

Figure 7 (a) Schematic procedure of the preparation of epoxy vitrimer nanocomposites cured by 4-AFD. Reproduced with permission from Ref. [120], © Elsevier Ltd. 2023. (b) Preparation of epoxy vitrimer/CNTs composite and the mechanism of network rearrangement enabled by disulfide metathesis and carboxylate transesterification. Reproduced with permission from Ref. [124], © Wiley-VCH GmbH 2024.

vitrimer network exhibited a glass transition temperature (T_g) of approximately 95 °C. In contrast, Lorero et al. [121] synthesized an epoxy vitrimer by stoichiometrically mixing DGEBA with 4-AFD and curing the mixture, achieving a T_g of about 141 °C. Incorporation of only 0.1 wt.% CNTs increased the T_g from 141 °C (pure vitrimer) to 165.3 °C. Although both vitrimers utilize the same curing agent, the significant T_g difference likely arises from the distinct structures of the epoxy monomers used. Furthermore, the authors investigated the effect of excess 4-AFD on the network's thermal properties. Compared to the stoichiometric formulation (T_g = 141 °C), a 10% excess of curing agent raised the T_g to 155 °C, whereas a 20% excess lowered it to 137 °C. These findings indicate that the properties of epoxy vitrimers can be effectively tuned not only by the choice of epoxy oligomer structure but also through precise control of curing agent stoichiometry.

Furthermore, nanocomposites cured with 2,2′-diaminodiphenyl disulfide (2-AFD) operate on principles similar to the 4-AFD system [122, 123], differing primarily in the position of the amine substituent. This structural isomerism influences the conformation of molecular chains within the crosslinked network, thereby modulating the final material properties. Beyond amine-based curing agents, disulfide bonds can also be incorporated into ester-linked networks through the molecular design of diacid curing agents. Employing dicarboxylic acids containing disulfide moieties to cure epoxy resins yields crosslinked networks possessing both ester and disulfide dynamic covalent bonds (DCBs), which can synergistically enhance the dynamic properties [47, 124]. For instance, Zhang et al. utilized 4,4′-dithiobutanedioic acid (DTDA) to cure epoxy resin, producing a vitrimer where the curing agent introduces both ester crosslinks and segment-integrated disulfide bonds as concurrent dynamic exchange sites (Fig. 7(b)) [124]. With TBD as a catalyst, both disulfide and ester bonds in the network can undergo simultaneous exchange upon heating, significantly enhancing the material's dynamic characteristics. This design of a dual-dynamic-bond network exemplifies a strategic route to achieving highly dynamic epoxy vitrimer matrices. In a related approach, Zheng et al. developed a novel method for preparing disulfide-containing polyacid curing agents (Fig. 8) [125]. They synthesized poly(α-lipoic acid) (PLA) via ring-opening polymerization of α-lipoic acid, resulting in a polymer with

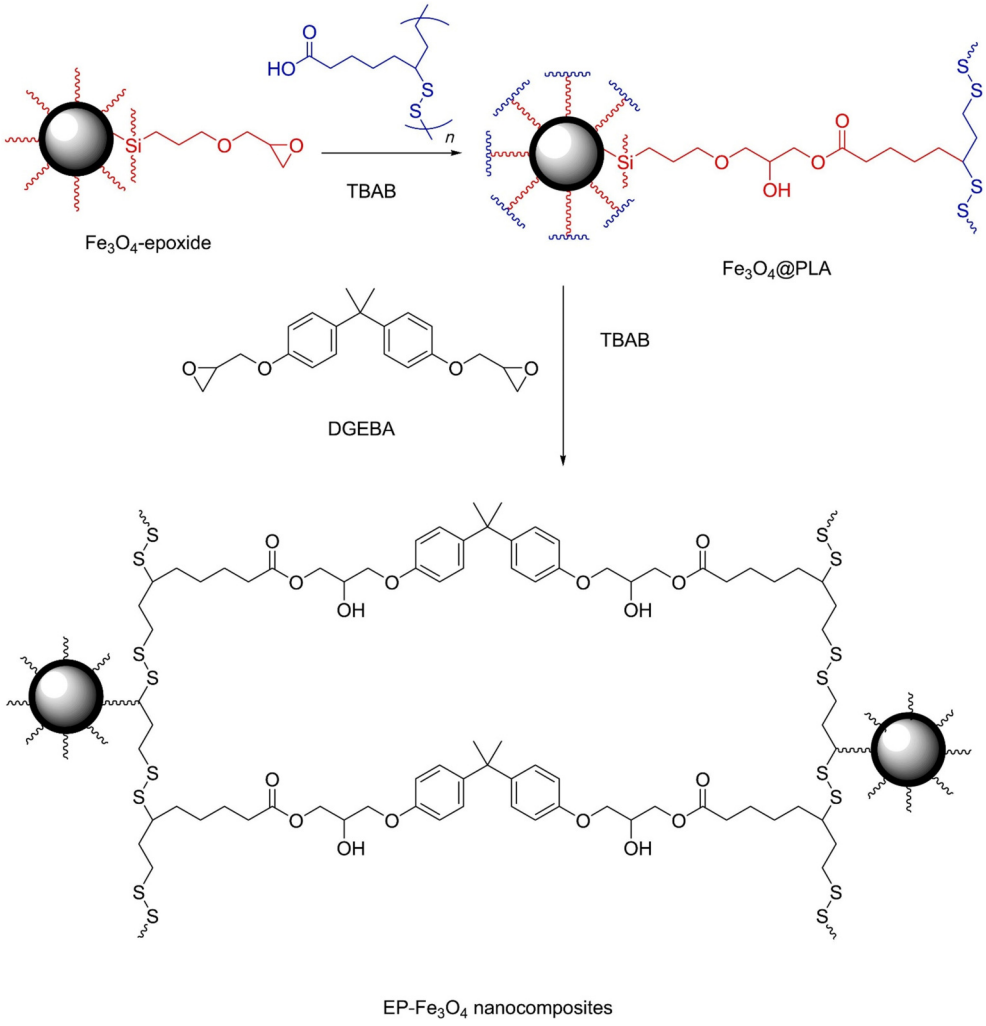

Figure 8 Synthesis of epoxy vitrimer nanocomposites with a polymeric hardener carrying disulfide bonds. Reproduced with permission from Ref. [125], © Elsevier Ltd. 2023.

dynamic disulfide bonds along its backbone and terminal carboxyl (–COOH) groups. This PLA was subsequently employed as a curing agent for epoxy resins, reacting with DGEBA to form epoxy vitrimers.

In summary, dynamic covalent chemistry based on disulfide bonds provides versatile design strategies for constructing epoxy vitrimer networks. By incorporating exchangeable disulfide bonds into either epoxy monomers or curing agents while retaining the original curing chemistry, recyclable vitrimers and their composites emerge as promising alternatives to conventional epoxy resins in many applications. The high reactivity of aromatic disulfide bonds not only ensures efficient bond exchange but also facilitates mild chemical recycling of disulfide-containing vitrimers. This capability supports responsible end-of-life management of related products and enables the potential recovery and reuse of embedded nanofillers. Meanwhile, aliphatic disulfide-based systems retain significant research and practical value, particularly in applications where higher repair or reprocessing temperatures are operationally acceptable or required.

2.3 Imine-based epoxy vitrimer substance

Imines, also known as Schiff base structures [20, 126], constitute another highly reactive class of dynamic covalent bonds (DCBs) capable of undergoing bond exchange without requiring external catalysts or thermal input. They offer advantages including straightforward preparation, rapid exchange kinetics, and high efficiency in material repair. Formed via dehydration condensation between primary amines and aldehydes, imines are inherently reversible linkages and are susceptible to hydrolysis in the presence of water. In the dynamic formation–cleavage equilibrium, acidic conditions generally accelerate imine hydrolysis, while neutral to slightly alkaline environments enhance their stability. This pH responsiveness renders imine bonds ideal reversible units for constructing pH-sensitive materials. Additionally, the molecular structure significantly influences imine stability. When both the aldehyde and amino groups are attached to aromatic rings, the resulting imine typically exhibits greater stability [127]. Conversely, imines derived from aliphatic counterparts tend to undergo spontaneous hydrolysis. Leveraging the reversible formation–hydrolysis equilibrium of imines enables the design of moisture-driven polymer networks, allowing moist powders to be molded into cohesive sheets without external heating. Moreover, the pH responsiveness of imine bonds can be exploited as degradable sites within crosslinked polymers, facilitating efficient network depolymerization [128].

The dynamic behavior of imine bonds extends beyond

formation and hydrolysis to encompass transamination and imine metathesis reactions (Fig. 9) [20]. In transamination, free amine groups displace the original amines within an imine, generating a new imine bond while releasing the former amine—a process analogous to the exchange between esters and hydroxyl groups in transesterification. The most direct application of imine chemistry lies in synthesizing reparable and recyclable polyimine vitrimers. Capitalizing on the highly efficient dynamic character of imines, numerous high-performance reversible epoxy vitrimers have also been developed. Since epoxy resins do not inherently contain aldehyde groups, the introduction of aldehydes into the crosslinked network requires deliberate molecular design of the monomers.

Consequently, imine-mediated dynamic covalent epoxy networks generally cannot employ industrially prevalent epoxy monomers directly. Instead, specific molecular architectures must be designed to integrate both imine and epoxy functionalities into the monomer or curing agent. In this context, vanillin emerges as a prominent building block [128–133]. Derived from bio-based lignin, it possesses multiple readily modifiable functional groups, making it a frequent starting point for constructing imine-based dynamic epoxy networks. Both epoxy monomers and curing agents have been designed from vanillin derivatives. For example, Wang et al. [134] synthesized a fully bio-based dynamic curing agent (VDA) containing imine linkages via a condensation reaction between vanillin and 1,10-diaminodecane. This agent was subsequently cured with epoxidized soybean oil (ESO) at 150 °C (Fig. 10). The dihydroxy structure of the designed curing agent necessitates a hydroxyl-epoxy ring-opening reaction for curing, which exhibits lower reactivity compared to conventional amine curing and therefore requires a catalyst. Leveraging the hydrolysis of imines under weakly acidic conditions enables closed-loop recycling of the nanofillers and composites, aligning with sustainable design principles. The prevalent use of raw materials such as vanillin and ESO in imine-based epoxy vitrimers has further stimulated interest in bio-based feedstocks, aiming to reduce dependence on non-renewable petrochemical resources.

While imine bonds exhibit high dynamic efficiency, their intrinsic reactivity also compromises the stability of the resulting materials, thereby limiting their practical application scope. For instance, exposure to acidic environments or high humidity must be avoided during service, as these conditions promote imine bond cleavage. This humidity and acid sensitivity can also be leveraged in various application scenarios such as environmental monitoring devices.

2.4 Siloxane-based epoxy vitrimer substance

Siloxane exchange involves the nucleophilic attack of silanolate anions on siloxane bonds, leading to siloxane bond rearrangement [64, 135, 136]. Its mechanism is analogous to a bimolecular nucleophilic substitution reaction [135]. Although this reaction was reported as early as the 1950s, it garnered little attention initially. With advancing technological demands, the emergence of self-healing silicone elastomers as a research focus has drawn renewed interest to the dynamic silanolate exchange within siloxane networks. The first incorporation of siloxane chemistry into

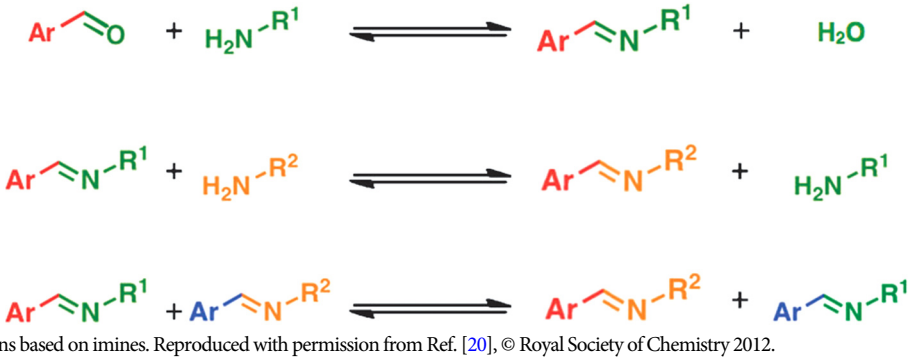

Figure 9 Dynamic reactions based on imines. Reproduced with permission from Ref. [20], © Royal Society of Chemistry 2012.

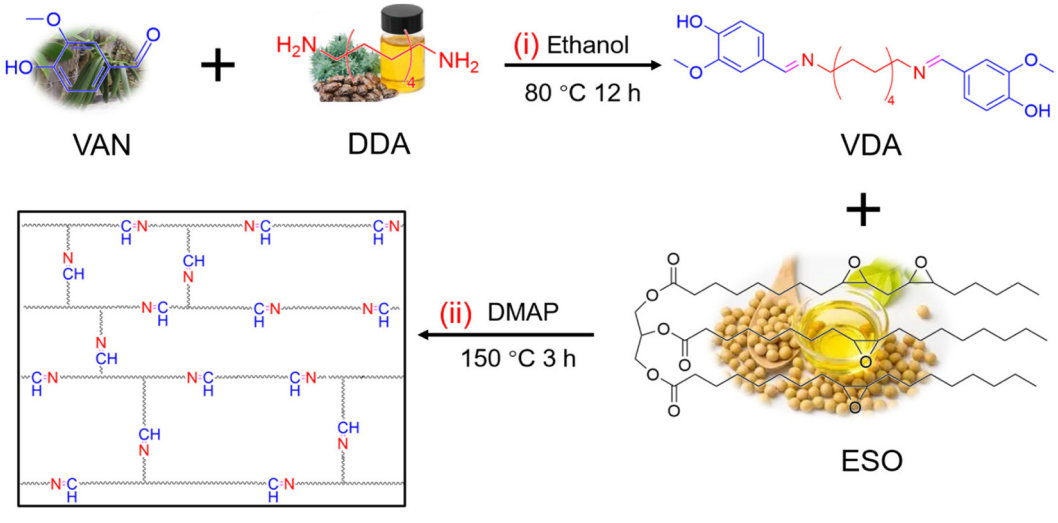

Figure 10 Preparation of all-biobased curing agent (VDA) and epoxidized soybean oil-based vitrimer. Reproduced with permission from Ref. [134], © American Chemical Society 2024.

dynamic materials was reported in 2012 [136], after which studies exploring siloxane and silyl ether chemistry in CANs have gradually emerged. Nevertheless, compared to other dynamic bond exchange reactions, the exchange rate of siloxane bonds in polymeric materials is relatively slow, limiting their processability and broader utility. To address this, several studies have focused on accelerating the silanolate exchange in vitrimers, primarily through optimizing catalyst loading and developing more efficient catalytic systems [64, 65, 135].

Siloxane exchange has also been implemented in epoxy vitrimer nanocomposites. Dong et al. [137] reported an innovative design in which imine and siloxane bonds were integrated into a single epoxy vitrimer network. Here, the imine moiety functioned as a reversible carrier for the catalyst, establishing a unique "shuttle catalyst" mechanism that significantly accelerated the siloxane exchange rate (Fig. 11). The researchers synthesized a novel curing agent (APDI) containing both imine and siloxane dynamic bonds. When cured with DGEBA epoxy resin using TBD as catalyst, it produced a vitrimer network incorporating this dual-dynamic covalent architecture. The resulting epoxy vitrimer exhibits high mechanical strength (tensile strength of 43.75 MPa; T_g of 78.1 °C) combined with a rapid dynamic response (stress relaxation time of 384 s at 100 °C). The material achieves substantial self-healing within only 10 min at a mild temperature (90 °C), displays excellent thermal reprocessability (1 h at 120 °C under 16 MPa), and can be chemically degraded and recovered under acidic conditions. Furthermore, the incorporation of nanoparticles extends its functionality, enabling features such as light-triggered repair.

