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## Advances in plant oil-based polymeric materials with dynamic covalent bonds

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With the current unstable supply of petroleum resources, the environmental pollution problems, as well as the increasing demand for sustainable development, plant oils, which are natural, renewable resources, have been regarded as potential alternative resources to petrochemical-based materials for fabricating polymers due to their advantages, such as renewability and wide availability. However, traditional plant oil-based polymer materials have limitations in terms of recyclability and reprocessability. In recent years, dynamic covalent bond polymers have undergone rapid development and hold significant promise for addressing the inherent trade-off between performance and sustainability in traditional polymeric materials, thereby offering innovative solutions for sustainable development. Herein, we have summarized the fundamental characteristics of plant oil resources, along with the evolution and characteristics of dynamic covalent bonds. The two primary exchange mechanisms of covalent adaptable networks (CANs) and recent advances in plant oil-based dynamic covalent polymer materials are also reviewed. Additionally, we discuss the utilization of various dynamic covalent bonds in polymer material design while providing perspectives on future developments in this field.

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### Sustainability spotlight

With the continuous advancement of sustainable development in recent years and the increasingly prominent limitations of traditional petroleum-based materials, the efficient utilization and functional expansion of biomass materials, such as plant oils, have become a research hotspot, and the dynamic covalent bond polymers have emerged as an effective solution that balances strength and sustainability. In order to present the cutting-edge work of plant oil-based biomass materials in the field of dynamic covalent polymer research, this review surveys the innovative use of plant oils in fabricating polymeric materials with dynamic covalent bonds. Our review also presents several prospects for the future development of plant oil-based dynamic covalent polymers, aiming to provide inspiration for advancement in related fields.

## 1. Introduction

With the continuous advancement of sustainable development, biomass resources have attracted widespread attention in the development of polymers. Based on the processing properties, polymeric materials can be classified into two main categories: thermoplastics and thermosets. Thermoplastics soften and can be reshaped repeatedly when heated, but their poor heat resistance limits their application in high-temperature environments. In contrast, once molded and cured, thermosets neither soften nor melt when heated, which renders them non-recyclable and makes waste disposal challenging, thereby increasing the risks of resource waste and environmental pollution.<sup>1</sup> Additionally, the processing of thermosets involves a long curing time and low production efficiency, and harmful gases may be released during

the curing process, posing certain environmental and health hazards. Therefore, utilizing green and renewable resources to produce polymers can not only reduce the dependence on limited petroleum resources but also effectively decrease carbon emissions, mitigate the greenhouse effect, and foster the prosperity of agriculture-related industries.

In the field of polymer science, the emergence of dynamic covalent bond polymers (DCPs) has narrowed the gap between thermosets and thermoplastics. While maintaining or even enhancing mechanical properties, DCPs exhibit exceptional self-healing and recyclability capabilities. Meanwhile, plant oils, with their characteristics of renewability, wide availability, low cost, and environmental friendliness, have become crucial feedstocks for DCPs. Plant oils are rich in unsaturated fatty acids, which can be chemically modified to construct polymer networks with specific dynamic covalent bonds (DCBs). Common DCBs in the preparation of plant oil-based DCPs include hydroxyl-ester bonds, imine bonds (Schiff bases), disulfide bonds, urethane bonds, boronic ester bonds, Diels-Alder adducts, and sterically hindered urea bonds. The

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introduction of DCBs endows materials with self-healing, recyclability, and reprocessable properties,<sup>2</sup> which may also impact the chemical properties and stability of materials to some extent. Research in this field still faces significant challenges to achieving the overall optimization of material properties.

## 2. Plant oil resources

Plant oils are a type of natural oils extracted from the seeds, fruits, or germ of plants and include castor oil (CO), epoxidized soybean oil (ESO), tung oil (TO), palm oil (PO), epoxidized linseed oil (ELO), and epoxidized canola oil (ECaO). According to the prediction of the United States Department of Agriculture (USDA), in the 2025/26 sales year, the global oilseed production is expected to increase to 6.93 billion tons (Fig. 1); the global plant oil production will reach nearly 2.35 billion tons, and the consumption is expected to reach 2.289 billion tons.<sup>3</sup> In global plant oil consumption, the food sector accounts for 72%, while the industrial usage (including biofuel production) is also continuously increasing and is expected to reach 641 million tons. The double growth in both production and consumption reflects the significant position of plant oil in the global market and its wide application in both the food and industrial sectors.

The main component of plant oils is triglycerides, which are esters formed by the three hydroxyl groups of the glycerol molecule and fatty acids. They are widely used as raw materials for the production of multifunctional elastomers, coatings, nutritional products, and medicines, demonstrating significant industrial value in corresponding fields. The unsaturated fatty acids in plant oils can be modified through various chemical reactions, thereby introducing different functional groups (Fig. 2). For example, the carbon-carbon double bonds in plant oils undergo epoxidation reactions to form epoxy groups, which can react with carboxyl groups, hydroxyl groups, amino groups, *etc.*, to form DCBs. The ester bonds of triglycerides are introduced through esterification reactions, and the hydroxyl groups can react with boronic acid to form boronic ester bonds. These ester bonds and boronic ester bonds can undergo dynamic exchange reactions under acidic or basic conditions, confirming that plant oils can be transformed into high-value chemicals

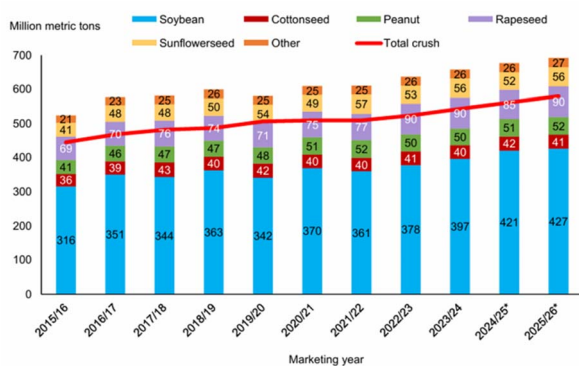


Fig. 1 Global major oilseed production and processing. Reproduced with permission.<sup>3</sup> Copyright 2025, the U.S. Department of Agriculture, Economic Research Service.

through chemical modification and the introduction of DCBs. These chemicals can be used as monomers, oligomers, or intermediates, and are further utilized to prepare various polymers, such as polyurethanes,<sup>4</sup> epoxy resins,<sup>5</sup> alkyd resins,<sup>6</sup> *etc.* Therefore, with the pursuit of sustainability, based on the features of plant oils, the research and development of plant oil-based polymers can provide new solutions for environmental protection and resource utilization.

## 3. DCPs

As an important part of DCPs, DCBs are a special type of covalent bond capable of reversible cleavage and reformation under stimuli like light, heat, pH changes, and redox reactions. Their bond energy lies between stable covalent bonds and non-covalent bonds (*e.g.*, ionic bonds, coordination bonds, hydrogen bonds, or van der Waals forces). While the reversible DCBs were discovered as special cases in the mid-20th century, their broader significance was later recognized. Subsequently, in 1999, Lehn<sup>7</sup> introduced DCBs into the research of supramolecular chemistry. In the 21st century, the research on DCBs witnessed a breakthrough. In 2001, the Nobel Prize winner in Chemistry, Barry Sharpless, proposed the concept of “Click Chemistry”,<sup>8</sup> emphasizing the construction of complex molecular structures through rapid, efficient, and selective chemical reactions. This idea coincides with the characteristics of DCBs, providing new ideas for their application. Subsequently, Kloxin *et al.*<sup>9</sup> introduced the concept of “Covalent Adaptable Networks” (CANs), which is specifically used to construct cross-linked polymer networks containing reversible covalent bonds. At this point, chemists began to introduce DCBs into various fields such as materials science, organic synthesis, and biochemistry. For instance, Leibler *et al.*<sup>10</sup> designed an epoxy resin with DCBs that cross-linked with acid anhydrides to form a dynamic covalent network. This network could rearrange its topological structure through ester exchange reactions at high temperatures without depolymerization, exhibiting glass-like fluidity at high temperatures and combining the advantages of thermosets and thermoplastics. Leibler *et al.* referred to such polymers as “vitrimers”, which are composed of vitreous and polymer components. Subsequently, researchers gradually explored more types of DCBs, such as Diels–Alder adducts, hydroxyl–ester bonds, imine bonds (Schiff bases), disulfide bonds, urethane bonds, sterically hindered urea bonds, boronic ester bonds, and carbamate bonds, further expanding the performance and application scope of vitrimers.