2.5 Boronic ester-based epoxy vitrimer substance

Boronic ester bonds, formed by the reaction of boronic acid with diols, represent a class of reversible DCB [138–141]. Similar to the exchange mechanisms of ester and imine bonds, hydroxyl groups within the network can substitute those in the existing boronic ester to participate in bond exchange reactions. During the design of epoxy vitrimers, a common strategy to incorporate boronic ester bonds involves structurally modifying either the epoxy monomers or the curing agents, while retaining the original curing chemistry and crosslinked network architecture.

The reaction between boronic acid groups and hydroxyl groups to form boronic esters can also be designed to serve as a novel epoxy curing mechanism, wherein boronic esters themselves act as crosslinking sites within the epoxy network.

This approach enables the preparation of vitrimers containing dynamic boronic esters by crosslinking linear epoxy prepolymers bearing hydroxyl groups with polyboronic acids.

In nanocomposites, boronic esters can also serve as the dynamic covalent component of the vitrimer matrix. In the work reported by Huang et al., 4-carboxyphenylboronic acid pinacol ester (CAPE) was employed as a linker molecule [142]. Its carboxyl group first underwent a ring-opening reaction with the epoxy groups of epoxidized natural rubber (ENR), thereby introducing boronic ester moieties onto the rubber chains. Subsequently, surface-modified silica nanoparticles rich in hydroxyl groups were covalently linked to the boronic ester groups on the modified rubber side chains via an exchange reaction, constructing an interface-crosslinked vitrimer network based on dynamic covalent boronic ester bonds—where the silica nanoparticles act as crosslinking points within the covalent network (Fig. 12). Through this ingenious chemical design, the incorporation of nanofillers and the synthesis of the vitrimer were accomplished in a single integrated process. The nanofillers were designed to function as crosslinking nodes within the network, enabling the preparation of the nanocomposite. Compared to physically dispersed composites, this covalent incorporation of nanofillers into the vitrimer network significantly enhances material stability. Benefiting from the dynamic exchange of boronic ester bonds, the material exhibits self-healing at elevated temperatures (achieving 98% strength recovery after 24 h at 150 °C) and can be thermally reprocessed (retaining over 90% strength after recovery at 190 °C). Moreover, it can be dissolved and recycled via solvent-assisted transesterification reactions. Notably, this system operates without external catalysts by directly utilizing the surface hydroxyl

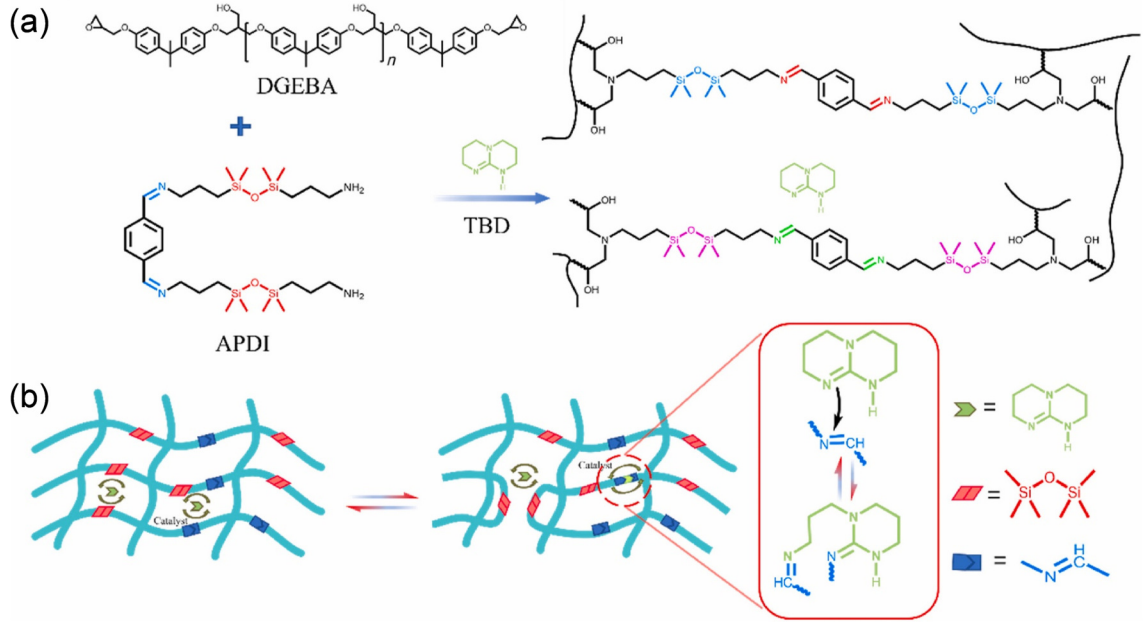

Figure 11 (a) Synthesis and chemical structure of dynamic epoxy network based on TBD catalyzed imine and siloxane exchanges. (b) Illustration of the self-healing and recovery of epoxy vitrimer catalyzed by TBD shuttle catalyst. Reproduced with permission from Ref. [137], © Elsevier Ltd. 2025.

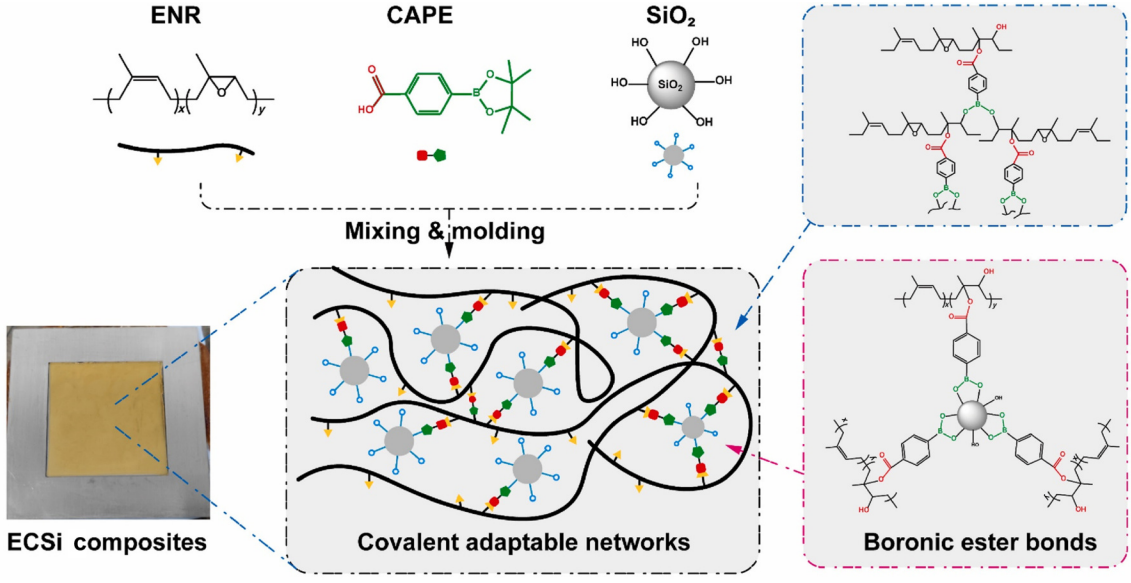

Figure 12 Construction of epoxidized rubber vitrimer based on boronic esters. Reproduced with permission from Ref. [142], © Elsevier Ltd. 2022.

groups of commercial silica, offering a simple and versatile dynamic interfacial crosslinking strategy for constructing high-performance, recyclable rubber-based vitrimer elastomers and flexible electronic devices.

3 A brief introduction of vitrimer-based nanocomposites and preparation process

The various epoxy vitrimer types discussed in previous section serve as matrixes for nanocomposites, where understanding of vitrimer design principles lays the groundwork for developing composite materials. In the field of advanced composites, material design primarily follows two distinct pathways. The first employs continuous fibers as reinforcement to fabricate structural composites [143–146]. The second utilizes nanofillers as functional modifiers to prepare nanocomposites with enhanced or novel properties [72, 147–152]. These two categories differ fundamentally in their reinforcement mechanisms, target property profiles, and underlying design philosophies.

In traditional fiber-reinforced epoxy composites—such as those using carbon or glass fibers—the design focuses on mechanical performance at the macroscopic scale. In these systems, high-performance fibers constitute the majority of the volume and serve as the primary load-bearing elements, forming the structural skeleton of the material. They efficiently transfer and carry the bulk of the applied stress. Here, the epoxy matrix acts principally as an effective load-transfer medium and a geometrically stable phase. It impregnates, bonds, and anchors the fibers together, thereby translating the intrinsic superior properties of the fibers into the overall macroscopic performance of the composite. Consequently, the mechanical properties of such materials are predominantly determined by the characteristics, orientation, and volume fraction of the reinforcing fibers.

By contrast, the design of epoxy-based nanocomposites centers on interfacial interactions and nanoscale effects. Rather than incorporating macroscopic load-bearing elements, this approach utilizes the exceptionally high specific surface area and unique functional properties of nanofillers—such as carbon nanotubes and graphene—to reconstruct and enhance the epoxy matrix itself at the molecular and nanoscopic levels. Even at very low filler loadings, these materials can induce qualitative transformations in matrix properties. They not only improve mechanical properties—such as strength, modulus, and fracture toughness—and thermal stability in a synergistic manner, but also introduce functionalities originally absent in the neat epoxy, including electrical conductivity, thermal conductivity, and enhanced barrier performance.

Crucially, within the vitrimer matrix, nanofillers can participate in the bond exchange reactions of the dynamic network and enhance the adaptability of vitrimer network, endowing the material with richer dynamic functionality. Simultaneously, the vitrimer matrix can exert a reciprocal effect on the nanofillers, endowing them with recyclability and reusability—a feat exceptionally challenging to achieve with traditional epoxy thermosets. As previously highlighted, these two aspects are mutually reinforcing. In contrast, the interaction in conventional permanently cross-linked epoxies is largely unidirectional, with the influence primarily flowing from the nanofillers to the matrix.

Given the diverse structures and properties of nanofillers, there is currently no one-size-fits-all doping method at the industrial scale. Instead, specific approaches must be tailored to the characteristics of each filler. Nevertheless, many preparation methods share common underlying principles or procedures, differing mainly in their practical implementation details.

A direct and widely applicable method for introducing nanofillers into vitrimer networks is physical blending [35, 89, 153, 154]. This approach is typically carried out in the monomer melt or prepolymer prior to the curing of the vitrimer network. Nanofillers are uniformly dispersed within the system via mechanical stirring, ultrasonication, or solvent-assisted dilution. Subsequently, the vitrimer network undergoes *in-situ* polymerization and crosslinking—catalyzed when necessary (Fig. 7(a)). Achieving homogeneous dispersion of nanofillers remains a key challenge in physical blending due to strong interfacial interactions at the nanoscale. During this process, nanofillers are primarily immobilized through physical entrapment within the polymer network, generally without forming strong chemical bonds with the matrix.

Our group has previously employed physical blending to

incorporate various nanomaterials—such as CNTs [41, 89], magnetite [155, 156], and silica [154]—into different dynamic vitrimer networks. Although these studies utilized matrices other than epoxy vitrimers (including polyurethane-based networks), they demonstrate the general applicability of physical blending for integrating diverse nanoparticles into vitrimer matrices. This strategy is particularly suitable for nanoparticles that do not chemically react with the network. For epoxy vitrimers, both carbon-based materials and other inorganic nanoparticles inert to the monomers can be incorporated via pre-polymerization blending, yielding nanocomposites with a wide range of functionalities.

Notably, many nanofillers tend to aggregate or restack due to their strong intrinsic interactions. For example, the pronounced π–π interactions between graphene layers make it difficult to achieve homogeneous dispersion in the monomer melt or prepolymer mixture during physical blending with the vitrimer matrix. This often leads to restacking and aggregation during subsequent curing, resulting in a non-uniform distribution of filler aggregates and consequently inhomogeneous nanocomposites [157–160]. Furthermore, it is challenging to maintain stirring or ultrasonication throughout the curing process, as prolonged agitation can introduce numerous air bubbles. Once agitation stops, nanoparticle fillers tend to reagglomerate, adversely affecting the final material properties. To mitigate these issues, many current methods utilize compression molding to cure the mixture, which promotes more thorough filler dispersion. However, this approach limits large-scale production at engineering levels and significantly restricts the attainable geometries of the resulting composites.

Many carbon nanomaterials and inorganic nanospheres possess surfaces that are chemically reactive or readily functionalizable [80, 82, 161]. Chemical modification of these nanofillers can suppress their agglomeration by enhancing their compatibility and interactions with the polymer network. Moreover, suitably functionalized surfaces may participate directly in network formation during curing, becoming covalently integrated into the polymer architecture. For example, graphene-based nanomaterials are typically chemically treated and functionalized prior to their incorporation into nanocomposites. Specific surface-modification strategies and advanced structural control will be discussed in subsequent sections. The chemical modulation approaches developed for graphene offer a valuable framework for the surface engineering of other nanoparticle fillers, based on their inherent surface chemistry and structure.

Beyond conventional physical blending and chemically modified mixing, nanofiller layers can also be deposited onto the surface of already-formed vitrimer polymers through specific techniques to create multilayer composite architectures. Our group previously employed an aqueous immersion method to form a polydopamine (PDA) nanocoating on flexible polymer films [162]. During immersion in a stirred dopamine solution, dopamine undergoes oxidative self-polymerization, forming a uniform and strongly adherent PDA nanocoating on the film surface. This design strategy offers a transferable approach for depositing surface nanocoatings on epoxy vitrimer materials. Such solvent-assisted nanoparticle deposition onto molded polymer surfaces follows essentially similar principles. The resulting multilayer structure mitigates the risk of inhomogeneity associated with pre-cure physical mixing. For film-based materials, the functional distinction between a surface-adhered layer and a uniformly dispersed composite is often minimal, with no significant differences observed in macroscopic performance—including stimulus responsiveness and thermal transport characteristics.

Beyond the methods discussed, other innovative strategies exist for incorporating nanofillers into vitrimer crosslinked networks. Chemical modification significantly expands the scope for tailoring filler surface properties and architectures. Furthermore, the development of novel design and preparation routes for nanofillers promises access to diverse morphologies and highly functional nanocomposites.

4 Carbon nanomaterial/epoxy vitrimer composites

Incorporating carbon nanomaterials into thermosetting polymer matrices is a well-established strategy for enhancing material performance and imparting electrical or thermal conductivity, with extensive research and mature industrial applications [35, 78, 158, 160, 163–168]. For stimulus-responsive vitrimers, the high electrical conductivity and efficient photothermal conversion typical of many carbon nanomaterials not only broaden the functional scope of the vitrimer but also diversify the modes of stimulus response available for regulating dynamic bond exchange. Traditional vitrimers are often constrained by their limited stimulus modalities, making the integration of functional nanofillers an attractive solution. For example, in many thermally sensitive electronic devices, direct heating of the material may be impractical, and uniformly heating large or irregularly shaped components can be challenging—factors that otherwise restrict the application of vitrimers. In contrast, light stimulation offers precise, remote, and localized control, enabling actuation in scenarios where conventional heating is infeasible. Moreover, the versatile chemical and spatial structures of carbon nanomaterials allow for the grafting of functional molecules via dynamic covalent bonds, facilitating their on-demand, reversible release within the stimulus-responsive framework of the vitrimer network.