CANs can be classified into dissociative and associative types based on the exchange mechanism.<sup>11</sup> The dissociative exchange involves the breaking of existing bonds before forming new ones, and the viscosity drops rapidly due to the decreased crosslink density (Fig. 3a). When the temperature rises, the equilibrium shifts towards dissociation, significantly lowering viscosity and imparting transient thermoplastic-like behavior. The Diels–Alder addition product is a representative example of the dissociative CANs. As the core mechanism of CANs, the associative exchange mechanism enables dynamic topological rearrangement while maintaining mechanical properties at elevated temperatures (Fig. 3b). DCBs, such as hydroxyl–ester



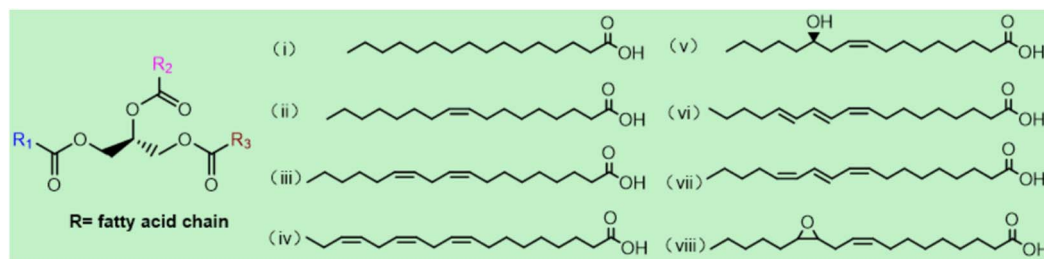


Fig. 2 Schematic of the structure of plant oils. (i) Palmitic acid. (ii) Oleic acid. (iii) Linoleic acid. (iv) Linolenic acid. (v) Ricinoleic acid. (vi) Tung oil acid. (vii) Pomegranate acid. (viii) Vernonia acid.

bonds and boronic ester bonds, follow the associative exchange mechanism. The dynamic reactions of classic DCBs are illustrated in Fig. 3c. Moreover, the exchange mechanism of DCBs is influenced by multiple factors, including the dynamic bond type, the presence of catalysts, the network topology, temperature, and environmental conditions. Generally, the tunability of CANs remains a key advantage, and the specific application requirements can be met by strategically selecting the bond type, catalysts, and reaction conditions.

## 4. Plant oil-based polymers containing dynamic covalent bonds

### 4.1 Hydroxyl-ester bonds

Due to the presence of natural hydroxyl groups, double bonds, and ester groups, plant oils can be used to effectively construct

CANs of hydroxyl-ester systems with potential for extensive chemical modification. The dynamic transesterification reaction of hydroxyl-ester bonds enables topological rearrangement at elevated temperatures, imparting thermoplastic-like processability. However, traditional transesterification reactions usually require a relatively high temperature (above 100 °C) and a catalyst (such as zinc acetate<sup>13</sup>) to accelerate the reaction rate, increasing energy consumption and the risk of catalyst degradation and loss, which compromises long-term stability and compatibility. To mitigate these issues, catalyst-free synthesis of hydroxyl-ester polymers can be achieved by introducing excessive hydroxyl groups or catalytically active tertiary amines, thereby reducing potential environmental impacts.

**4.1.1 Epoxidized soybean oil.** Epoxidized soybean oil (ESO) is derived from soybean oil extracted from soybean seeds. The reactive epoxy groups in ESO enable ring-opening reactions with amino groups, carboxyl groups, phosphoric acid, and

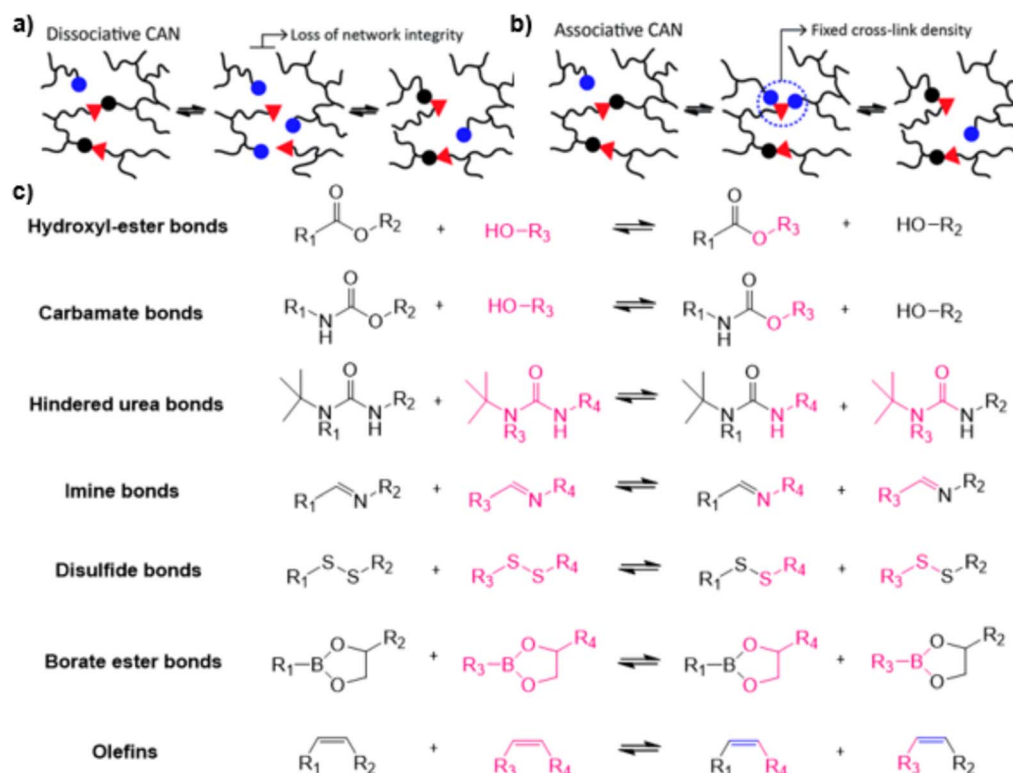


Fig. 3 (a) and (b) Two exchange mechanisms of CANs. Reproduced under the terms of the CC-BY license.<sup>12</sup> Copyright 2016, Wim Denissen, The Royal Society of Chemistry. (c) The dynamic reactions of some DCBs.



phosphate esters, *etc.* Its application in environmentally friendly materials can be further expanded by modification, meeting the requirements of sustainable development. In this regard, Wu *et al.*<sup>14</sup> synthesized a fully biobased and recyclable dynamic covalent network vitrimer *via* the esterification reaction between glycyrrhizic acid and ESO. The rigid framework of glycyrrhizic acid imparted the vitrimer with excellent thermal stability and mechanical properties, and the vitrimer demonstrated biocompatibility and chemical degradability in ethylene glycol. Similarly, Neymara *et al.*<sup>15</sup> selected ESO and L-tartaric acid as raw materials to fabricate a catalyst-free and reprocessable biobased system. The additional hydroxyl groups in L-tartaric acid significantly promoted the ring-opening of the epoxy groups and the ester exchange reaction, increasing the curing and dynamic exchange rates. The resultant material exhibited excellent thermal stability ( $T_{d5\%} = 262\text{--}287\text{ }^{\circ}\text{C}$ ) and still maintained high performance after repeated processing, providing a new pathway for the development of sustainable biobased thermosetting materials.

To further expand the biobased monomer options, Li *et al.*<sup>16</sup> turned their attention to natural sugar alcohols, such as glycerol, erythritol, xylitol, and sorbitol. They combined these sugar alcohols with ESO to synthesize all-biobased epoxy resin matrix materials (Fig. 4). These bio-based vitrimers exhibited good mechanical properties at room temperature, and all samples except Xyl0.6 can effectively repair cracks within 1 h at 160  $^{\circ}\text{C}$  owing to the transesterification reactions. To simultaneously enhance the thermal resistance and mechanical strength of the material, Yang *et al.*<sup>17</sup> incorporated rosin-derived rigid monomer fumaric acid into ESO to prepare a fully biobased epoxy resin. The study found that the introduction of fumaric acid significantly increased the glass transition temperature ( $T_g$ ) to 65  $^{\circ}\text{C}$  and enhanced the mechanical properties, with the tensile strength reaching 16 MPa. Transesterification reactions enabled this material to exhibit self-healing, triple shape memory, and repeatable processing characteristics.

Beyond structural composites, ESO-based dynamic networks show great potential in functional materials and advanced

manufacturing technologies. Grauzeliene *et al.*<sup>18</sup> synthesized polyethylene glycol/ESO monomers and developed a flexible phase-change composite material with shape stability, high thermal conductivity, and 3D printing capability. Leveraging the synergistic effect of photoinitiators and transesterification catalysts, the material had both shape memory (100% shape recovery rate) and recyclability (*via* alcoholysis). Subsequently, Grauzeliene *et al.*<sup>19</sup> synthesized a bio-based resin using 2-hydroxy-3-phenoxypropyl acrylate, tetrahydrofurfuryl methacrylate, and acrylated epoxidized soybean oil (AESO) as raw materials through a free radical polymerization reaction. The incorporation of tetrahydrofurfuryl methacrylate significantly enhanced the tensile strength and reprocessing efficiency. This material exhibited excellent performance in digital light processing 3D printing, including high resolution, rapid curing, and material diversity.