Materials such as CNTs and graphene exhibit exceptional photothermal conversion efficiency, functioning as nanoscale heat sources that efficiently absorb photons from the visible to near-infrared range and convert them into substantial thermal energy. When uniformly dispersed as fillers within a vitrimer matrix, these carbon nanomaterials can instantaneously transform incident light into localized heat. This rapid heating triggers dynamic covalent bond exchange within the vitrimer network, enabling remote and fast network rearrangement, reshaping, and photo-welding/repair of the polymer material through light irradiation. Our group pioneered the use of this photothermal conversion effect to activate and manipulate chemical reactions in polymer networks, demonstrating light-driven self-healing and welding in epoxy vitrimers [41]. This approach extends the functionality of vitrimers beyond conventional single-stimulus modes. The CNT-vitrimer composite was prepared by physical blending, in which nanofillers were thoroughly mixed with the vitrimer monomers prior to curing under compression molding into thin films. Under infrared irradiation, the composite film achieves localized heating, enabling rapid welding and efficient self-healing of surface scratches.

Photo-induced dynamic bond exchange not only enables material self-healing and welding but also holds broader application potential. Our group previously utilized the photothermal effect of vitrimer nanocomposites to actuate liquid crystal elastomers

(LCEs). In CNT-functionalized exchangeable LCE composites (CNT-*x*LCEs), reversible bending, stretching, and rotational motions can be achieved by modulating light intensity and irradiation position, paving the way for applications in soft robotics and actuators (Fig. 13) [89]. Notably, CNT reinforcement concurrently enhances the mechanical properties of the composite, allowing the structure to bear loads substantially exceeding its own weight and perform practical work. Furthermore, the CNT photothermal effect can trigger bond exchange in vitrimers even at cryogenic temperatures, significantly extending the applicability of such three-dimensional smart actuators. This enables stable functionality in extreme environments, such as polar regions or outer space. The successful integration with LCE vitrimer networks demonstrates that carbon nanomaterial doping exhibits universality across diverse polymer-based dynamic networks, endowing materials with complex three-dimensional motility and versatile application scenarios.

Beyond their photothermal properties, CNTs possess inherently high electrical conductivity. When dispersed within typically insulating epoxy vitrimer matrices—particularly those cured via epoxy–dicarboxylic acid systems—they act as countless microscopic conductive pathways. Lorero et al. [121] observed that when the CNT content in epoxy vitrimers exceeds 0.1 wt.%, the nanotubes contact and overlap, forming a percolated three-dimensional conductive network throughout the material. Leveraging the Joule heating effect enables efficient electrical heating, which in turn can trigger and control dynamic covalent bond (DCB) exchange within the nanocomposite. This electrothermal capability, analogous to the photothermal effect, opens avenues for CNT-reinforced vitrimer nanocomposites in applications such as conductive heating, electrical welding, and other functions within sustainable reconfigurable structures and flexible electronics.

Graphene, a two-dimensional material consisting of a single layer of sp^2-hybridized carbon atoms, exhibits outstanding mechanical, thermal, and electrical properties, sharing functional similarities with CNTs [35, 158, 161]. In vitrimer nanocomposites, graphene serves as a multifunctional nanofiller that not only enhances mechanical strength but also imparts electrical/thermal conductivity and photothermal/electrothermal responsiveness, enabling remote and precise control of dynamic covalent bond exchange reactions. In the graphene–vitrimer composites prepared by Yang et al. [35], graphene was incorporated into the vitrimer matrix via physical blending. Even at a low loading of only 1 wt.%, significant performance improvements were achieved: the yield strength and fracture strain increased substantially from 12.0 MPa and 6% (neat vitrimer) to 22.9 MPa and 44%, respectively. The addition of

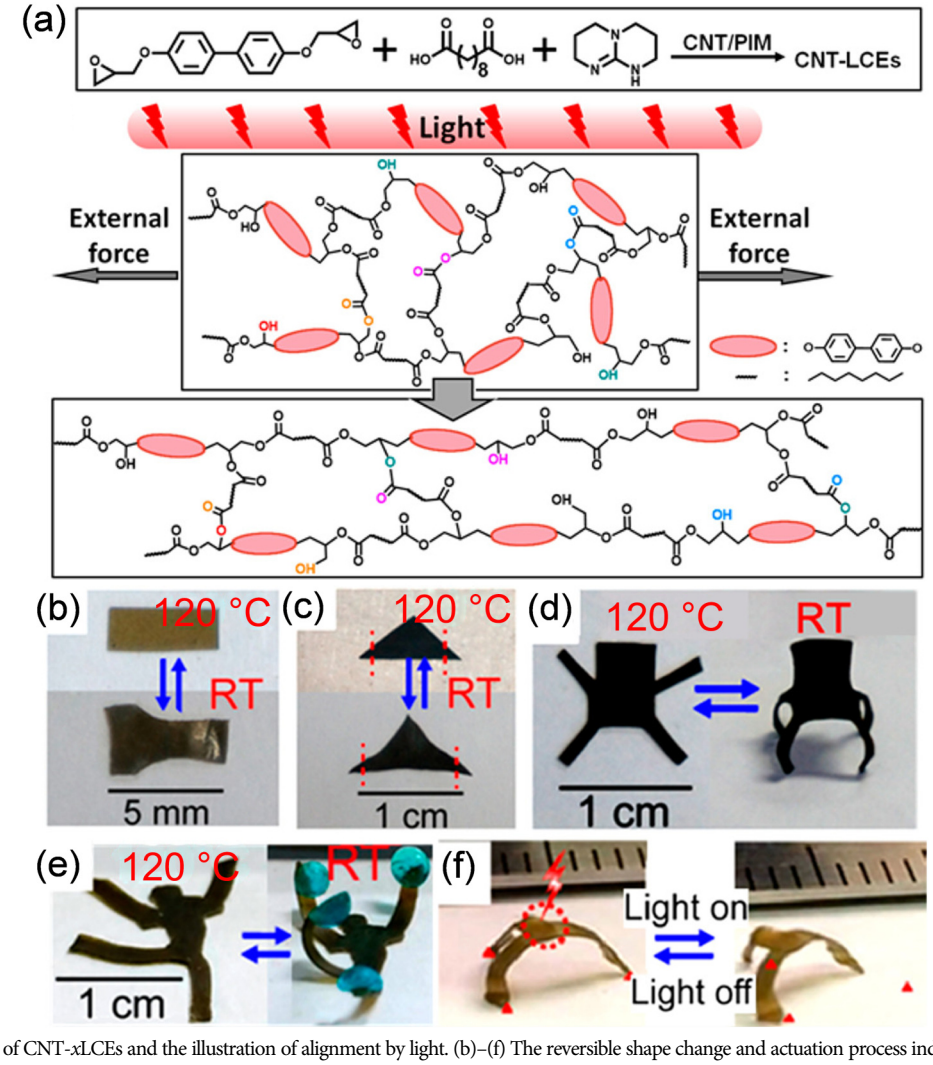

Figure 13 (a) Synthesis of CNT-*x*LCEs and the illustration of alignment by light. (b)–(f) The reversible shape change and actuation process induced by light. Reproduced with permission from Ref. [89], © American Chemical Society 2016.

graphene altered the failure mode of the material, transforming the intrinsically brittle pure vitrimer into a tough material that exhibits clear yielding and a dimpled fracture surface. This effective strengthening and toughening provide the essential mechanical foundation for achieving large-strain shape memory and reconfiguration in the material.

However, the strong π–π interactions between graphene layers pose a significant challenge for achieving uniform dispersion within a polymer matrix during composite processing [160, 169]. These interlayer forces cause graphene sheets to tend to restack and agglomerate in the uncured mixture. To address this, chemical modification strategies have been developed and widely adopted for graphene–vitrimer nanocomposites [161, 170–172]. Modified graphene exhibits reduced aggregation while enhancing interfacial compatibility with the polymer, often enabling covalent bonding with the network. Importantly, such modifications generally preserve the inherent functional properties of graphene.

Graphene oxide (GO), a two-dimensional derivative obtained from the oxidation of graphite, represents one of the most direct chemical routes to functionalized graphene [173–177]. The surface of GO is rich in oxygen-containing groups such as hydroxyls, epoxides, and carboxyls, which significantly improve its compatibility with polymer matrices. Krishnakumar et al. [161] prepared GO nanosheets by oxidizing graphite in strong acids and oxidizing agents, thereby introducing oxygen functional groups between the layers. The GO was then uniformly dispersed into an epoxy prepolymer, with aggregation notably reduced during mixing. Subsequent curing with 2-AFD as a curing agent yielded a catalyst-free epoxy vitrimer nanocomposite based on disulfide exchange (Fig. 14). Compared to the GO-free reference, the composite showed significant enhancements in storage modulus, flexural strength, and flexural modulus. Moreover, owing to the high photothermal conversion efficiency of GO, the nanocomposite also exhibited low-temperature shape-memory properties.

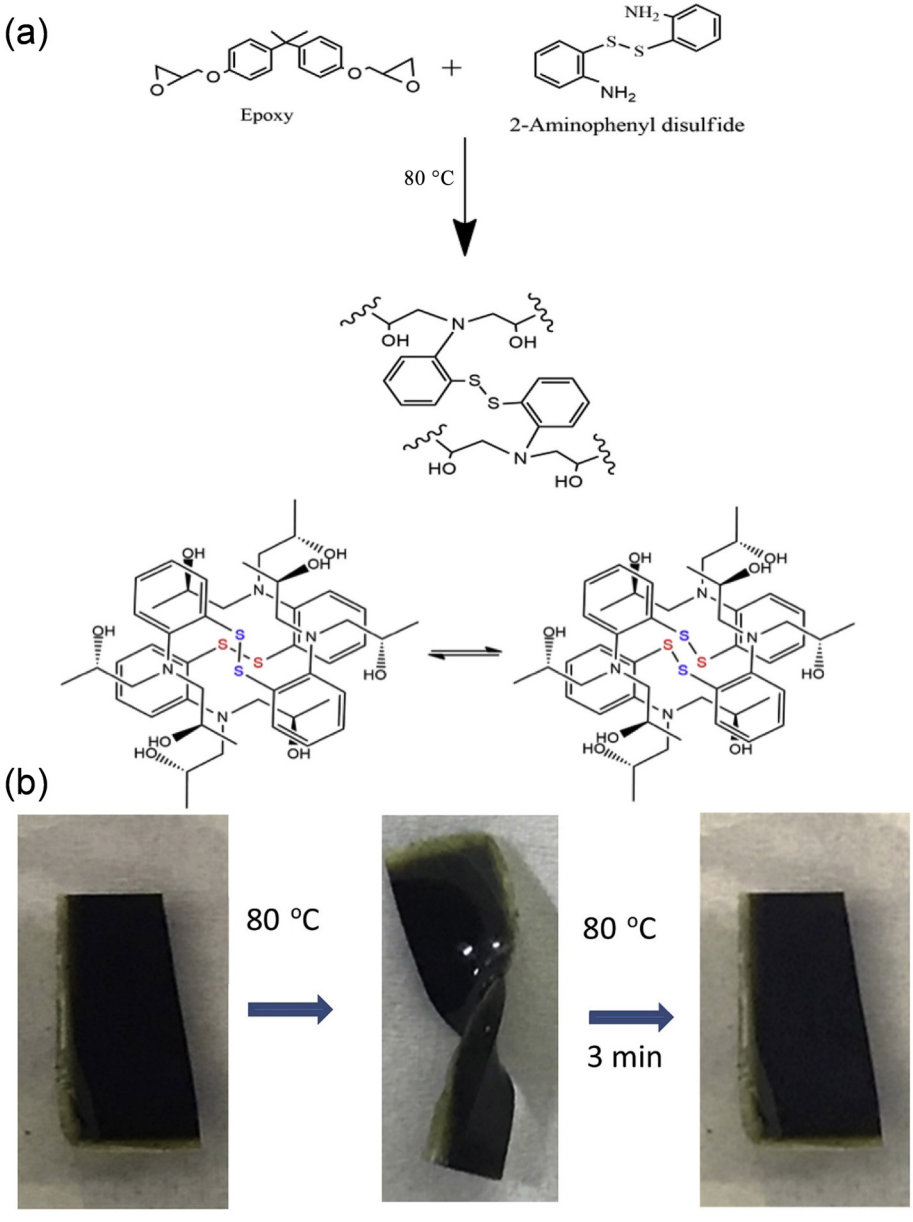

Figure 14 (a) Synthesis and chemical structure of epoxy vitrimer and (b) the shape recovery of GO-vitrimer composites. Reproduced with permission from Ref. [161], © Elsevier Ltd. 2020.

The addition of nanofillers not only imparts new properties to vitrimers but also can enhance the dynamic exchange within the network itself. Park et al. [178] employed molecular dynamics simulations to elucidate the mechanism by which GO promotes dynamic disulfide exchange. Their findings indicate that incorporating GO lowers the T_g of the dynamic network and creates a van der Waals excluded volume at the filler-matrix interface. This interfacial region enhances the mobility of nearby molecular segments, thereby accelerating the dynamic disulfide exchange reaction (Fig. 15). Simulations further demonstrated that, under identical self-healing conditions, the number of newly formed disulfide bonds in the GO/vitrimer system significantly exceeds that in the neat vitrimer, with the most active exchange occurring within the interfacial zone. This reveals that GO not only improves macroscopic self-healing performance but also provides atomistic-level insights into modulating the reaction kinetics and dynamic properties of vitrimer networks through nanofillers. Moreover, the oxygen-rich surface of GO allows for further chemical functionalization to strengthen its interaction with the polymer network, enabling it to participate directly in network formation or bond exchange. Bohra et al. covalently grafted organic molecular chains onto the GO surface using 4-AFD functionalization. This increased steric hindrance between graphene sheets not only improved dispersion but also allowed the functionalized graphene oxide (FGO) to participate in the epoxy vitrimer curing reaction (Fig. 16) [120]. Once the network was formed, the disulfide bonds on the FGO could engage directly in the dynamic exchange of the vitrimer matrix. This design strategy transforms FGO from a mere filler into an integral, covalently bound component of the network, thereby stabilizing the filler-matrix interaction.

Notably, even unmodified graphene nanomaterials can enhance composite properties through alternative strategies, such as hybrid integration with other nanofillers to introduce multiple functionalities simultaneously. Ren et al. first prepared gold nanoparticle–graphene hybrid fillers (AuNPs/GNPIs) by *in-situ* reduction of gold acetate on graphene nanosheets (Fig. 17) [179]. In this process, defects on the graphene surface served as nucleation sites, followed by growth of gold nanoparticles via Ostwald ripening, resulting in a patchy, anchored coating rather than full coverage. The hybrid filler was then uniformly dispersed into a vitrimer matrix through shear mixing and ultrasonication to form nanocomposite films. This unique hybrid architecture ingeniously combines the localized surface plasmon resonance (LSPR) of gold nanoparticles with graphene's broad-spectrum light absorption and high thermal conductivity, producing a pronounced synergistic

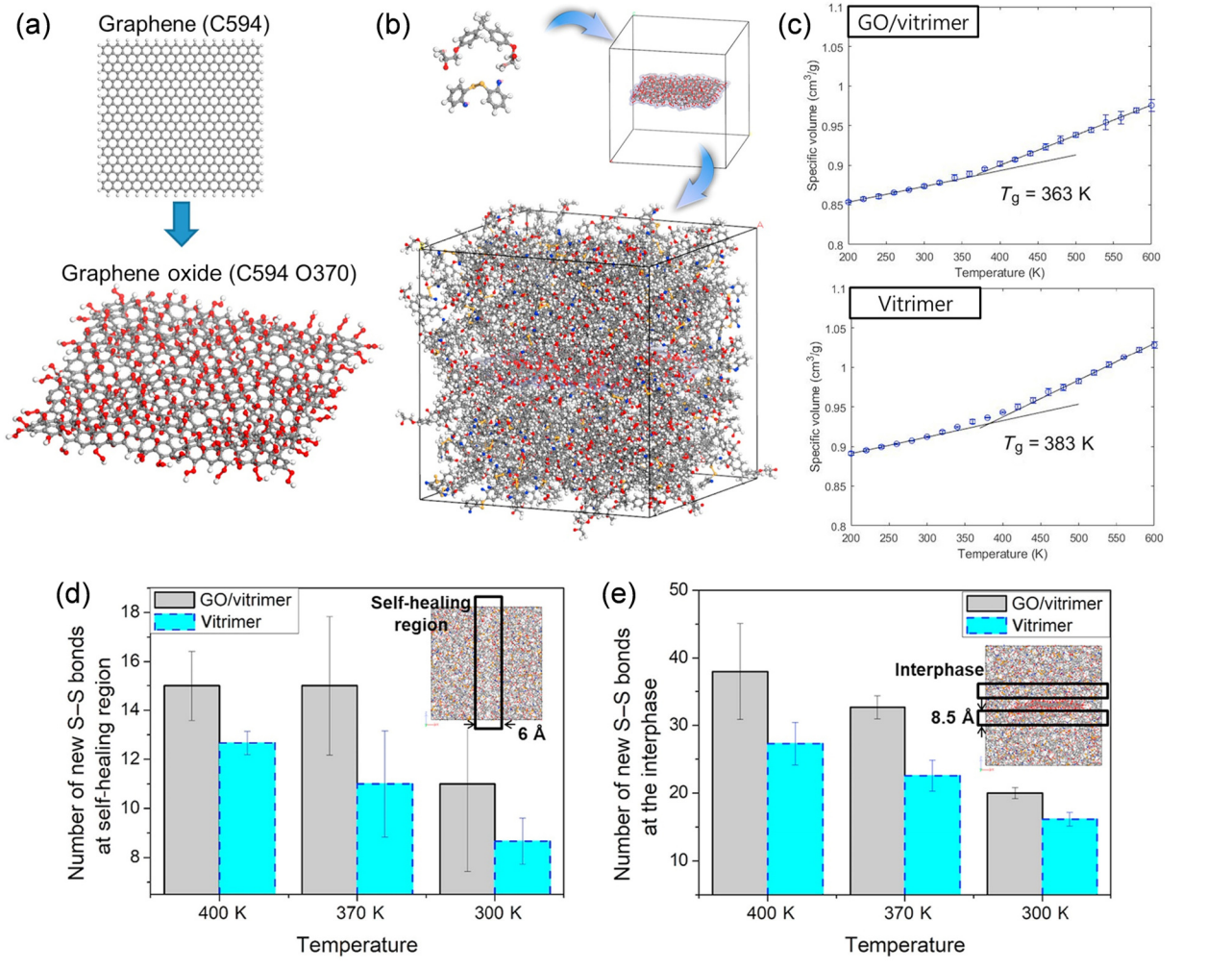

Figure 15 (a) Initial structure of GO/vitrimer. (b) A unit cell containing the GO is packed with the DGEBA and AFD molecules followed by the dynamic crosslink simulations to form a crosslinked structure having the same crosslink density with vitrimer models. (c) Specific volume vs temperature graph of GO/vitrimer and vitrimer. Calculated T_g of GO/vitrimer and vitrimer. Reproduced with permission from Ref. [178], © Elsevier Ltd. 2020.

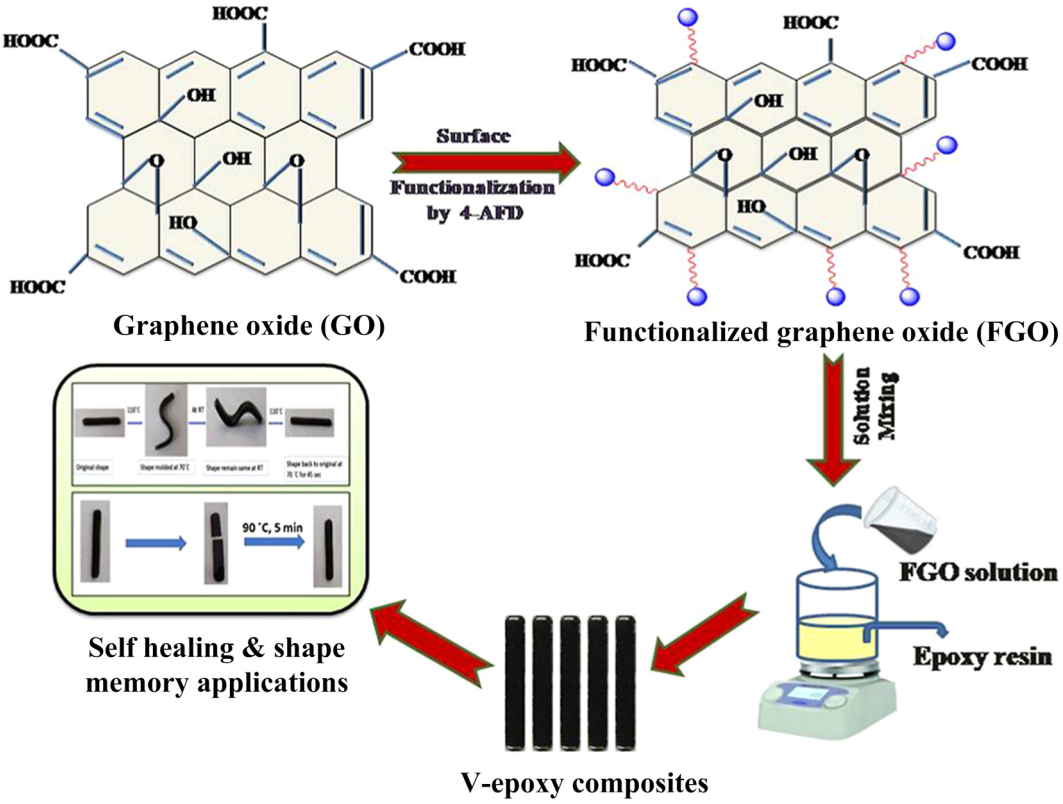

Figure 16 Schematic procedures of the functionalization of GO into FGO, the preparation of composites and applications of them. Reproduced with permission from Ref. [120], © Elsevier Ltd. 2023.

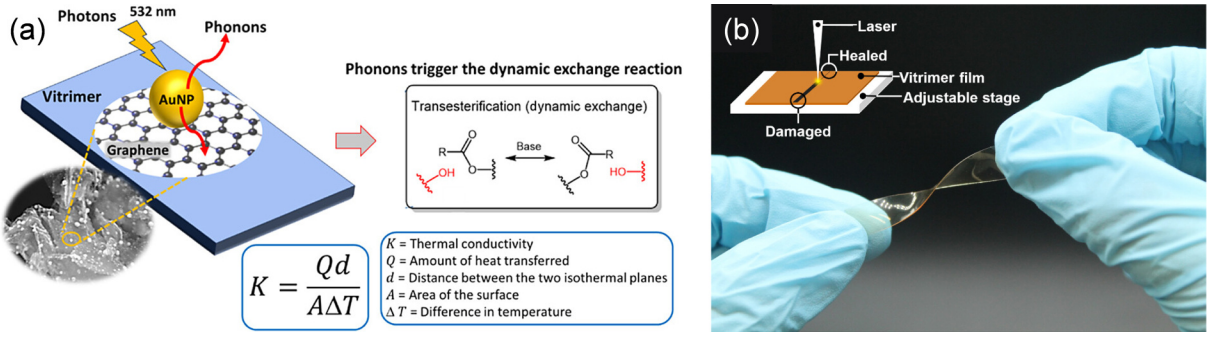

Figure 17 (a) Schematic presentation of photo-thermal conversion and structure of nanocomposite. (b) The transparency and flexibility of vitrimer film and the damage healing process with laser irradiation. Reproduced with permission from Ref. [179], © American Chemical Society 2024.

effect. The resulting composite achieves ultrafast, localized photothermal self-healing within milliseconds (as short as 100 ms) under 532 nm laser irradiation, with a healing efficiency reaching 90.9%—far exceeding that of single-filler systems. Moreover, while maintaining film flexibility and partial transparency, the material offers a novel platform for on-demand, precise coating repair.

In addition to incorporating hybrid fillers, controlling the higher-order self-assembled architectures of graphene sheets themselves offers an alternative route to achieve distinctive enhancement effects. For instance, graphene aerogel (GA) is a three-dimensional porous network material formed by the self-assembly of graphene sheets [180–182]. Its interconnected sheets constitute an ultralight solid with a hierarchical porous structure, exhibiting high electrical conductivity, large specific surface area, and excellent mechanical flexibility. Luo et al. [183] incorporated annealed graphene aerogel (aGA) as a nanofiller into a polyimide vitrimer matrix, and its synergistic reinforcing effect far surpassed that of conventional fillers such as graphene powder and carbon nanotubes. At a GA loading of 5 wt.%, the composite achieved a high electrical conductivity of 161 S/m—eight times that of composites with graphene powder and over thirty times that of carbon nanotube composites. Even after nine consecutive reshaping cycles, the conductivity showed no significant degradation. Under 10% tensile strain, the conductivity decreased by only 7%, demonstrating exceptional stability. Similarly, introducing graphene-based materials with diverse morphologies into epoxy vitrimers holds promise for developing highly efficient, reinforced vitrimer nanocomposites.

Carbon nanodots (CDs) have emerged in recent years as another promising nanocarbon material and are being increasingly employed in the construction of nanocomposites with broad potential. CDs possess a conjugated structure typically below 10 nanometers in size. They exhibit not only strong fluorescence, chemical stability, ease of functionalization, and excellent

biocompatibility, but also a surface chemistry that can be deliberately tailored. Tang et al. [82] functionalized the surface of carbon nanodots to enrich them with carboxyl groups, which subsequently reacted with epoxy groups in an epoxy-modified natural rubber (ENR) matrix to form β-hydroxy ester bonds (Fig. 18). In this design, the CDs act as crosslinking nodes within the network, endowing the final material with outstanding mechanical properties and a distinctive toughening mechanism. Under stress, the relatively short rubber chains connecting adjacent CDs undergo pronounced stretching and preferential cleavage, thereby impeding crack propagation. Meanwhile, longer molecular chains retain their elasticity, preserving structural integrity and establishing an effective energy-dissipation pathway. Furthermore, the nanocomposite exhibits intrinsic fluorescence derived from the CDs. The uniformity of the luminescence confirms the excellent dispersion of the CDs within the rubber matrix, and this built-in fluorescent labeling capability holds promising value for applications in sensing, anti-counterfeiting, and traceability.

In summary, vitrimer-carbon nanocomposites harness the unique functionalities of nanofillers, such as exceptional photothermal conversion efficiency. This enables remote and spatially controlled optical manipulation of topological rearrangement within the dynamic vitrimer network, effectively overcoming the limitations of conventional thermal stimulation. These composites exhibit outstanding low-temperature self-healing, welding, and shape-reconfiguring capabilities, along with significant mechanical reinforcement. Graphene-based fillers provide a rich platform for designing photoresponsive vitrimer nanocomposites and offer valuable insights for the functionalization of other nanofillers, many of which possess readily modifiable surface chemistry. Beyond surface modification, multicomponent hybrid fillers can be engineered to yield stronger synergistic effects than single-filler systems. Furthermore, multilevel structural assembly of carbon materials represents another effective strategy for performance enhancement, with numerous possibilities yet to be explored. Future research on carbon-nanomaterial-based vitrimer smart composites may focus on further improving photothermal conversion efficiency, optimizing nanofiller dispersion and matrix-filler interfacial interactions, and developing novel photoresponsive dynamic bond systems alongside multifunctional integration strategies. These advances will help expand their application potential in demanding environments and next-generation smart devices.

5 Non-carbon nanomaterials/epoxy vitrimer composites

5.1 Magnetic nanoparticles/epoxy vitrimer composites

The incorporation of magnetic nanoparticles into vitrimer matrices using specific methods enables the fabrication of magnetically responsive and controllable nanocomposites. These composites synergistically combine the advantages of magnetic stimulation—such as remote, non-contact control, rapid response, high biocompatibility, and strong penetration—with the inherent reprocessability, self-healing, and shape-reconfiguring capabilities of vitrimers, thereby expanding stimulus modes and application scenarios. The magnetically responsive vitrimer-based nanocomposites can serve unique functions in fields such as magnetic monitoring and magnetic sensing.

Magnetically responsive nanocomposites typically operate through two principal mechanisms [184–189]. The first is the magnetothermal effect, in which alternating magnetic fields induce heating within magnetic nanoparticles, thereby triggering reversible bond exchange reactions that enable material healing, welding, recovery, shape memory, and deformation. The second is direct magnetic guidance, where forces or torques exerted by static magnetic fields on magnetic particles induce immediate macroscopic motion and complex shape changes in the material. In contrast to vitrimer composites doped with carbon nanomaterials—where photothermal stimulation is typically confined to the surface and lacks penetration—magnetic fields offer distinct advantages. Light-activated systems often exhibit non-uniform responsiveness, with bond exchange reactions progressing more slowly in the bulk than at the irradiated surface. For thicker materials, it is difficult to activate internal transesterification via surface illumination, resulting in pronounced response lag. Under a uniform magnetic field, however, all dispersed magnetic nanoparticles are activated simultaneously, enabling a homogeneous, coordinated response throughout the entire material volume. These characteristics endow magnetically responsive vitrimer nanocomposites with significant application potential in

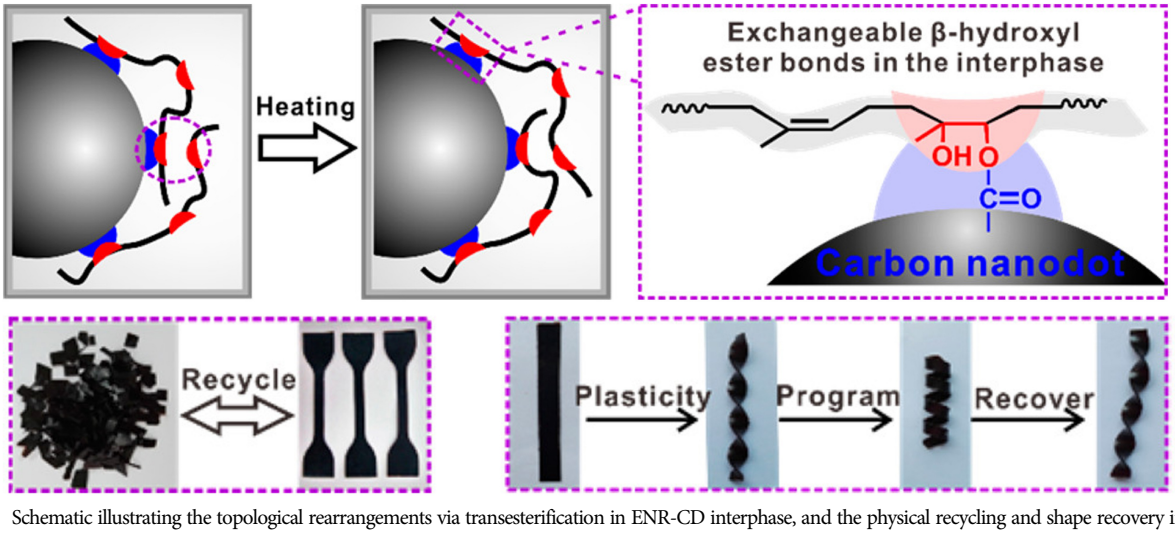

Figure 18 Schematic illustrating the topological rearrangements via transesterification in ENR-CD interphase, and the physical recycling and shape recovery induced by heat. Reproduced with permission from Ref. [82], © American Chemical Society 2017.

advanced fields such as soft robotics, biomedical devices, and flexible electronics.