**4.1.2 Castor oil.** Castor oil (CO), a prominent non-edible plant oil, contains various active groups such as ester groups, hydroxyl groups, and carbon-carbon double bonds, which enable it to synthesize a variety of derivatives with specific properties, thus exhibiting the potential to develop high-value-added polymers.<sup>20</sup> Maleated castor oil (MCO) was grafted onto post-consumer recycled polypropylene (PCR-PP) in the molten state by Dey *et al.*<sup>21</sup> to synthesize a polypropylene-based vitrimer containing dynamic ester exchange bonds (Fig. 5). This design not only solved the chain breakage problem caused by traditional grafting methods, but also introduced single or double dynamic CANs. The mechanical performance recovery rate remained above 90% after five recycling cycles, which was conducive to the closed-loop recycling of polypropylene.

Further expanding CO's applications, Amornkitbamrung *et al.*<sup>22</sup> synthesized a benzoxazine monomer by the Mannich reaction using vanillin, furfurylamine, and epoxidized castor oil (ECO), and carried out the ring-opening polymerization reaction with the ECO and introduced glutaric anhydride for

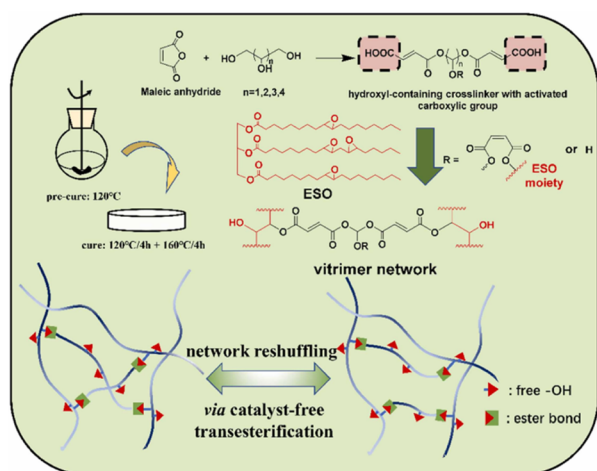


Fig. 4 The synthesis route of the ESO-SA polymer. Reproduced with permission.<sup>16</sup> Copyright 2025, Elsevier.

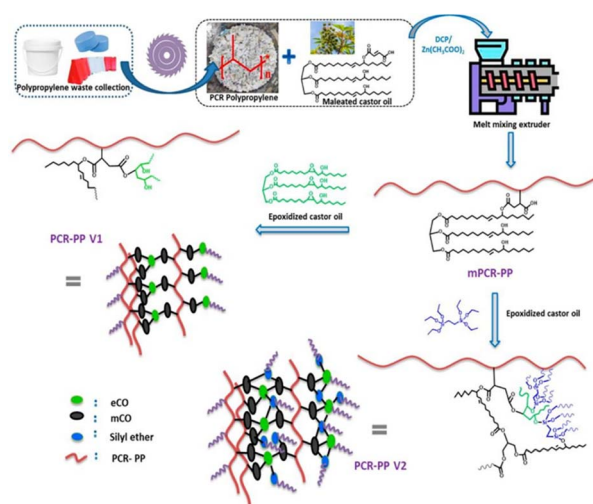


Fig. 5 Synthesis of the PCR-PP-based vitrimers with single and dual CANs. Reproduced with permission.<sup>21</sup> Copyright 2023, ACS Publications.



functionalization to prepare a benzoxazine/epoxy copolymer without fillers. This material achieved crack healing through the ester exchange reaction of the glutaric anhydride functional group, offering a strategy for the development of light-responsive self-healing polymers without NIR photothermal conversion fillers. Bergoglio *et al.*<sup>23</sup> synthesized solvent-free bio-based vitrimers *via* ultraviolet (UV) light-induced curing of ECO, utilizing the photoinitiator triphenylsulfonium hexafluorophosphate and the catalyst dibutyl phosphate. This material exhibited a high heat resistance of up to 200 °C, which is suitable for environmentally friendly coatings and adhesives. Cellai *et al.*<sup>24</sup> used ECO, a novel eugenol-based phosphate ester, and recycled carbon fibers to develop a fully bio-based adhesive. The bonding strength of the adhesive on ceramic composites reached 0.19 MPa, and the bonding strength on aluminum reached 0.14 MPa. After two re-bonding cycles at 90 °C, the initial strength of the adhesive could still be maintained from 74% to 91%. This demonstrated that combining bio-based monomers, recycled carbon fillers, and CANs can produce sustainable and reprocessable adhesives that are suitable for flexible electronics and circular composite applications. Most recently, CO was utilized to fabricate epoxidized methacrylated castor oil (EMCO) by Zhang *et al.*<sup>25</sup> *via* methacrylic anhydride modification and epoxidation. Then, it was combined with itaconic acid through an epoxy-acid ring-opening reaction and free radical polymerization to prepare a fully bio-based vitrimer. In summary, this material achieved efficient closed-loop recycling without significantly sacrificing mechanical properties, thus extending the service life and promoting the sustainable development of CO-based polymer materials.

**4.1.3 Tung oil.** Tung oil (TO) is an important renewable plant oil resource that possesses unique chemical properties due to the multiple conjugated double bonds and ester groups in its molecules. Researchers have developed various types of TO-based materials with diverse properties and great potential for applications through ingenious molecular design and dynamic chemistry. In the field of basic high-performance vitrimers, Li *et al.*<sup>26</sup> developed a recyclable and degradable epoxy vitrimer based on TO and bisphenol A diglycidyl ether. The resulting material exhibited a tensile strength of 59.21 MPa and could be repaired at 180 °C for 6 h. Moreover, it achieved shape memory (triple shape memory performance) and mechanical recycling functions through dynamic ester exchange reactions.

Moving towards application-oriented materials, Bei *et al.*<sup>27</sup> addressed the limitations of lignin-based photothermal materials (low strength, unstable response, and poor recyclability) by developing a lignin-tung oil covalent adaptable network through crosslinking with glycidylated lignin and tung oil triacid. The tensile strength of the material reached 19.97 MPa, and it also had high adhesion strength (6.80 MPa) and chemical degradability. When combined with a thermoelectric generator (LOT system), a stable open circuit voltage (125 mV) can be generated. This material provided a sustainable, high-performance solar energy collection solution with a biomass content of 75%. Zhang *et al.*<sup>28</sup> employed TO and malic acid, microwave-assisted synthesis, and dynamic covalent chemistry techniques to synthesize UV-curable oligomers and bio-based

reactive diluents to prepare UV-curable coating materials. The self-healing efficiency, welding efficiency, and shape fixation rate of this material were 92.5%, 171.8%, and 98.2%, respectively. This coating integrates environmental friendliness, high performance, and intelligent response, providing a sustainable high-performance coating for construction and automotive applications.

Further expanding the functionality and sustainability of TO-based vitrimers, Chen *et al.*<sup>29</sup> synthesized two tung oil derivatives (tung oil maleic anhydride (TOMA) and epoxidized tung oil ethanalamides (ETOE)), and prepared degradable tung oil-based vitrimers (PETVs) by combining them with multi-walled carbon nanotubes (MWCNTs) (Fig. 6). The composites exhibited excellent electrical conductivity and thermal conductivity, and could achieve rapid self-healing under various stimuli, such as electricity, infrared radiation, and heat. For instance, the self-healing efficiency of the PETV-1 reached 94.7% by heating for 10 min at 160 °C. More importantly, this material can be completely degraded under alkaline conditions, enabling the recycling and reuse of MWCNTs, facilitating the design of green and high-performance thermosetting composites.

The intelligent response characteristics of TO-based materials have been further explored for applications in functional devices. TO-based triglycidyl ester, isophorone diamine, and isophorone diamine carbonate were used by Tian *et al.* to prepare a degradable and programmable foam with dynamic hydroxyl-ester bonds.<sup>30</sup> This foam could achieve shape-color dual-response functionality by introducing thermochromic microcapsules, enabling its application in intelligent sensing fields such as high-temperature warning systems.

Progressing towards sustainable and catalyst-free vitrimers, Xu *et al.*<sup>31</sup> synthesized the TO-based triglycidyl ester (TOTGE) with terminal epoxy groups, and a fully bio-based and catalyst-free vitrimer was prepared by combining it with citric acid (CA). This material can achieve self-healing (the self-healing efficiency reached 93.2% at 180 °C for 15 min), shape memory, and recyclability without external catalysts, and can be applied in

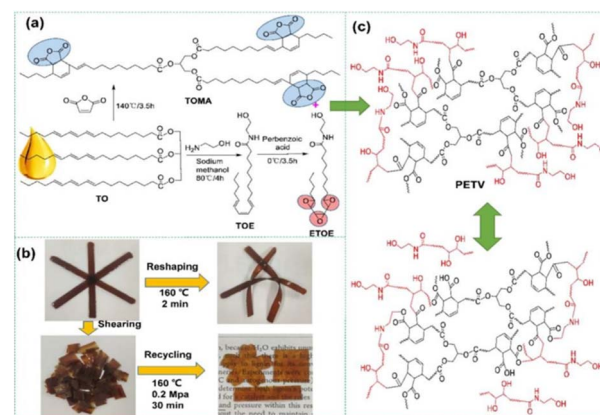


Fig. 6 (a) Synthesis route of TOMA and ETOE; (b) schematic diagram of reshaping, cutting, and recycling of PETV-1; (c) mechanism of transesterification in polymer networks. Reproduced with permission.<sup>29</sup> Copyright 2023, ACS Publications.



fields such as adhesives and sensors. Extending towards high-performance composites based on this catalyst-free approach, Xu *et al.*<sup>32</sup> used TOTGE and 1,8-diamino-*p*-menthane methyl diamine (MDA) to prepare the TOTGE-MDA vitrimer with dynamic ester exchange reaction ability, and applied it to carbon fiber-reinforced composite materials (CFRCs). The tensile strength of TOTGE-MDA vitrimers can reach 57.6 MPa, and the obtained CFRCs not only had a strength of 541 MPa, but also possessed self-healing (the self-healing efficiency reached 94.5% at 180 °C for 30 min), shape-memory, and recyclability properties, providing a new strategy for high-performance recyclable composite materials in fields such as aerospace and automotive industries.

**4.1.4 Epoxidized linseed oil.** The molecules of linseed oil contain unsaturated carbon-carbon double bonds, and epoxidized linseed oil (ELO) can be fabricated after ring oxidation treatment, which can undergo ring-opening reactions with various compounds *via* epoxy groups. ELO is widely used as a plasticizer, polyvinyl chloride heat stabilizer, in coatings and inks, and can also be used in cosmetics and food packaging materials. Shaikat *et al.*<sup>33</sup> synthesized the acrylated flaxseed oil (AELO) monomer using ELO and acrylic acid, and subsequently prepared a dynamic thiol-acrylate photoinitiating polymer. Through dynamic ester exchange reactions, the material can achieve stress relaxation and network rearrangement at high temperatures, thereby endowing it with self-healing (it can be thermally activated and healed at 180 °C) and shape memory functions, which can be utilized as flexible electronic skin and soft robots.

**4.1.5 Epoxidized canola oil.** Epoxidized canola oil (ECaO) is usually obtained by subjecting canola oil to ring oxidation using oxidants (peroxides, peracetic acid, *etc.*) in the presence of acidic or alkaline catalysts. It can be used in preparing biobased polyether polyols for the production of polyurethane foams and further lubricants, coatings, and plastic additives. Rodrigues *et al.*<sup>34</sup> developed bio-based vitrimers based on ECaO and lactic acid (LA), which had rubber-like flexibility, relatively low  $T_g$ , and a degradation temperature of approximately 300 °C. The thermal expansion analysis confirmed the good recyclability and reusability.

## 4.2 Imine bonds

The imine bond (C=N) is formed by the condensation of an amine and an aldehyde or ketone<sup>35</sup> and can undergo thermodynamically reversible formation and cleavage reactions. Aldehydes are more likely to form imine bonds than ketones, and primary amines rather than secondary amines tend to form imine bonds. The imine covalent bond can undergo the following three dynamic reactions: (1) the imine bond reacts with a primary amine within the system under acidic or neutral conditions to undergo transamination. (2) A double decomposition reaction occurs between two imine bonds. (3) Under acidic or alkaline conditions, it hydrolyzes into an amino group and a carbonyl compound, and re-forms the imine bond under neutral conditions. In research on developing imine bonds, vanillin and its derivatives play a core role in constructing

recyclable bio-based DCPs.<sup>36</sup> The dynamic imine bonds provided by them are the basis for achieving reversibility and functional diversity, and the design concept has given rise to various high-performance and sustainable material systems.

**4.2.1 Epoxidized soybean oil.** Capitalizing on vanillin's role in providing imine bonds, researchers have extensively utilized ESO to develop sustainable vitrimers. Wang *et al.*<sup>37</sup> used a vanillin-based dynamic curing agent and 1,10-fatty diamine to cure ESO, creating a fully bio-based dynamic cross-linked vitrimer. They innovatively grew MWCNTs on the surface of the vitrimer to prepare a superhydrophobic, photothermally active coating *via* the solvent-induced self-formation of micro-nano structures. The best superhydrophobic coating (VEV-CNTs-3) had a contact angle of 152.1° and a sliding angle of 9°. This coating exhibited excellent anti-icing and deicing properties (the ice delay time reached 622 s, and the ice adhesion strength was only 11.2 kPa), and the internal dynamic imine bonds imparted the coating with closed-loop recycling ability. Similarly, focusing on ESO-vanillin systems but exploring structural control, Li *et al.*<sup>38</sup> synthesized dynamic crosslinking materials using ESO, vanillin, and different structural diamines (aromatic/alkaline) through a melt reaction (Fig. 7). The mechanical properties of the materials (high strength, low ductility, and high flexibility) were controlled by precisely selecting the type of diamine. Thanks to the dynamic imine bond, the vitrimers exhibited excellent reconfigurability, thermal stability (the initial decomposition temperature exceeded 200 °C), adhesion performance, and outstanding recyclability (with performance even improving after recycling), laying the foundation for the development of a high-performance sustainable adhesive.

By further refining the curing chemistry, Zhao *et al.*<sup>39</sup> prepared the vanillin-based Schiff base (VSB) through the reaction of vanillin with 4,4'-methylene diphenylamine, and applied it to cure ESO and synthesize sustainable epoxy vitrimers. The phenolic hydroxyl groups in VSB significantly enhanced its reactivity with the epoxy groups of ESO. The mechanical properties of the vitrimers could be easily regulated (ranging from soft to hard) by adjusting the feeding ratio of ESO and VSB. Their initial decomposition temperatures were about 400 °C, conveniently facilitating the on-demand design of

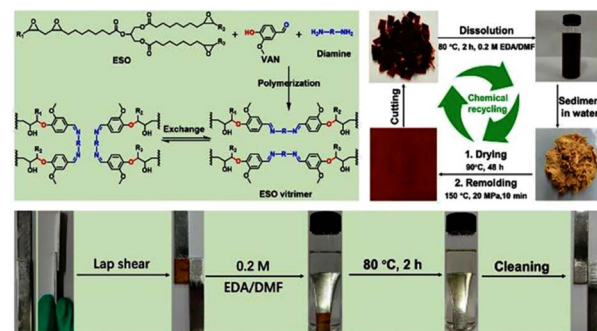


Fig. 7 Synthesis of the ESO vitrimer through a one-pot solvent-free approach. Reproduced with permission.<sup>38</sup> Copyright 2024, ACS Publications.



material properties. To balance the sustainability of bio-based materials with the demand for high performance, Veloso-Fernandez *et al.*<sup>40</sup> also synthesized a bio-based vitrimer (VESOV) using ESO and vanillin-derived Schiff bases (VSB) and commercial epoxy resin (ER). The study showed that the  $T_g$  and storage modulus were significantly improved as the ER content increased. For example, the  $T_g$  of VESOV was 28.7 °C, which increased to 48.2 °C after adding 30 wt% ER. At 30 wt% ER, the recyclability and mechanical properties of the material achieved an optimal balance, and the mechanism for achieving recyclability through dynamic Schiff base exchange reaction was revealed.

To expand the functionality towards flexible applications, Safarpour *et al.*<sup>41</sup> used modified ESO, vanillin, bio-based diamine, and oleic acid to synthesize flexible and stretchable vitrimers based on green chemistry principles, and the mechanical and processing properties could be adjusted by precisely controlling the amount of oleic acid. These vitrimers have excellent reprocessability, recyclability, oxygen and water vapor barrier properties, comparable to low-density polyethylene, and have extremely low migration in food simulants (much lower than the limit of 10 mg dm<sup>-2</sup> stipulated by the European Union), thus meeting food packaging requirements. More importantly, the materials are biodegradable in seawater and are expected to solve the problem of plastic marine pollution.