Chen et al. [47] employed a physical blending method, in which all reactants—including magnetite nanoparticles, the difunctional epoxy resin DGEBA, the dithiolane derivative DTDA (containing dynamic disulfide bonds), and the organic catalyst TBD—were mixed prior to curing. This procedure yielded an epoxy vitrimer network capable of dual dynamic bond exchange, namely transesterification and disulfide metathesis, while simultaneously introducing magnetic responsiveness to create a multifunctional nanocomposite (Fig. 19). The resulting material not only allows flexible modulation of dynamic properties such as characteristic relaxation time and T_v, but also exhibits multi-stimuli responsiveness, enabling magnetically driven actuation, near-infrared photothermal conversion, and thermal/optical healing and welding. This combination highlights its considerable potential for reconfigurable, self-healing smart materials. As noted earlier, physical blending serves as a versatile preparation route for nanocomposites and can be readily extended to other epoxy-based or vitrimer networks for the incorporation of magnetic nanoparticles. Our group has successfully introduced Fe_3O_4 nanoparticles into a polyurethane-based vitrimer network using the same physical blending strategy [155]. By integrating nanocomposite regions with different nanoparticle loadings into a single device, selective activation of individual areas can be achieved by tuning the magnetic field strength, creating localized, sequential actuation akin to "logic switches". This approach offers a design principle for constructing locally responsive, multi-level controlled devices that can adapt to dynamic environments, and provides a conceptual framework for developing integrated, multifunctional, and multi-responsive systems based on epoxy vitrimer nanocomposites.

It is noteworthy that when iron oxide particles are reduced to a specific size range (typically 3–15 nm), they exhibit superparamagnetic behavior at room temperature and are referred to as superparamagnetic iron oxide nanoparticles (SPIONs) [190–192]. This phenomenon arises primarily from size effects. Conventional iron oxide nanoparticles are usually larger (> 30 nm) and contain multiple magnetic domains—small regions within the particle where magnetic moments are aligned. These domains often cancel each other out, leading to a weak overall magnetization. Under an external magnetic field, domain walls shift, eventually aligning most magnetic moments along the field direction. When the field is removed, a high energy barrier prevents the moments from easily returning to their original state, leaving behind residual magnetization (remanence). In contrast, SPIONs are so small that each particle effectively behaves as a single magnetic domain; all atomic magnetic moments within a particle align in the same direction. This single domain acts like a tiny magnet with a strong net magnetic moment. Under an external magnetic field, SPIONs rapidly magnetize and display strong ferromagnetic-like response. Once the field is removed, thermal energy randomizes the magnetic moments, causing the macroscopic magnetism to vanish almost instantly with no remanence—a property termed superparamagnetism. Precisely because SPIONs leave no residual magnetism, they can be stably and uniformly dispersed within a matrix. This avoids mutual attraction or aggregation, which would otherwise severely compromise material homogeneity and

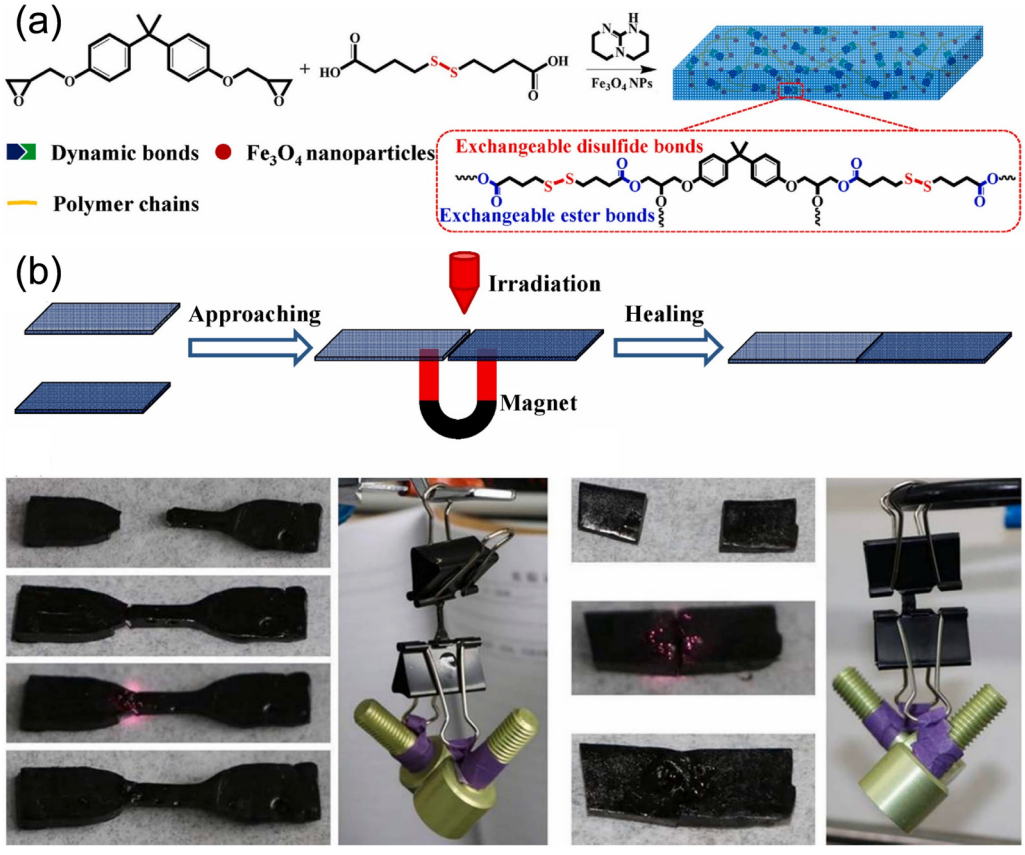

Figure 19 (a) The preparation of vitrimer-based nanocomposites. (b) Fracure-welding of the nanocomposites by the combination of near infrared and magnet. Reproduced with permission from Ref. [47], © Elsevier Ltd. 2022.

mechanical properties. Owing to this unique combination of strong magnetic response and zero remanence, SPIONs currently show great promise in fields such as biomedicine [193–195] (e.g., MRI contrast agents, targeted drug delivery, magnetic hyperthermia), catalysis [196, 197], and environmental remediation [198, 199].

Signorato et al. [200] successfully covalently integrated superparamagnetic iron oxide nanoparticles (SPIONs) into a vinyl-urea vitrimer matrix by designing a novel ligand (PAAcAc) containing a phosphate-anchoring group and an acetoacetic ester terminal group, thereby constructing a magnetically responsive composite vitrimer (Fig. 20). This design not only ensures uniform and stable dispersion of SPIONs within the matrix—avoiding the particle agglomeration often encountered in traditional physical blending—but also significantly enhances the mechanical properties of the material, increasing its fracture stress by up to 244%. Moreover, the composite fully retains the reshaping, self-healing, and recyclable characteristics of the vitrimer, ensuring sustainability throughout the material's life cycle. Crucially, the incorporated SPIONs endow the composite with rapid responsiveness to external magnetic fields, enabling its use as a magnetically driven actuator for soft robotics with precise controllability. Their superparamagnetic nature guarantees complete demagnetization once the magnetic field is removed. While SPION-epoxy vitrimer composites have not yet been reported, this work provides an exemplary model for the covalent integration of SPIONs into epoxy-based vitrimer systems.

In addition to the prevalent use of magnetite (Fe_3O_4) as the magnetic component, other types of magnetic nanoparticles also show promise for creating magnetically responsive composites when incorporated into polymer matrices. For instance, Kuang et al. [201] constructed a magnetically responsive dynamic polymer (MDP) by embedding ferromagnetic NdFeB microparticles into a reversible crosslinked dynamic polymer (DP) based on the Diels–Alder reaction. Incorporating such magnetic particles into an epoxy vitrimer matrix is similarly feasible in principle. The resulting magnetic vitrimer composite allows bond cleavage and reformation upon heating, enabling three key functions: seamless magnetic-guided welding between components, *in-situ* reprogramming of magnetization direction, and magnetically driven permanent structural reconfiguration. The MDP composite not only permits the programmable fabrication of complex geometries and magnetization distributions, but also extends its applicability to reconfigurable deformable structures, multistable origami/kirigami architectures, and remotely controlled soft robotics.

In addition to these examples, other magnetic nanomaterials—such as manganese ferrite ($MnFe_2O_4$) [202, 203], carbonyl iron [204, 205], nickel rods [206, 207], and barium ferrite [208, 209]—also exhibit distinct application potential. Although magnetically responsive nanocomposites have not yet been directly synthesized within epoxy vitrimer networks, ongoing exploration and development of magnetic inorganic nanoparticles, metallic particles,

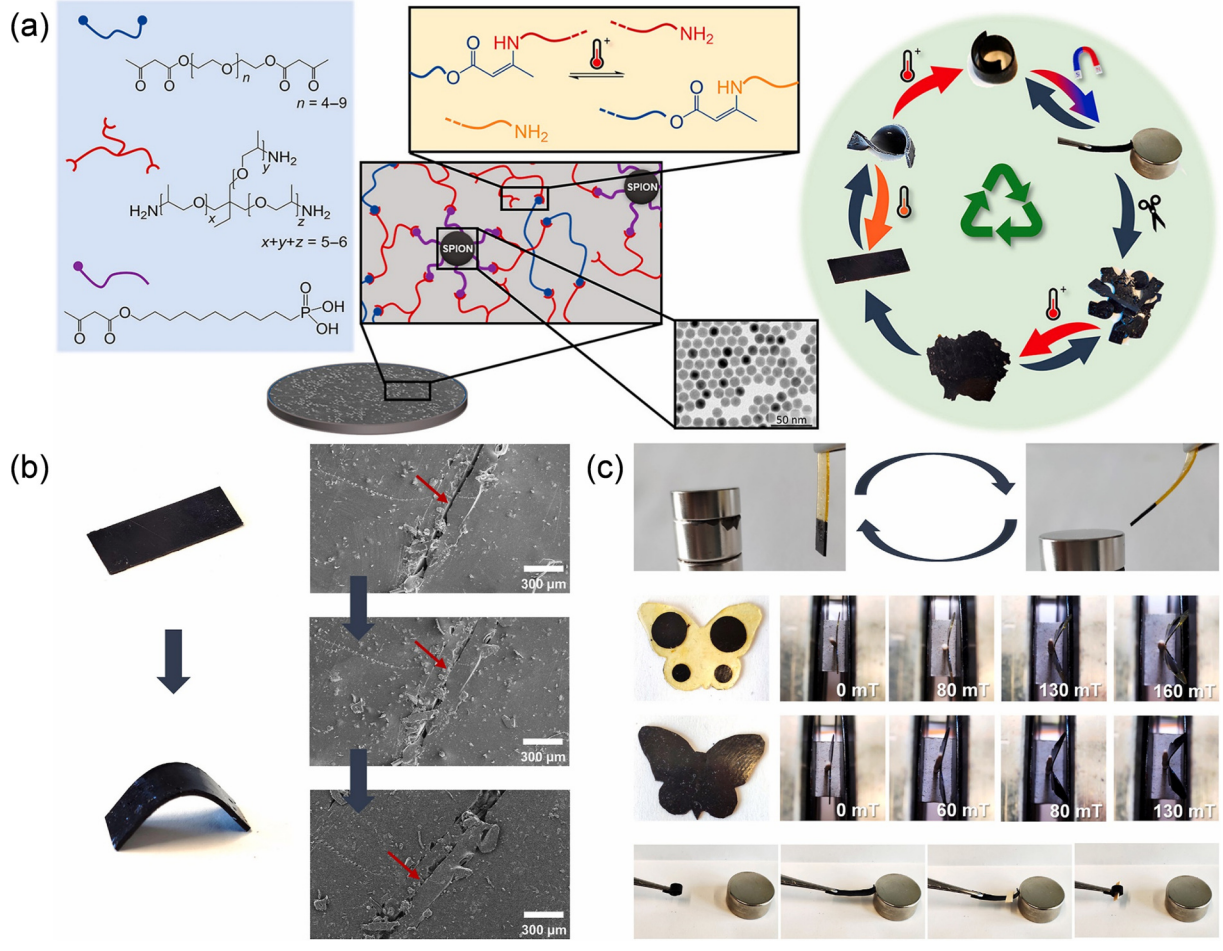

Figure 20 (a) Schematic structure and applications of vitrimer-SPION nanocomposites. (b) The reshaping and self-healing process of the composites. (c) Magnetic control of nanocomposite and its actuator application. Reproduced with permission from Ref. [200], © Elsevier Ltd. 2025.

and alloy particles will provide a rich library of fillers for designing such systems and expand their potential application scope.

5.2 Silica nanoparticles/epoxy vitrimer composites

Beyond incorporating intrinsically functional nanoparticles into vitrimer matrices to extend stimulus-responsive self-healing capabilities, certain nanoparticles—which lack specific functionality—are frequently employed as nano-reinforcing agents. These primarily enhance the mechanical and thermal properties of the polymer matrix, with silica nanoparticles being the most representative example. Widely used as a nano-reinforcing phase in composites, inorganic silica nanoparticles offer advantages such as low cost, straightforward synthesis, controllable size, and ease of surface functionalization. Nanocomposites formed with vitrimers find extensive application in adhesives, protective coatings, paints, and related fields, and hold broader prospects for industries such as civil engineering.

Similarly, unmodified silica nanoparticles are commonly incorporated into vitrimer crosslinked networks through physical blending and are widely used in the preparation of epoxy composites [210–214]. Our group successfully introduced silica nanoparticles into a liquid crystal epoxy-based dynamic network to produce reinforced liquid crystal elastomers (*x*LCN/SNP) [154]. Compared with the neat *x*LCN, the composite exhibits a lower liquid crystal–isotropic transition temperature (T_i) and a higher thermal decomposition temperature (T_d). The addition of 1 wt.% silica nanoparticles increases T_d by approximately 30 °C, significantly improving thermal stability. In terms of mechanical properties, the composite shows enhanced tensile strength and increased elongation at break at elevated temperatures, both above T_g and even above T_i. These improvements are substantial and directly enhance the performance of *x*LCN materials during high-temperature molding and reshaping processes, thereby providing a more reliable material foundation for smart devices that require complex shape processing and excellent thermal stability.