**4.2.2 Castor oil.** In terms of enhancing the performance and sustainability of traditional epoxy resins, Sankar Lal *et al.*<sup>42</sup> mixed CO-derived bio-resin (EMR) with epoxy resin and a vanillin-based Schiff base curing agent to construct a dynamic imine bond crosslinking system. The addition of EMR reduced the relaxation time of the epoxy resin, improved the efficiency of dynamic bond exchange, and significantly enhanced the impact strength of the epoxy resin (the impact strength of the epoxy vitrimer with 20% EMR increased by 37.8%). Moreover, when further combined with flax fibers, the resulting green composite material exhibited excellent interfacial adhesion and chemical recyclability. Subsequently, Shao *et al.*<sup>43</sup> designed a dual dynamic network structure of ester-imide bonds and aromatic Schiff base crosslinkers using ricin oil-derived epoxy resin, and synthesized ricin oil-based epoxy resin (ASB-ECO), shown as Fig. 8. This material had a self-healing rate of up to 100% under visible light, a low dielectric constant ( $D_k = 5.54$ ), and low dielectric loss ( $D_l = 0.025$ ), with the potential for application in the field of electronic materials.

CO, cysteamine, and vanillin were used to fabricate a fully biobased polyimide trimer by Li *et al.*<sup>44</sup> via a Schiff base cross-linking reaction. This material can achieve self-healing within 30 minutes at 80 °C and an antibacterial rate of over 90% against *Staphylococcus aureus* and *Escherichia coli*. Moreover, this material can be recycled multiple times through hot pressing, reprocessing, and solvent treatment, with a monomer recovery rate of up to 92.7%, showing the potential of biobased thermosetting materials with both antibacterial functions and sustainability.

To balance the recyclability and mechanical properties for sustainable development and efficient material utilization, Wu

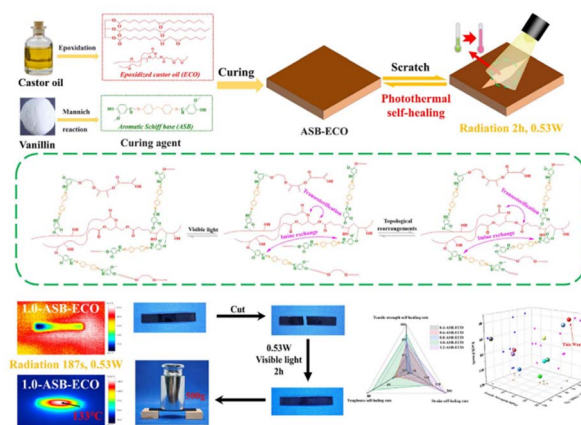


Fig. 8 Schematic of the ASB-ECO synthesis process. Reproduced under the terms of the CC-BY license.<sup>43</sup> Copyright 2025, Yingqing Shao, MDPI.

*et al.*<sup>45</sup> used CO, vanillin-derived diols (VAN-AM), and isophorone diisocyanate (IPDI) to synthesize biobased polyurethane foams containing dynamic imine bonds. By adjusting the ratios of VAN-AM and CO, the density (apparent density as low as 56.6 kg m<sup>-3</sup>), cell structure, and mechanical properties of the foam were regulated. The experimental results showed that BPUFs could achieve efficient recovery through hot pressing at 150 °C for 20 min while maintaining excellent mechanical properties, thus resolving the contradiction between their recyclability and mechanical properties. Furthermore, Xie *et al.*<sup>46</sup> developed a sustainable polyurethane network with adjustable mechanical properties based on CO and vanillin. By varying the VSB content, the mechanical properties can be regulated from elastomer to rigid plastic, with the tensile strength and modulus being significantly increased, and the elongation at break showing a trend of first increasing and then decreasing. For example, the tensile strength and Young's modulus of CPUV-5 reached 52.14 MPa and 1027.47 MPa, respectively, which were 73 times and 347 times that of unmodified PU, respectively. Moreover, this material exhibited good degradability under acidic conditions, further enhancing its sustainability. Sun *et al.*<sup>47</sup> used CO, vanillin, and *m*-xylylenediamine to synthesize the vanillin diol monomer through dynamic imine coupling, and then prepared bio-based glassy polyurethane. Thanks to the dynamic imine bond, the material displayed reconfigurability, weldability, good antibacterial ability (antibacterial rate of 87%), and degradability (within 1 h at 60 °C). Moreover, the material still retained 86% of its original tensile strength even after three crushing/molding cycles, providing inspiration for the development of sustainable polyurethane thermosetting materials. Similarly, Akdogan *et al.*<sup>48</sup> developed a dual-crosslinked bio-based polyurethane by incorporating CO, IPDI, 2,2'-dithiodianiline, as well as disulfide and imine bonds. The dual-crosslinked polyurethane achieved balanced performance, exhibiting moderate strength (43.5 MPa), high gel content, and excellent acid degradability. It retained good mechanical properties (13.8 MPa) after reprocessing at 155 °C, confirming that combining dual dynamic



bonds can optimize the trade-off between mechanical strength and recyclability in bio-based thermoset materials.

**4.2.3 Tung oil.** Jia *et al.*<sup>49</sup> synthesized the trimer monomer of vanillin methacrylate based on TO and vanillin, and prepared a bio-based Schiff base glassy polymer as shown in Fig. 9. This material achieved self-healing when heated at 120 °C for 120 min, and was gradually degraded through ester bond hydrolysis under alkaline conditions. Moreover, combined with graphene and graphene oxide, the material displayed the functions of electrothermal-triggered self-healing, temperature sensing, and fire warning, with the potential for application in fireproof building materials, intelligent sensors, electronic skin, and soft robots.

### 4.3 Disulfide bonds

The disulfide bond is typically formed by oxidizing two thiol groups (–SH), and can be cleaved into two mercapto groups through reduction reactions.<sup>50</sup> The oxidation reaction of mercapto groups leads to a compact structure, while the reduction reaction of the disulfide bond results in a looser structure. Therefore, by introducing polymers containing disulfide bonds, the materials can undergo structural changes according to the oxidation–reduction changes, and the breakage and reformation of the disulfide bonds can promote the self-healing of the damaged material.<sup>51</sup>

By leveraging the dynamic potential of disulfide bonds, researchers have developed innovative CO-based materials. Zhou *et al.*<sup>52</sup> synthesized a polyurethane acrylate resin (COPUA-SS) containing dynamic disulfide bonds using the “one-pot solvent-free” method, and subsequently prepared a photopolymer material suitable for UV-curing 3D printing. The tensile strength of this material reached 70.4 MPa, and it also had a high bio-based content (40.3%). Moreover, its 100% self-healing efficiency and the ability to be recycled multiple times significantly expanded the application scope of bio-based materials in precision 3D printing (such as the manufacturing

of artworks and architectural models). To address the mechanical property limitation, Chen *et al.*<sup>53</sup> prepared polyurethane (PUU) network materials by using CO and 4-aminophenyl disulfide (APD). As the APD content increased, the tensile strength, Young's modulus, and elongation at break were improved, and the performance of COPUU-3 had improved most significantly. Moreover, the material can be restored to nearly 100% of its original mechanical properties under hot pressing at 180 °C, confirming the contribution of the dynamic disulfide bonds to the re-manufacturability.

Two dynamic curing agents (amino phenyl disulfide and amino phenyl diisocyanate) were utilized by Zhou *et al.* to cure CO,<sup>54</sup> and a polyurethane thermosetting material was synthesized based on dual-locking covalent adaptable networks with programmable degradation and closed-loop recycling properties. The tensile strength of the material was  $42 \pm 2$  MPa, which remained stable under a single-chemical stimulus, degraded under dual stimuli, and could achieve closed-loop recycling by regenerating the dynamic bonds through heating. Moreover, this material also possessed multifunctional characteristics such as UV shielding, self-healing, and long-term stability. Similarly, Wei *et al.*<sup>55</sup> developed polyurethanes with varying disulfide contents using CO, 4,4'-diphenylmethane diisocyanate (MDI), and 4-aminophenyl disulfide, and investigated their thermoreversible behavior under different combinations of disulfide and Diels–Alder bonds. The optimum formula contained 4.22% thioether, which had the shortest stress relaxation time and the largest creep range in the range of 60 to 100 °C, and restored the melt processing performance at 135 °C. This study facilitated the design of recyclable polyurethane with adjustable viscoelastic transition and fast thermal response. Exploiting synergistic dynamic chemistries, Zhang *et al.*<sup>56</sup> used epoxidized methacrylated castor oil (EMCO), itaconic acid (IA), and 4,4'-dithiobisbenzidine (AFD) to construct a dual dynamic bond vitrimer (EMCO–GMA–IA–AFD) through free radical polymerization and epoxy–carboxylic acid, epoxy–amine reactions (Fig. 10). The synergistic mechanism of the disulfide bond and ester bond enabled the material to achieve a self-healing efficiency of 94.6%, and it could be repeatedly processed through hot pressing, providing a new solution for the recycling of high-load components. Kashyap *et al.*<sup>57</sup> developed a fully bio-based CAN film based on acetylated castor oil (ACO) and cysteamine. The obtained CAN films exhibited no performance degradation after 10 cycles of recycling, and their inherent hydrophobicity was successfully applied in the field of metal corrosion prevention, marking a significant breakthrough in the transition of dynamic materials from the laboratory to engineering protection scenarios.

Based on the versatility of ESO and disulfide chemistry, Daugelaite *et al.*<sup>58</sup> used AESO, 2-hydroxy-3-phenoxypropyl acrylate, and diallyl disulfide (DADS) to synthesize a bio-based photopolymer compound through photopolymerization reactions, which could be patterned with high precision using micro-contact printing technology. The study found that the material began to restore its permanent shape within 30 s, recovered half within 60 s, and fully recovered within 90 s, showing good shape memory performance. At the same time,

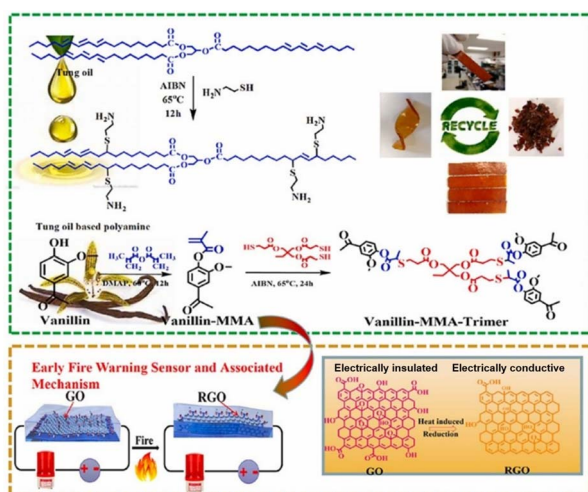


Fig. 9 Synthesis of the monomer of vanillin methyl acrylate trimer. Reproduced with permission.<sup>49</sup> Copyright 2022, Elsevier.



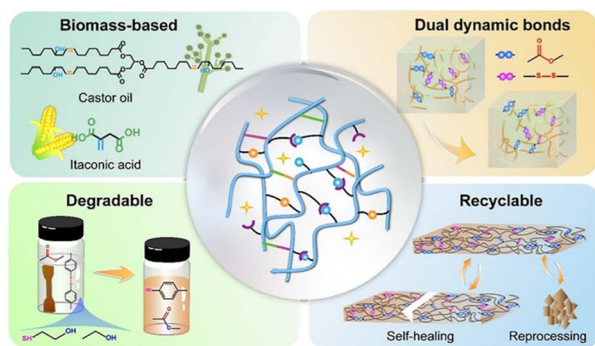


Fig. 10 Schematic of EMCO-GMA-IA-AFD. Reproduced with permission.<sup>56</sup> Copyright 2024, ACS Publications.

the inhibition rates against *Escherichia coli*, *Staphylococcus aureus*, and *Aspergillus* were all over 97%. In addition, an increase in DADS content would reduce the crosslinking density and increase flexibility (an increase of 53%). Shifting focus to composite reinforcement, Ye *et al.*<sup>59</sup> synthesized an epoxy vitrimer (ESO-S) by curing ESO with APD to prepare composites. The introduction of ESO-S not only enhanced the thermal stability and melting point of the composites but also provided a theoretical basis for designing high-performance biobased composites. Shan *et al.*<sup>60</sup> used a dimer aliphatic acid and aromatic disulfide to co-crosslink ESO, thereby preparing a room-temperature self-healing elastomer (the scratches disappeared within 5 min). The tensile strength of the elastomer could still be restored to 91% of the original material after reprocessing three times. Additionally, by simply using vacuum filtration and transfer processes, the carbon nanotube (CNT) layer could be combined with the elastomer to fabricate a highly sensitive and long-term stable strain sensing performance. Zhao *et al.*<sup>61</sup> synthesized a bisphenol derivative containing imine bonds using tyramine and *p*-phenylenediamine, and cured it with APD to prepare an epoxy resin (ETA) with dual dynamic bonds. The ETA-3 exhibited excellent thermal stability ( $T_{d5\%} = 319.17$  °C) and storage modulus (2085.35 MPa), and its short relaxation time ( $\tau = 32.24$  s) and low activation ( $E_a = 70.07$  kJ mol<sup>-1</sup>) enabled the material to repair scratches within 30 min at 100 °C.

Extending the design of multi-dynamic-bond polymers to systems, Xu *et al.*<sup>62</sup> synthesized a bio-based glassy polymer (TOTGE-APD) with two dynamic network structures (hydroxyl ester and disulfide bond) by using TOTGE and APD, and the carbon fiber-reinforced polymer (CFRP) was prepared. The study found that an increase in APD content would enhance the crosslinking density and increase the  $T_g$ . TOTGE-APD also had self-healing functionality, capable of rapidly repairing scratches at 160 °C, and the tensile strength could be restored to 97.5% of the original sample after hot pressing for applications in aerospace and automotive fields.

Complementing previous efforts in vitrimer reprocessing, Di Mauro *et al.*<sup>63</sup> used ELO and aromatic dicarboxylic acid with disulfide bonds to prepare thermosetting resins, and the effects of different initiators on the material properties were

investigated. Among them, when imidazole was used as the initiator, the resin exhibited excellent reprocessability and could be completely chemically recycled at 80 °C in 1 N NaOH within 3 days without the need for additional chemicals. After 10 reprocessing cycles, the  $T_g$  of the material was still 76 °C, and the  $T_{d5\%}$  was equivalent to that of the original material (275 °C). This provides an important reference for the development of sustainable biobased thermosetting resins.

#### 4.4 Carbamate bonds

The formation of carbamates usually involves the reaction between alcohols and isocyanates ( $-NCO$ ), and the bonds exhibit moderate formation and cleavage kinetics under mild conditions, requiring lower temperatures than other DCBs. However, their stability is relatively weak, and in the selection and use of catalysts, factors such as toxicity and cost must be considered. Consequently, how to balance the reversibility and stability of carbamate bonds remains a critical challenge for practical applications.

Research using CO has demonstrated one approach to this challenge. Shi *et al.*<sup>64</sup> used CO, bisphenol, and IPDI to synthesize reprocessable cross-linked polyurethane materials through the cross-linking reaction of dynamic phenol-aminocarboxylate networks. The dynamic bond dissociation temperature and the rate of network rearrangement can be adjusted within a wide range by changing the types and proportions of bisphenol, which has the potential for application in 4D printing, recyclable materials, and intelligent devices.

Parallel research with ESO has explored alternative dynamic chemistries. Yang *et al.*<sup>65</sup> used CO<sub>2</sub> to catalyze the cyclocondensation of ESO for synthesizing cyclic carbonate-functionalized soybean oil (CSBO) and then cross-linked it with a dithiol-containing diamine (APD) and a dithiol-free diamine (4,4'-diaminodiphenyl methane, DDM) to prepare two NIPUs (CSBO-APD and CSBO-DDM). The experimental results showed that CSBO-APD exhibited a faster molecular chain rearrangement ability due to the presence of both dynamic disulfide bonds and carbamate bonds, with a shorter stress relaxation time (3.15 min at 140 °C), lower activation energy (74.22 kJ mol<sup>-1</sup>), and the ability to self-heal at 50 °C for 1 h and recycle at 100 °C for 1 h. In contrast, CSBO-DDM required a higher temperature to achieve self-healing because it only contained amide ester bonds. Liu *et al.*<sup>66</sup> synthesized a fully biobased polyhydroxyurea (PHU) based on natural biomass by reacting MDA, derived from turpentine oil, with CSBO (Fig. 11). This non-isocyanate polyurethane (NIPU) exhibited excellent thermal stability and mechanical properties without using toxic isocyanates. Moreover, the amino groups in MDA can catalyze the dynamic covalent bond exchange reaction between CSBO and amino groups at room temperature, thereby endowing the material with self-healing, recyclability (the recovery rate of mechanical properties of CSBO-MDA was more than 85%), and shape memory functions. By adjusting the ratio of amino groups to cyclic carbonate, the crosslinking density and performance of the material can be optimized, providing a new



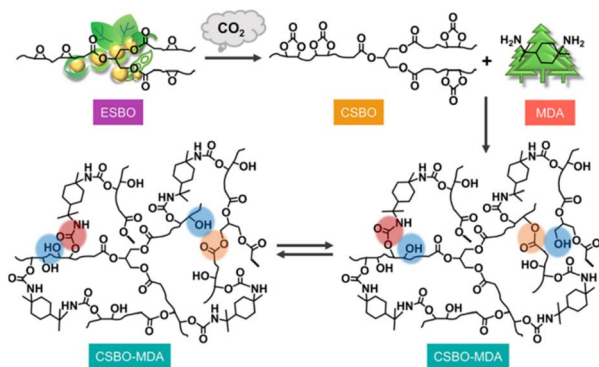


Fig. 11 The formation of the cross-linked CSBO–MDA polymer networks. Reproduced with permission.<sup>66</sup> Copyright 2021, ACS Publications.

idea for the development of sustainable high-performance biobased materials.