The doping content of nano-silica in vitrimer nanocomposites can be adjusted over a broad range. Furthermore, the inorganic interface can be chemically modified to improve compatibility with the organic matrix, underscoring the material's practical versatility. Barabanova et al. [153] prepared uniformly dispersed silica-reinforced epoxy vitrimer networks via physical blending. Compared to the neat vitrimer matrix, the addition of 5 wt.%–10 wt.% silica increased the elastic modulus by 44% (from 1.8 to 2.6 GPa) and improved the tensile strength by 20%–25% (from 54 to 65 MPa). Thermal properties were also enhanced, reflected in a reduced coefficient of thermal expansion and an elevated T_g (Fig. 21). Improved weldability was demonstrated by the fact that welded specimens containing SiO_2 fractured outside the joint interface, indicating that the weld strength exceeded that of the pure vitrimer matrix. This reinforcement mechanism likely stems from hydrogen bonding interactions provided by silica and its participation in interfacial transesterification reactions. Increasing the loading of silica nanofillers further can also yield nanocomposites with excellent properties and practical value. However, excessively high filler contents tend to accentuate incompatibility with the organic matrix, often resulting in heterogeneous composite performance. In the study by Legrand et al. [80], through rational interface design—specifically, surface functionalization—nanocomposites with both outstanding mechanical properties and vitrimer-like dynamic behavior were

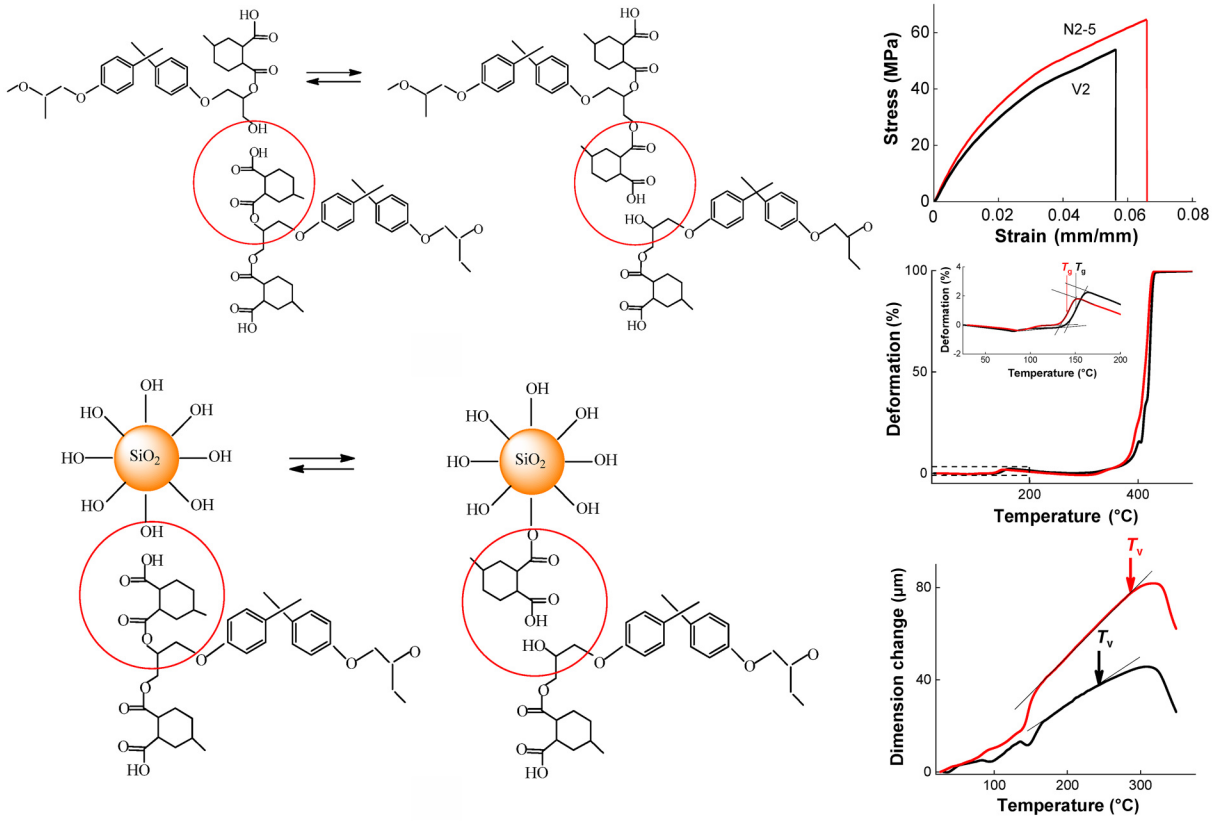

Figure 21 Schematic representation of transesterification reaction between epoxy chains and on the epoxy/silica interface, and the influence of introducing nano-silica into vitrimer matrix toward the mechanical and thermal properties. Reproduced with permission from Ref. [153], © MDPI 2021.

successfully prepared even at inorganic filler loadings as high as 40 wt.%. The silica nanoparticles were surface-modified with epoxy groups. During the formation of the vitrimer network, these epoxy groups participated in the curing reaction, becoming covalently integrated into the crosslinked structure and enabling reversible transesterification, thereby imparting reshapeability to the material. Moreover, at such high nanoparticle loadings, both modulus and strength were significantly enhanced, preserving the composite's practical utility. The abundant silanol groups on the silica surface offer rich possibilities for chemical modification, making surface functionalization a versatile strategy to address dispersion and compatibility challenges of inorganic fillers in organic matrices. By chemically bridging the interface between inorganic filler and organic vitrimer, the composite benefits from the stability of the inorganic phase and the reversibility of the dynamic covalent network, leading to comprehensive improvements in material performance and structural uniformity.

Beyond conventional approaches such as physical blending and chemically modified crosslinking, novel doping methods have been developed that offer broader perspectives for designing innovative nanocomposite fabrication processes. Kimura et al. [215] reported a one-pot strategy using a commercial polyester to successfully prepare silica-nanoparticle-reinforced vitrimer composites (Fig. 22). The key advantage of this method lies in its remarkable simplicity: neither the polyester nor the silica filler requires complex pre-modification. Crosslinking of the polymer network and incorporation of the filler are achieved simultaneously through simple blending followed by thermal curing. The addition of silica nanoparticles not only significantly enhances the mechanical properties of the vitrimer composite—including storage modulus, Young's modulus, and ultimate stress—but also induces distinct changes in its dynamic exchange behavior. Most notably, the "constrained rubber phase" formed at the filler-matrix interface not only provides reinforcement but also broadens the distribution of stress-relaxation times in the material. This indicates that, while achieving macroscopic reinforcement and toughening, the composite exhibits more complex and tunable relaxation kinetics on the microscopic scale compared to the neat vitrimer matrix.

As a classic inorganic filler for enhancing mechanical and thermal properties, silica is extensively employed in polymer-based nanocomposites. While low-loadings of nano-silica serve as an effective strategy for property improvement, high-loadings achieved through chemical modification and tailored synthesis routes offer considerable potential for exploration, promising the development of numerous novel materials. Furthermore, inspired by hybridization strategies used for graphene nanofillers, the combined use of silica with other types of nanofillers for synergistic reinforcement represents another highly promising research direction.

5.3 other inorganic nanomaterials/epoxy vitrimer composites

The extensive family of inorganic nanomaterials—including various oxides, salts, and other compounds—provides a wide range of options for enhancing polymer matrices. Many of these materials possess tunable structures that allow flexible chemical control, offering significant potential for developing inorganic-reinforced vitrimer nanocomposites. For instance, natural silicates exhibit rich structural diversity and are highly amenable to design, synthesis, and modification. Halloysite nanotubes (HNTs) are a naturally occurring nanotubular aluminosilicate clay mineral, traditionally used in ceramics and refractories, with chemical properties similar to kaolinite. Owing to their hollow tubular morphology and distinct inner/outer surface chemistry, HNTs have recently gained attention as nanocontainers, catalyst supports, polymer fillers, hydrogen-storage materials, and templates for nanomaterials, demonstrating considerable practical value [216–219]. Both the inner lumen and

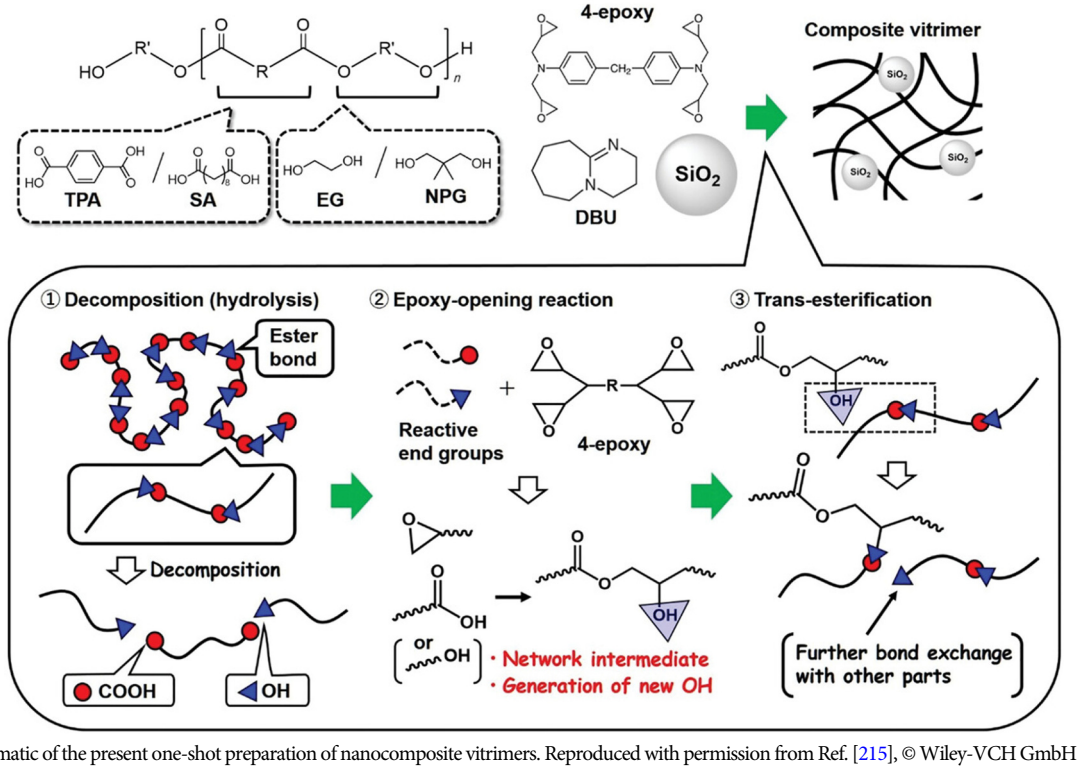

Figure 22 Schematic of the present one-shot preparation of nanocomposite vitrimers. Reproduced with permission from Ref. [215], © Wiley-VCH GmbH 2023.

outer surface of HNTs offer versatile design opportunities, enabling the introduction of specific functions into the final composite. Moghari et al. [220] successfully prepared HNTs-vitrimer nanocomposites by modifying the outer surface of HNTs with amino groups via silane coupling. This functionalization allowed the HNTs to participate in and promote the curing reaction of the epoxy network. Simultaneously, loading 8-hydroxyquinoline (8-HQ) into the lumens of the HNTs enabled pH-responsive release in corrosive environments, imparting self-healing and anti-corrosion properties to the epoxy coating. At a low loading of only 0.1 wt.%, the activation energy of the curing reaction decreased by 14% compared to that of the neat epoxy resin. Through differential functionalization of its inner and outer surfaces, HNTs act as multifunctional fillers that achieve multiple objectives at very low loadings—not only accelerating the curing process but also significantly enhancing the corrosion resistance of the material.

MXene, an emerging two-dimensional inorganic nanomaterial, possesses unique properties such as metallic-like high conductivity and excellent photothermal conversion efficiency [221–224]. In recent years, it has been widely incorporated into polymer matrices to enhance performance or introduce new functionalities. Its general chemical formula is $M_{n+1}X_nT_z$, where M stands for an early transition metal (e.g., Ti, V, Nb), X represents carbon or nitrogen, and T denotes surface functional groups (e.g., –O, –OH, –F). Carey et al. [225] integrated multiple functions—conductivity, mechanical reinforcement, and photothermal self-healing—into a nanocomposite by incorporating MXene into a vitrimer polymer network. This approach addresses common shortcomings of vitrimer materials, such as insufficient conductivity and limited mechanical strength, while introducing the highly practical capability of light-triggered repair. Functionally, MXene behaves similarly to carbon nanomaterials like CNTs within the composite. However, MXene offers unique advantages: its two-dimensional layered structure provides higher intrinsic electrical conductivity and a lower percolation threshold, enabling conductive pathways to form at lower loadings. Moreover, its surface is rich in functional groups, allowing stable and uniform dispersion in water and various polar solvents without requiring complex covalent modification. This greatly simplifies the processing of unmodified MXene into polymer matrices via physical blending and expands the feasibility of coating, spinning, and other fabrication techniques. In their work, MXene–vitrimer composites were prepared using a distinctive powder-coating and hot-pressing strategy (Fig. 23). First, micron-sized prepolymer powder was synthesized via precipitation polymerization. MXene nanosheets were then selectively coated onto the powder surface through acid-induced precipitation in an aqueous phase. Finally, the coated powder was consolidated by hot-pressing. During this process, MXene is driven to the boundaries between the vitrimer particles, spontaneously forming a continuous, biomimetic Voronoi-like conductive network. This design achieves multiple functionalities—conductivity, reinforcement, and photothermal self-healing—at extremely low filler content. The composite reaches the electrical percolation threshold with only 0.19 vol.% MXene, significantly lowering production costs while demonstrating that high-performance composites can be built with minimal filler loading. This approach substantially broadens the application potential of vitrimer composites in advanced fields such as wearable electronics, soft robotics, smart coatings, and aerospace.

In summary, the diversity of inorganic nanomaterials provides multiple avenues for designing nanofiller systems for vitrimers, encompassing both naturally occurring nanocrystals and synthetically engineered materials. This diversity not only enables the continuous development of composite systems with enhanced properties and unique stimulus-responsive functionalities, but also proves instrumental in addressing the multifaceted and complex demands of nanocomposite applications. In traditional epoxy thermoset nanocomposites, many nanofillers—such as titanium dioxide, boron nitride, and aluminum oxide—have been widely used. Their incorporation into epoxy vitrimers is equally feasible and highly promising. Furthermore, the sophisticated micro- and nano-structures found in natural inorganic materials offer a source of biomimetic inspiration, guiding the design and application of bio-inspired nanofillers. This significantly expands the scalability of vitrimer nanocomposite systems and underscores their considerable potential for future advancement.

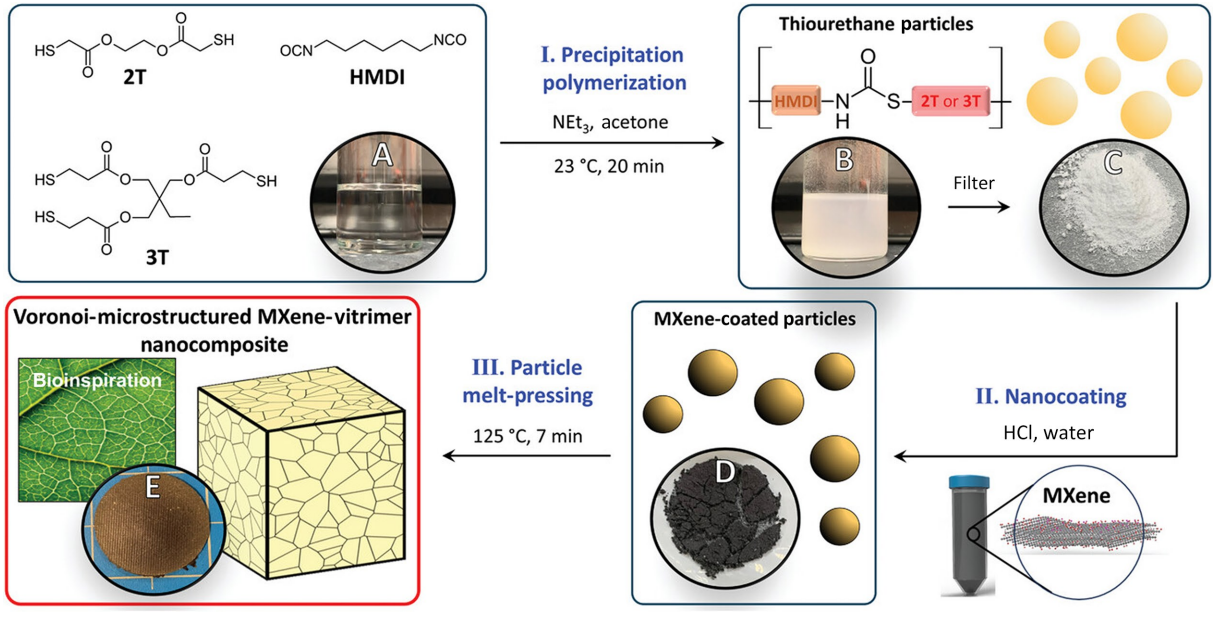

Figure 23 Fabrication of biphasic Mxene-coated vitrimer nanocomposite. Reproduced with permission from Ref. [225], © Wiley-VCH GmbH 2024.

6 Applications of epoxy vitrimer nanocomposites

As usage environments become increasingly complex and material requirements continue to rise, the demand for more advanced and versatile smart materials persists. By leveraging the multifunctional attributes of nanofillers and the tunable performance of polymer matrices, epoxy vitrimer–nanofiller composites exhibit a diverse range of functionalities and demonstrate broad application potential in many areas of daily life, with their scope continuously expanding into new fields. This section highlights several representative cutting-edge applications of epoxy vitrimer nanocomposites, illustrating the powerful functional capabilities that nanofillers impart to the vitrimer matrix.

6.1 Soft actuators

LCEs are specialized materials that combine the orientational order of liquid-crystal mesogens with the elasticity of cross-linked polymer networks [226]. The liquid-crystal units within LCEs can change their alignment in response to external stimuli such as heat, light, or magnetic fields, leading to reversible and often large macroscopic deformations. In soft robotics, LCEs are considered ideal materials for fabricating artificial muscles and flexible actuators, capable of achieving various biomimetic motions such as crawling, grasping, and rolling. Liquid crystal epoxy resins are also commonly used liquid crystal precursors [227, 228]. These are small molecules or oligomers bearing both liquid-crystal units and terminal epoxy groups, which can be cured to form thermosetting liquid-crystal polymers that retain local order within liquid-crystal domains. Such materials enable a variety of smart functions, including shape memory and soft actuation. Integrating vitrimer chemistry with LCEs not only endows the resulting materials with stimulus-responsive reversible actuation but also introduces self-healing and three-dimensional shape-reconfiguring capabilities.

Our group pioneered the integration of vitrimer chemistry with liquid crystal networks, incorporating the photothermal effect of CNTs into LCE actuators to achieve light-controlled reversible shape programming and actuation (Fig. 24) [89]. Irradiating stretched films with infrared light locally triggers transesterification within the vitrimer network, thereby inducing alignment of the liquid crystal units in selected regions. This process effectively "writes" predetermined bending or stretching deformations into the planar film, enabling reversible actuation under thermal or optical stimuli. The approach allows mold-free, light-directed

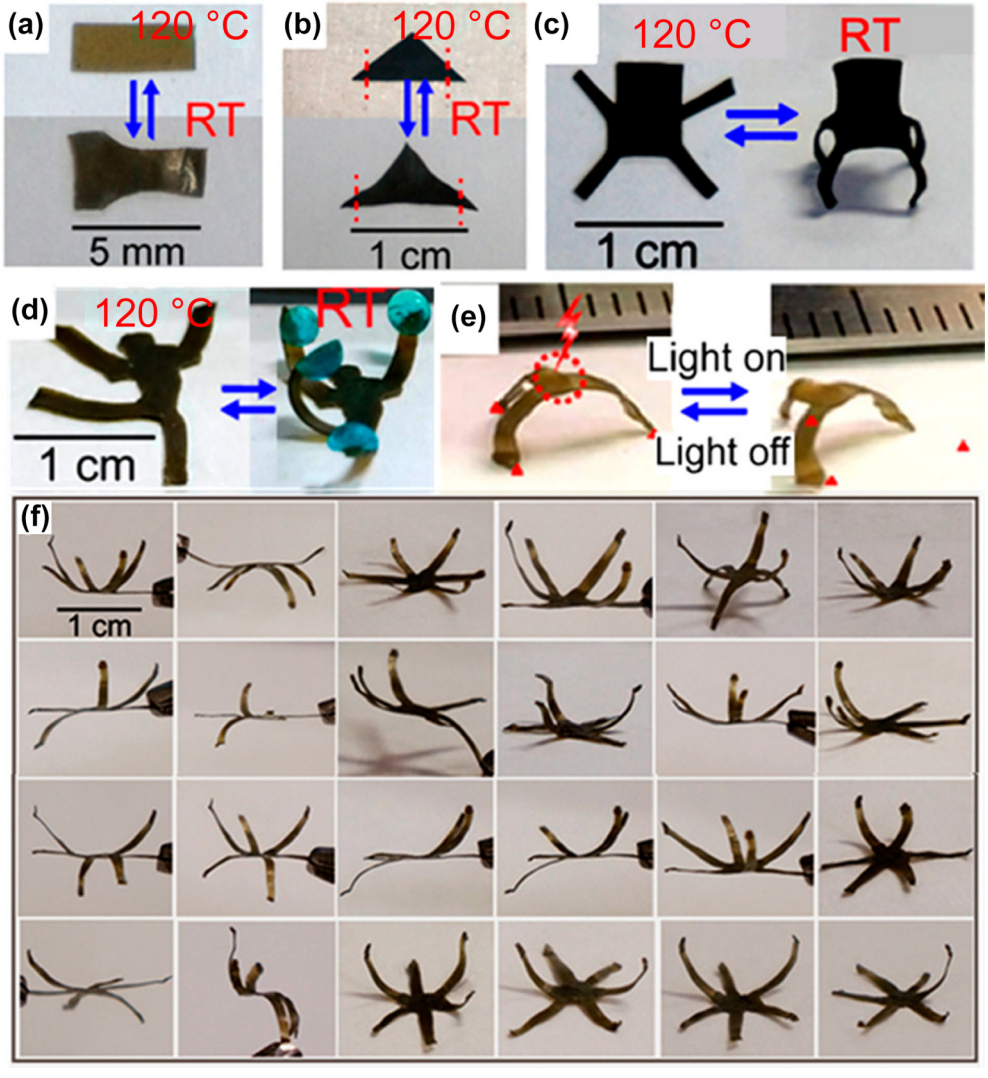

Figure 24 The reversible actuation of CNT-*x*LCEs based on different reversible shape changes under heat or light control. Reproduced with permission from Ref. [89], © American Chemical Society 2016.

programming of three-dimensional architectures. Moreover, the material permits photo-erasure and reprogramming, photo-welding of damaged areas, and photo-repair of microcracks—all achievable at remarkably low temperatures (e.g., −130 °C).

We have further developed magnetically responsive LCE actuators by physically blending paramagnetic Fe_3O_4 nanoparticles into a polyurethane-based LCE vitrimer network, creating magnetically responsive LCE-vitrimer composites [155]. Although this system is not based on an epoxy vitrimer, it offers valuable design principles for vitrimer materials across different matrices. The uniform dispersion of Fe_3O_4 nanoparticles enables the composite to utilize the synergistic interplay between the magnetocaloric effect and liquid-crystal phase transitions for actuation: under an alternating magnetic field, heat generated by the nanoparticles drives the liquid-crystal phase from an anisotropic to an isotropic state, inducing reversible macroscopic deformation. By programming the initial orientation of the liquid-crystal domains, we successfully realized diverse and complex actuation modes, including large contraction (up to ~ 80%), bidirectional contraction, helical elongation, and dynamic three-dimensional patterning. More importantly, by integrating regions with different Fe_3O_4 loadings into a single device, we exploited their distinct magnetothermal response thresholds to achieve localized and sequential actuation within a magnetic soft actuator—a first in such systems. contents into a single device, they exploited their distinct magneto-thermal response thresholds to achieve localized and sequential driving control over specific regions within a magnetic soft actuator for the first time. This breakthrough provides crucial insights for designing liquid crystal epoxy nanocomposites and opens avenues for developing integrated actuators with precisely controlled, multi-sensitivity responses. Inspired by this design of integrating composites with varying nanoparticle doping levels within a single device, we can envision integrating epoxy LCE units with diverse response modes—magnetic, optical, thermal—through spatially controlled doping or heterostructure welding. This approach could enable next-generation intelligent soft robots and multifunctional devices capable of executing complex sequential actions or adapting to dynamic environments.

The development of novel liquid-crystalline epoxy monomers and flexible crosslinkers, together with diverse strategies for nanoparticle selection and doping, is expected to yield a wide range of unique LCE actuators [229]. These intelligent soft materials are poised to make distinctive contributions to fields such as wearable devices and flexible electronics.

6.2 Conductive and electromagnetic shielding materials

With the rapid advancement and widespread adoption of electronic devices, electromagnetic interference (EMI) generated by numerous electronic systems not only disrupts the operation of surrounding equipment but also poses potential risks to human health and other biological systems. The use of electromagnetic shielding materials to attenuate electromagnetic radiation represents an effective approach to mitigate these hazards[230]. Compared to traditional metallic shields, conductive polymer composites are attracting growing attention in the field of EMI shielding due to their advantages such as light weight, low cost, ease of processing, corrosion resistance, and tunable electrical properties.

As noted earlier, vitrimer composites incorporating various carbon nanomaterials and two-dimensional fillers can exhibit excellent electrical conductivity. These materials preserve the favorable mechanical properties of polymers while providing conductive functionality, making them highly suitable for use as robust, stable interlayer structures in electromagnetic shielding. This approach opens new pathways for exploring advanced EMI shielding applications. In the work by Fang et al. [165], multi-walled carbon nanotubes (MWCNTs) were introduced as conductive fillers into an epoxy vitrimer network to develop high-performance electromagnetic shielding composites (Fig. 25). Instead of conventional physical blending, they used a liquid-phase adsorption and deposition process to uniformly coat cured epoxy vitrimer particles with MWCNTs. Subsequent hot-pressing consolidated the material and fully established the conductive network, resulting in strong inter-particle bonding while retaining a layered architecture of the polymer matrix at the MWCNT interfaces. The resulting EMI shielding composite shows an ultralow electrical percolation threshold of 0.066 wt.%, meaning only trace amounts of MWCNTs are needed to form conductive pathways. At an MWCNT loading of 2 wt.%, the electromagnetic interference shielding effectiveness reaches 22 dB; increasing the content to 4 wt.% raises the shielding effectiveness to 31.5 dB. Analysis of reflectance, absorbance, and transmittance coefficients indicates that absorption is the dominant shielding mechanism. Moreover, the vitrimer nanocomposite exhibits excellent mechanical strength, a broad processing window, and good reprocessability and recyclability. This work provides new design concepts and material solutions for next-generation sustainable, high-performance flexible electronic packaging and electromagnetic protection.

6.3 Smart coatings

Nanoparticle-reinforced vitrimer composites can serve as self-healing and recyclable smart coatings, where the type of stimulus response is determined by the functional properties of the nanofillers, creating an on-demand, multifunctional, and recyclable responsive system. Many nanoparticles possess hollow or tubular structures whose cavities can encapsulate specific functional molecules. Upon stimulation, these molecules can be released to impart additional functionality to the coating. For instance, Moghari et al. [220] encapsulated the corrosion inhibitor 8-hydroxyquinoline (8-HQ) within the lumens of halloysite nanotubes (HNTs), conferring pH responsiveness. This strategy transforms corrosion protection from a passive mechanism—relying solely on the stable, crosslinked polymer matrix—into an active one through stimulus-triggered inhibitor release. This responsiveness also enables monitoring of the metal surface's corrosion state and facilitates repair of underlying damage. Furthermore, in passive protection systems based on densely crosslinked vitrimer networks, the incorporation of carbon materials with strong photothermal conversion efficiency can impart photothermally responsive reversibility while simultaneously reinforcing the matrix. Lorwanishpaisarn et al. [231] constructed a vitrimer nanocomposite filled with carboxylated multi-walled carbon nanotubes (COOH-MWCNTs). This material exhibits enhanced mechanical properties and achieves near-infrared (NIR) light-triggered self-healing (Fig. 26). Beyond its inherent strength and corrosion resistance, the dynamic nature of the vitrimer grants the coating sustainable self-repair and recyclability. Such light-responsive smart composite coatings hold broad application prospects in marine engineering, ship protection, pipeline systems, and related fields. Benefiting from high photothermal conversion efficiency, localized coating damage can be controllably repaired via

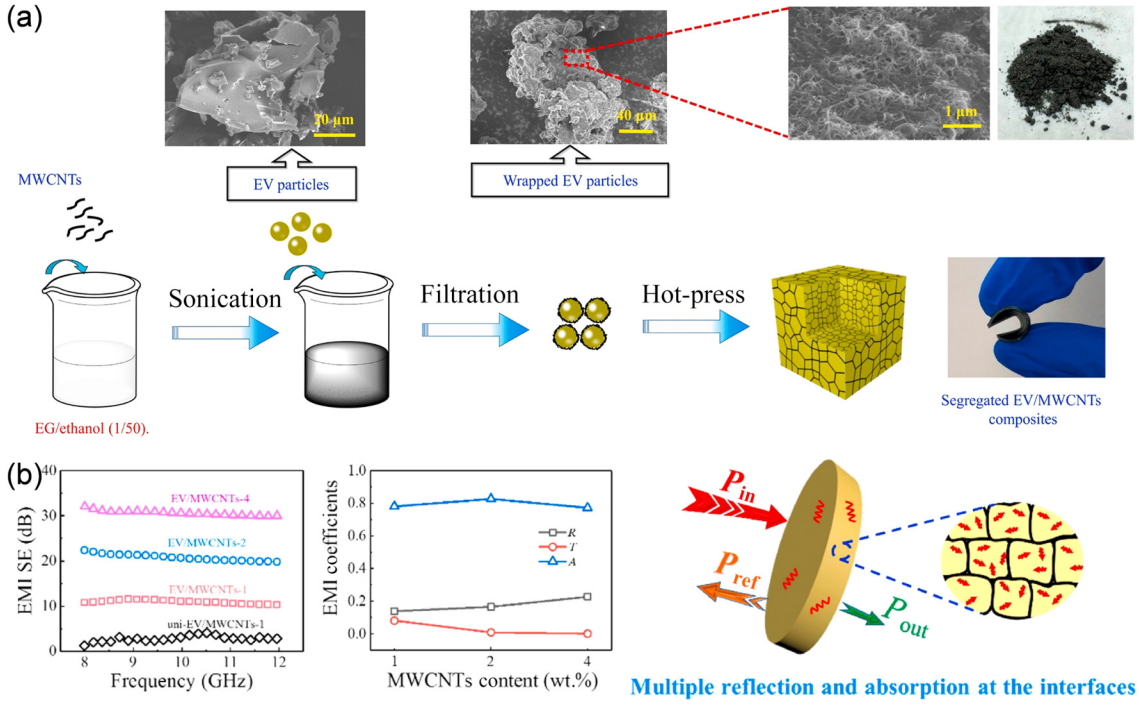

Figure 25 (a) Schematic illustration of the fabrication of the segregated MWCNTs-vitrimer nanocomposites by compression molding. (b) Electromagnetic interference shielding effectiveness of MWCNTs-vitrimer nanocomposites and schematic illustration of the electromagnetic interference shielding mechanism. Reproduced with permission from Ref. [165], © Elsevier Ltd. 2021.