PO is a common plant oil with wide applications and significant economic value, which contains a high proportion of saturated fatty acids. Through chemical modification and cross-linking reactions, it can be synthesized into high-performance biobased resin monomers, which can further be used to prepare new environmentally friendly composite materials. Geng *et al.*<sup>67</sup> synthesized a polyurethane acrylate (PUA) prepolymer through the polycondensation reaction between PO-based fatty acid diethanolamide (POEA), IPDI, and hexamethylene diisocyanate (HDI), and a photopolymerized PUA material was prepared. The dynamic exchange reaction of carbamate enabled the material to complete self-healing at 100 °C for 1 h, and the tensile strength was increased to 15.1 MPa after hot pressing (Fig. 12). The material's corrosion inhibition efficiency against Q215 steel reached 99.99%, and it can be applied in fields such as steel corrosion protection coatings, *etc.*

#### 4.5 Borate ester bonds

The borate ester bond is formed by the reversible condensation reaction between boronic acid, or its derivatives, and diols.<sup>68</sup> The formation and exchange reaction of borate ester bonds occur relatively quickly. At room temperature, bond formation can occur within a short time, and the reaction rate is faster than that of most DCBs. The dynamic behavior of borate ester

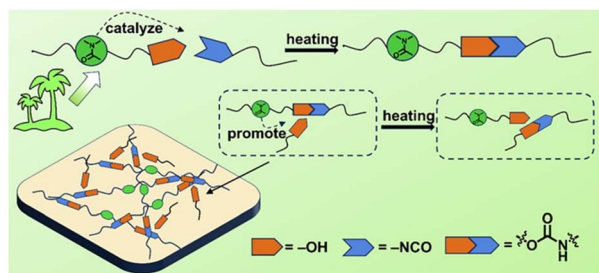


Fig. 12 Schematic of the dynamic exchange reaction of carbamate. Reproduced with permission.<sup>67</sup> Copyright 2024, ACS Publications.

bonds enables various exchange reactions. (1) Hydrolysis/re-esterification. Following the dissociation exchange mechanism, the crosslinking density depends on the balance between hydrolysis and re-esterification. (2) Ester exchange between diols and borate esters. Following the associative exchange mechanism, it is conducive to obtaining reversible crosslinked polymers with high crosslinking density and excellent mechanical properties. (3) Borate ester complexation. Following the same combination-exchange mechanism, dynamic behavior is achieved through direct complexation reactions between different borate ester compounds.

Research on CO demonstrates these dynamics in practical systems. Pan *et al.*<sup>69</sup> used CO, 2,2'-(1,4-phenyl)-bis[4-thio-1,3,2-dioxaborolane] and MDI as starting materials and carried out a one-pot reaction to synthesize a CO-based poly(sulfone-urea) network material. The material possessed a high decomposition temperature over 312 °C, high toughness (maximum > 250%), self-healing ability (the self-healing efficiency of COPU-80 was close to 100% at 120 °C for 60 min), shape memory, and reprocessability properties, which were expected to be applied in fields such as flexible electronic skins. Similarly, Han *et al.*<sup>70</sup> used CO, 1,4-benzene diboronic acid, epoxy resin, and 1-mercaptoglycerol to synthesize elastomer-modified bio-based polyurethane coating material through the borate ester cross-linking reaction. The surface of the modified coating material became smoother, the water contact angle increased, and the controlled release period of nitrogen fertilizer was extended to 126 days. This modification strategy improved the utilization efficiency of fertilizers and reduced environmental pollution, providing new theoretical and technical support for the development of controlled-release fertilizers based on bio-based materials.

Transitioning to ESO, Zych *et al.*<sup>71</sup> utilized the “thiol-epoxy” click reaction to synthesize a bio-based monomer containing dynamic borate ester bonds from AESO, and subsequently prepared a bio-based borate dynamic crosslinking network material (vitriimer) that was self-healable and recyclable. This material exhibited self-healing performance at room temperature, with a repair time of 6 h. Furthermore, this material can be reversibly hydrolyzed in 90% ethanol and then regenerated through solvent evaporation, demonstrating excellent recyclability. Recently, Bodhak *et al.*<sup>72</sup> also utilized the “thiol-epoxy” click reaction to prepare a bio-based acrylic monomer containing dynamic borate ester bonds using ESO and 2-hydroxy acrylate as raw materials, and subsequently synthesized the bio-based trimer (Fig. 13). This material can be reversibly hydrolyzed in 95% ethanol and regenerated through solvent evaporation, demonstrating excellent recyclability. Li *et al.*<sup>73</sup> used ESO, 1,4-phenylenediboronic acid, and 1-thioglycerol to synthesize epoxy resin monomers through the “thiol-epoxy” click reaction, and then prepared a recyclable bio-based epoxy adhesive. This resultant adhesive exhibited excellent bonding strength on various substrates such as glass, wood, and bamboo, and achieved a rearrangement of the network topology structure through the dynamic exchange reaction of borate ester bonds at high temperatures, thus possessing good recyclability and reprocessability.





Fig. 13 Schematic of the synthesis and research of the ESBO–HEA–DBDT biobased trimer. Reproduced with permission.<sup>72</sup> Copyright 2025, Elsevier.

Finally, in ELO systems, Sangaletti *et al.*<sup>74</sup> utilized the “thiol-epoxy” click reaction to develop a borate ester-based polymer monomer based on epoxy flaxseed oil and diboronic acid dithiol (DBEDT), and subsequently prepared recyclable carbon fiber-reinforced composites. The carbon fiber composite materials demonstrated comparable mechanical properties to traditional epoxy resin composite materials, and can be recycled under mild conditions without affecting the properties of the recycled carbon fibers and the resin matrix. This new type of polymer provided a new solution for sustainable and recyclable high-performance composite materials.

#### 4.6 Diels–Alder bonds

The Diels–Alder reaction is a classic [4 + 2] ring addition reaction.<sup>75</sup> In self-healing polymers, reversible dynamic crosslinking *via* the Diels–Alder reaction between furan and maleimide groups is widely studied.<sup>76</sup> Under appropriate conditions (*e.g.*, heating or catalyst effects), the six-membered ring generated by the Diels–Alder reaction can be reversibly decomposed. This dynamic reversibility enables the material to self-heal when subjected to external forces or damage, and it can be dissociated and reorganized through chemical methods to achieve the recyclability of the material.

Starting with ESO, Liu *et al.*<sup>77</sup> introduced a furan group and carried out a Diels–Alder reaction with bis-maleimide, thereby converting ESO into a pre-polymer (ESOF) containing Diels–Alder bonds for further preparing a bio-based adhesive with the capability of rapid reorganization. The adhesive completed self-healing at 140 °C and 30 min, which helped reduce energy consumption and environmental pollution, and showed low viscosity characteristics during the reorganization process, which was conducive to rapid reorganization self-healing. Further advancing ESO applications, Xi *et al.*<sup>78</sup> used poly(1,4-butanediol succinate) diol, soybean oil-based polyol (MESO), and toluene diisocyanate to synthesize a diol monomer, and then prepared a polyurethane adhesive. This adhesive showcased a tensile strength of up to 91.7 MPa, as well as good self-healing ability and thermal recoverability. The introduction of the Diels–Alder bond significantly increased the crosslinking density and mechanical properties, which was of great significance for the development of new environmentally friendly materials.

Expanding to CO, Costa Cornellà *et al.*<sup>79</sup> used three different renewable cyclic acid anhydrides to functionalize CO, which was cross-linked with a furan group to prepare a CO-based self-healing elastomer (Fig. 14). This elastomer had a tensile elongation rate of up to 487% and can act as a soft robot, and the developed soft pneumatic gripper exhibited desirable performances, including self-healing, full recyclability, and degradability, which can reduce the dependence on fossil-based plastics. Wei *et al.*<sup>80</sup> used polypropylene glycol, CO, and MDI to prepare a CO-based polyurethane vitriimer (C-PUV) and its modified asphalt (C-PUVAs), combined with disulfide bonds and Diels–Alder reactions. The initial decomposition temperature of C-PUVAs ranged from 241 °C to 270 °C, demonstrating excellent thermal stability, and their viscosity retention time at 100 °C exceeded 50 min, meeting the construction requirements. This material could be applied in road engineering, especially for steel bridge deck pavement, which could significantly improve the lifespan and maintainability of the road.

Regarding TO, Ma *et al.*<sup>81</sup> synthesized a recyclable and degradable TO-based vitriimer (PMFV) material with dynamic ester bonds and Diels–Alder bonds (Fig. 15). This material indicated excellent mechanical properties and thermal stability. When compounded with MWCNTs and GO, it exhibited excellent joule heating performance (it can reach 150 °C rapidly under 32 V voltage driving) and rapid temperature response capability (able to trigger an alarm within 2 s), and can be used in deicing equipment and fire/temperature warning sensors.