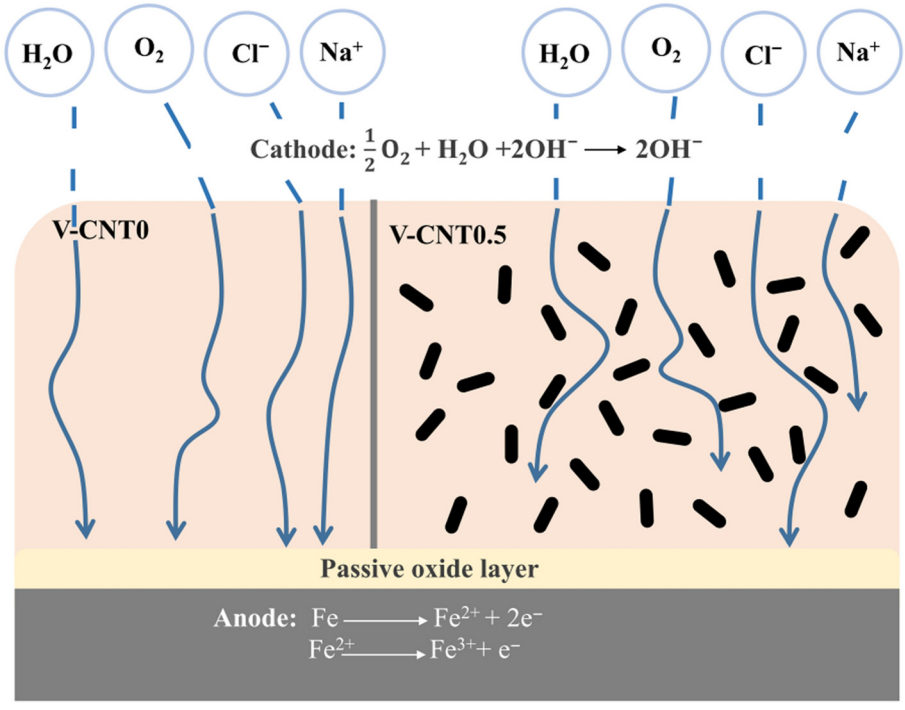

Figure 26 Corrosion protection of steel coated with nanocomposites. Reproduced with permission from Ref. [231], © Springer Nature 2022.

directional light irradiation, providing a key technological advance for sustainable coating development. When the bulk coating material is compromised, its dynamic covalent network allows the coating to be "erased", enabling both internal defect repair and full material recovery. This innovative design circumvents the challenge faced by traditional, non-reprocessable coatings, which must be discarded after a single service life.

6.4 Sensors

Due to the stimulus-responsive characteristics of vitrimers, nanocomposites doped with nanofillers inherently inherit all the properties and functions of the vitrimer matrix. Moreover, they acquire additional functionalities from the fillers, which significantly broaden their application scope. For instance, sensing in electronic devices relies largely on electrical conductivity. Nanoparticles such

as CNTs and graphene exhibit outstanding conductivity, thereby enhancing the sensing capabilities of the resulting nanocomposites. Consequently, nanocomposites combining carbon nanomaterials with vitrimers hold promising potential for use in electronic sensing devices. Jia et al. designed a bio-based vitrimer matrix and incorporated graphene and GO nanoparticles to prepare conductive nanocomposites, which were applied as coatings on steel plates (Fig. 27) [232]. Upon heating, the GO in the composite loses its oxygen-containing functional groups and converts into reduced graphene oxide (RGO), leading to a decrease in resistivity and the formation of conductive pathways. Based on this principle, the composite was employed as a temperature and fire alarm sensor: The triggered change in conductivity activates an external connected circuit, thereby setting off an alarm.

In recent years, flexible wearable electronic sensors have attracted extensive research interest. These devices are characterized by flexibility, stretchability, and even self-healing capability, allowing them to conform comfortably to human skin or textiles for real-time, continuous monitoring of physiological signals (e.g., heartbeat, muscle activity, joint movement) or environmental parameters. Nanoparticle-reinforced vitrimer materials represent promising candidates for such applications. Yang et al. [233] designed and synthesized a dynamic epoxy elastomer using disulfide bonds as the dynamic exchange linkage (Fig. 28). The resulting material exhibits high tensile strength, large strain capability, efficient self-healing, and recyclability. Differing from conventional approaches, their fabrication process first prepares a vitrimer elastomer, which is then pre-stretched. A dispersion of carboxylated MWCNTs is sprayed onto the stretched surface and dried. Upon release of the pre-stretch, the contracting elastomer causes the adhered CNT layer to buckle, forming firmly bonded conductive micro-wrinkles and protrusions. When the elastomer is stretched again, the contact points between these conductive features decrease, raising the electrical resistance and enabling strain sensing. This principle has been successfully applied to monitor human motion and recognize object shapes.

7 Conclusion and outlook

Epoxy vitrimer nanocomposites constitute an evolving frontier in polymer science and sustainable materials engineering. By merging the dynamic networks of vitrimers with the multifunctional attributes of nanofillers, this field has progressed beyond simply improving the properties of conventional epoxy resins. It now provides a feasible route to reconcile the long-standing trade-off between high performance and recyclability in thermosetting polymers. The synergistic interaction between the vitrimer matrix and the nanofillers—where each component actively influences and enhances the properties of the other—has unlocked novel functionalities, including remotely triggered self-healing, stimulus-

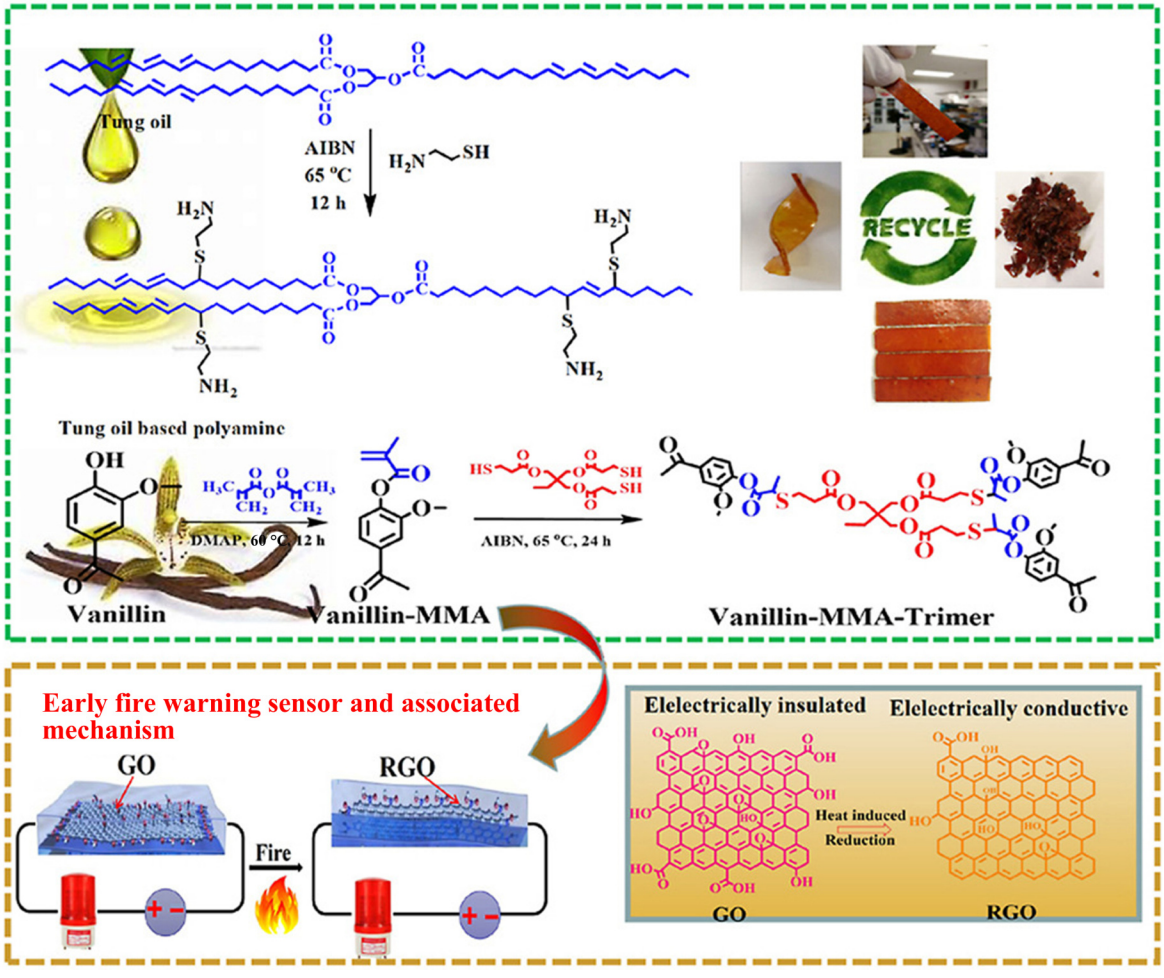

Figure 27 Preparation of vitrimer and schematic structure of GO-vitrimer nanocomposites and its application in early fire warning sensor. Reproduced with permission from Ref. [232], © Elsevier Ltd. 2022.

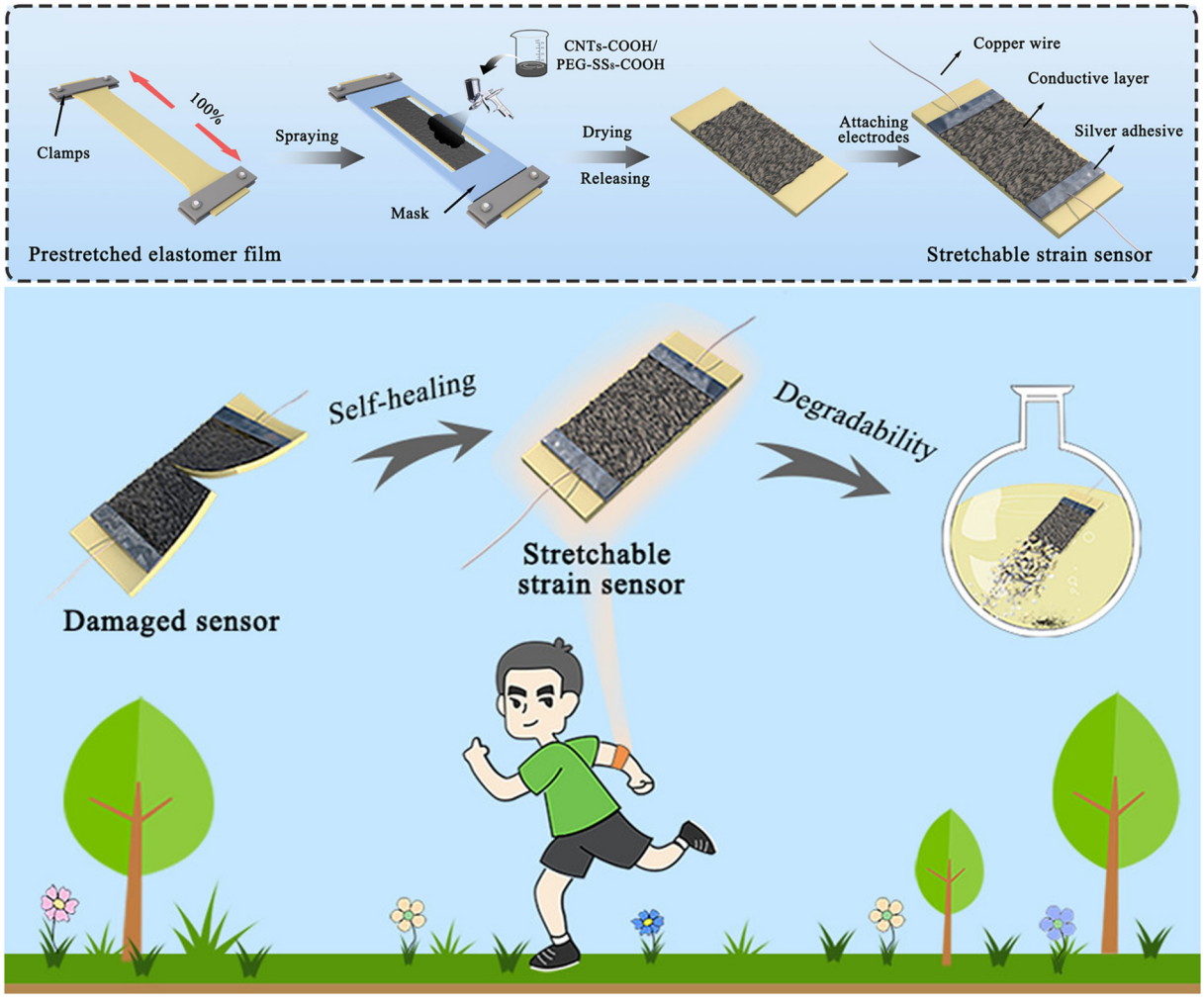

Figure 28 The preparation process of vitrimer elastomer film-based stretchable strain sensor nanocomposite and its potential application in wearable electronics. Reproduced with permission from Ref. [233], © American Chemical Society 2022.

responsive shape reconfiguration, and built-in electrical or magnetic response. This review aims to provide insights and a reference point for the development of vitrimer nanocomposites, extending even beyond epoxy-based systems. Many of the nanofiller incorporation strategies discussed here share common principles that can be adapted to other vitrimer matrices, thereby offering valuable experience for the design of more innovative and high-performance vitrimer nanocomposites in the future.

Despite the inspiring advances demonstrated at laboratory scale, significant challenges must be overcome to transition these materials from research prototypes into widely adopted engineering solutions. Foremost among these are the lack of standardized, scalable manufacturing processes, the need to verify long-term stability and durability under real-world service conditions, and the establishment of efficient closed-loop recycling infrastructures. Furthermore, a deeper understanding and precise tailoring of the interfacial interactions between nanofillers and the dynamic covalent network are essential, as these interactions critically govern both mechanical reinforcement and functional performance.

Looking ahead, the continued integration of dynamic covalent chemistry, nanotechnology, and green engineering principles will be essential for advancing this field. Future efforts should prioritize the design of bio-derived or intrinsically catalytic vitrimer systems, the development of industrially scalable composite fabrication routes, and the implementation of integrated lifecycle management strategies. As these challenges are addressed, epoxy vitrimer nanocomposites are poised to become key enablers in the transition toward a circular materials economy, with impactful applications in flexible electronics, soft robotics, sustainable coatings, and beyond.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

S. H. Z.: Writing, reviewing and editing the manuscript. Z. Y. L., J. Y. Z., E. J. H., Z. J. Y., Y. X. W., and W. D. T.: Reviewing the manuscript. Q. L. C., C. G., and G. L. W.: Project administration, funding acquisition. Y. W.: Reviewing the manuscript and supervision. Y. J.: Reviewing and editing the manuscript, project

administration, funding acquisition, supervision. All the authors have approved the final manuscript.

Use of AI statement

None.

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