#### 4.7 Sterically hindered urea bonds

The sterically hindered urea bonds (HUBs) are chemical bonds formed by the urea group (–NH–CO–NH–), which can be used to produce polyurea and poly(urea-aminocarbonate), enabling dynamic property changes and self-healing without catalysts at low temperatures. Ying *et al.*<sup>82</sup> first reported dynamic polyurea thermosetting materials based on HUBs, which can weaken the bonding energy of amide bonds and simplify the reverse reaction conditions, thereby enabling the reprocessing behavior of

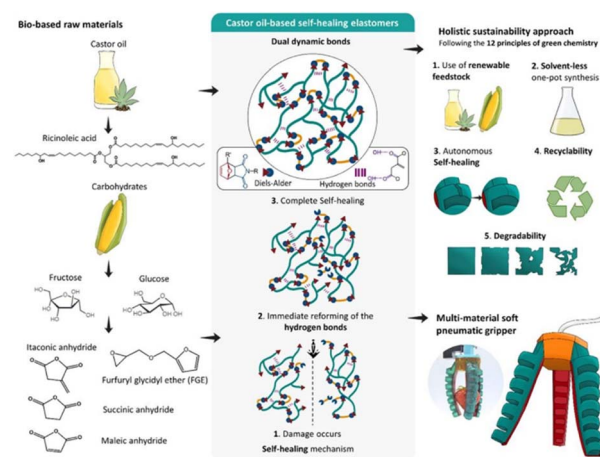


Fig. 14 Schematic of the preparation and research of castor oil-based elastomers. Reproduced with permission.<sup>79</sup> Copyright 2023, ACS Publications.



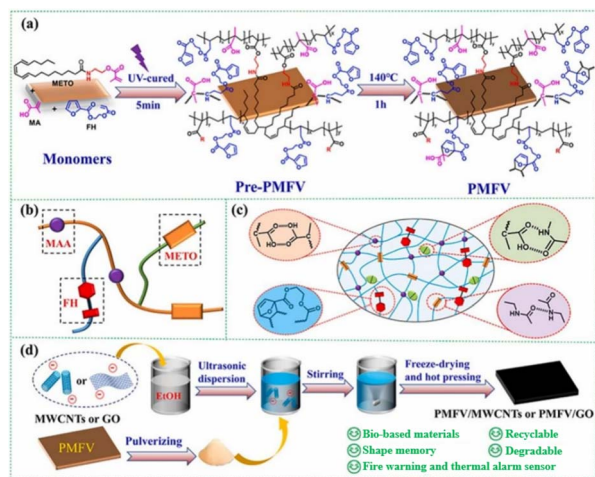


Fig. 15 Synthesis route (a), network structure (b), hydrogen bond network structure and DA bond (c) of PMFV; (d) the fabrication schematic diagram of PMFV/ MWCNTs and PMFV/GO composites. Reproduced with permission.<sup>81</sup> Copyright 2025, Elsevier.

thermosetting materials based on HUBs at room temperature.<sup>83</sup> Zhang *et al.*<sup>84</sup> used CO, IPDI, and polyethylene glycol to develop a recyclable, repairable, removable, and hydrophobic UV-curable polyurethane coating with dynamic HUBs. This coating featured a high bio-based content (45.1–47.6%), excellent thermal, mechanical, and coating properties, and hydrophobicity (the water contact angle was more than 90.8°). Moreover, due to the rapid dissociation or exchange of HUBs and intramolecular hydrogen bonds, the selected sample (PU-Si2) showed a scratch repair efficiency of up to 97.0% within 60 min at 140 °C and a welding efficiency of 91.3% within

60 min at 120 °C. Subsequently, Zhang *et al.*<sup>85</sup> studied a new bio-based polyurethane material based on CO, achieving self-healing, shape memory, and recyclability through HUBs and hydrogen bonds. This material could be recycled at least 4 times without significantly sacrificing tensile properties, and exhibited good shape memory behavior, with a shape fixation rate of over 88.4% and a shape recovery rate of over 81.3%. By adjusting the composition of HUBs, recyclable and moldable conductive composite materials can also be achieved. This work provides a simple and universal method for developing green and high-performance sustainable polyurethane materials. Zhu *et al.*<sup>86</sup> synthesized a recyclable and reusable biobased photopolymer (COIT) suitable for DLP 3D printing using CO, IPDI, and 2-(*tert*-butylamino)ethyl methacrylate (Fig. 16). The material exhibited rapid recyclability and reusability, and could be fully recovered within 4 h at 90 °C or 2 h at 100 °C, and the recovered resin had similar physical and chemical properties and printing resolution to the original resin. The excellent performances enabled the COIT to be used in complex model casting, information encryption/anti-counterfeiting, and other fields.

## 5. Conclusion and prospects

As a new type of polymer material, plant oil-based polymeric materials, DCPs, with DCBs were imparted with unique properties, such as self-healing, stimulus responsiveness, and recyclability, while maintaining excellent mechanical and thermal properties. This article reviews the synthesis methods, structural features, and application prospects of this class of polymers, highlighting their significant position in the fields of chemistry and materials science. However, despite significant progress, several critical challenges must be addressed to fully realize the potential of these materials. Moving forward, research should focus on the following strategic directions: (1) the optimization of reaction conditions. Many DCB exchange reactions still require relatively harsh conditions, and future work should prioritize the development of milder, catalyst-free systems and the exploration of alternative external triggers such as light, humidity, or microwave activation. (2) The improvement of stability and functionality. The long-term stability of DCPs under operational conditions (*e.g.*, thermal, oxidative, hydrolytic) remains a concern, particularly for applications in harsh environments. Strategies such as hierarchical network design, dual/multi-dynamic bond systems, and the incorporation of stabilizing nano-fillers can be employed to decouple dynamicity from degradation. (3) The in-depth exploration of the relationship between material properties and structure. Computational modeling and advanced feature technology should be used, integrating with the rational design of molecular structure, to dominate the macro properties and finally achieve tailor-made performance. (4) Expansion into high-value and emerging applications. Future opportunities lie in high-performance fields, including flexible electronic devices (self-healing circuits, stretchable sensors), biomedical materials (degradable implants, drug-eluting substrates), smart textiles, and energy storage devices. In conclusion, plant oil-based DCPs address existing challenges through interdisciplinary research.

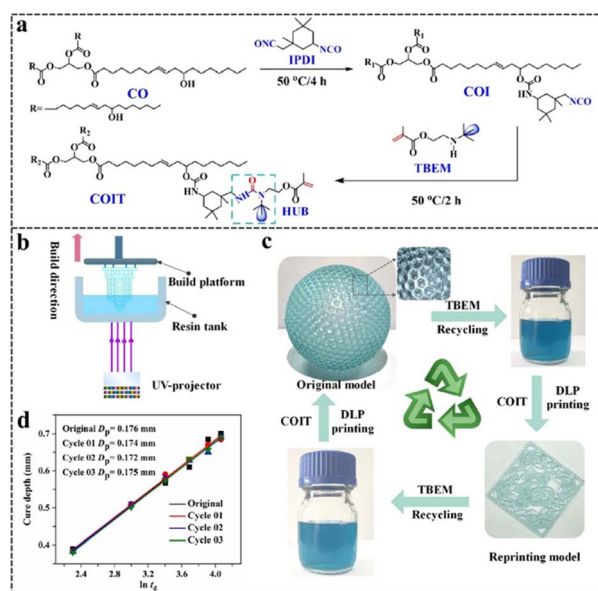


Fig. 16 (a) Synthesis of COIT oligomer; (b) schematic illustration of DLP printing; (c) recyclable DLP printing with COIT-T30; (d) working curves of original and recycled resins. Reproduced with permission.<sup>86</sup> Copyright 2023, Elsevier.



Their sustainable development will reduce dependence on fossil resources and also provide a new paradigm for material design, in which adaptability, functionality, and environmental harmony are intrinsically linked.

## Author contributions

Xiaohui Huang: writing the original draft, investigation, and formal analysis. Yanlin Li: conceptualization, supervision. Jing Liu: investigation and formal analysis. Hang Zhou: conceptualization and investigation. Kashif Khan: writing-review & editing. Yi Tan: supervision and writing-review & editing. Chengguo Liu: conceptualization, supervision, resources, funding acquisition, and project administration.

## Conflicts of interest

The authors declare no potential conflict of interest.

## Data availability

This is a review article. All the data reported herein can be found in the cited papers in the Reference section.

Supplementary information (SI): a list of chemical abbreviations. See DOI: <https://doi.org/10.1039/d5su00718f>.

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