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Recent advances and challenges in the mechanical and chemical recycling of vitrimers and fibre-reinforced vitrimer composites: A review

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ABSTRACT

Vitrimers, a class of covalent adaptable networks, have emerged as a promising alternative to traditional thermoset and thermoplastic polymers, owing to their unique reprocessability and recyclability. This review provides a comprehensive overview of mechanical and chemical recycling methodologies for vitrimers and their fibre-reinforced composites, highlighting recent advancements and innovations in the field. Firstly, the review classifies vitrimers based on their structure and dynamic covalent mechanisms and summarises their main properties. Subsequently, it gives an overview of the conditions and outcomes of both mechanical and chemical recycling processes for vitrimers. Then, after summarising the processing methods of vitrimer composites, it explores mechanical composite recycling techniques, which, while straightforward, often lead to diminished mechanical properties due to fibre length reduction. In contrast, chemical composite recycling methods demonstrate superior potential for preserving fibre integrity and recovering original monomers for closed-loop synthesis. Key studies showcasing successful recycling processes are discussed, underscoring the importance of developing closed-loop systems to enhance sustainability in material applications. Ultimately, this review aims to highlight the critical advancements in recycling strategies for vitrimers, positioning them as a vital component of sustainable composite material development.

1. Introduction

The use of thermoset resins and their respective composites is increasing year by year due to their outstanding mechanical performance, low density, dimensional stability, and chemical resistance. These properties made them essential materials for a wide range of industrial sectors and an integral part of everyday life, from transportation to wind turbines. The permanent crosslinking in thermoset composites hinders features such as self-healing, reshaping, and mechanical recycling by melting, which would be crucial to offering a sustainable approach [1]. During the last two decades, alternative materials have been designed to address the shortcomings that traditional thermosets face regarding their reuse and recycling. These materials incorporate reversible covalent bonds into the backbone structure or the crosslinks of the network to promote adaptability [2]. Reversible covalent bonds are covalent bonds that can reversibly break and reform under equilibrium conditions in response to external stimuli, serving as the core mechanism in dynamic covalent chemistry. Covalent adaptable networks (CANs) are defined as networks that contain a sufficient number and topology of reversible covalent bonds to enable the crosslinked network structure to respond chemically to an applied stimulus [3]. These dynamic networks can be categorised by their bond exchange mechanism, which can be associative and dissociative. In dissociative CANs, the bond-breaking precedes the reformation of the bond; therefore, when the bond breaks, the viscosity drops suddenly, and the material cannot maintain its dimensional stability. Typical examples of such CANs are polymers based on reversible Diels–Alder reaction between furans and maleimides [4–6]. On the other hand, in associative

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CANs, the new bond formation precedes bond breaking; thus, the crosslink density is unchanged during the reversible bond exchange. As a result, these materials remain intact and do not depolymerise [3,7].

This review focuses on associative CAN-based materials known as vitrimers and their recyclability. These materials behave as thermosets below their vitrimer transition temperature (T_v), while above the T_v, the increasing rate of the associative exchange reactions of permanent but dynamic crosslinks enable the vitrimers to flow, facilitating their reshaping and recycling. This review summarises the possible solutions of mechanical and chemical recycling in the case of vitrimer resins and their fibre-reinforced composites.

Although vitrimers show immense promise as recyclable alternatives to traditional thermosets, their broader industrial uptake remains limited. As discussed by Maes et al. [8] key challenges include maintaining performance while enabling reprocessing, ensuring compatibility with existing industrial workflows, and achieving cost-effective, scalable solutions. They also pointed out that while the chemical toolbox of dynamic covalent chemistry is expanding rapidly, few systems have yet demonstrated clear pathways to market, and closer alignment between fundamental research and industrial needs is urgently required. In this context, the present review contributes to closing this gap by providing a systematic overview of the current state of mechanical and chemical recycling strategies for vitrimers and their composites, with particular attention to practical aspects such as fibre recovery, matrix reusability, and recyclate performance. By identifying both the scientific advancements and the practical bottlenecks associated with vitrimer recycling, this article aims to support the development of more robust, scalable, and industrially relevant recycling solutions and thereby promote the broader uptake of vitrimer-based technologies in the circular economy.

2. Vitrimer resins

In this section, vitrimer resins are classified according to their structure and dynamic covalent mechanisms, and their main properties are summarised, followed by the possible solutions for their mechanical and chemical recycling.

2.1. *Classification and properties of vitrimer resins*

Vitrimers can be classified into several groups based on their respective bond exchange mechanism. The most researched dynamic covalent mechanisms include transesterification [9–34], imine exchange [35–56], disulfide metathesis [57–62], olefin metathesis [63,64], transalkylation exchange [65,66] and boronic ester exchange [67,68], transamination of vinylogous urethanes [69–72], diketoenamine bonds [73], transcarbamoylation [74–76], trithiocarbonate exchange [77], silyl ether metathesis [78], thiol-Michael addition [79], alkoxyamines [80,81], and carbonate exchange [82]. The academic literature on vitrimers pays considerably more attention to transesterification, disulfide exchange, and imine exchange mechanisms due to the ease of network exchanges in diverse environmental conditions, such as moderate (80 °C–160 °C [25,38]) to elevated temperatures (above 200 °C [9]), the presence of mild or no catalysts [17,27], or ambient atmosphere (without inert gas needed). Due to their straightforward synthesis and generally favourable thermal and mechanical properties, these systems are often preferred over vitrimers based on less established exchange chemistries. The reviewed vitrimer resin systems, classified on the basis of their dynamic mechanism, are listed in Table 1 along with their polymer components and main properties.

2.2. *Recycling of vitrimer resins*

Vitrimers can be recycled via mechanical and chemical methods. Mechanical recycling methods are usually based on size reduction and then reprocessing (melting and reshaping) the resin parts. In contrast, chemical recycling methods focus on dissolving the polymer resin with a suitable solvent or solvent mixture. Mechanical recycling aims at reprocessing the material without altering its chemical composition, whereas chemical recycling focuses on recovering the monomers for reuse in the synthesis of vitrimers. In the following sections, mechanical and chemical recycling methods are summarised. In this section, solutions dealing mainly with the recycling of the vitrimer matrix itself are summarised, studies focusing on the properties of the recycled fibres are discussed in section 3.

Key papers dealing with the recyclability of vitrimers are summarised in Table 2.

2.2.1. *Mechanical recycling*

Mechanical methods are generally the easiest way to recycle vitrimer resins. These recycling methods usually consist of two main steps. First, the vitrimer is mechanically machined into smaller particles by chip removal technologies such as milling, shredding or grinding [29,57] or mechanically broken [17]. The resultant characterising particle size varies in a wide range from 25 μm up to 8 mm, depending mainly on the applied machining technology (whether the tool is abrasive or has defined geometry, clearance angle, cutting edge radius, etc.) and process parameters (feed, cutting speed, depth of cut, pressure, etc.). Second, these particles are hot-pressed in a mould to reshape. The general process of mechanical recycling of vitrimer resins is depicted in Fig. 1. The benefit of these mechanical recycling methods lies in the absence of chemical reactions and the elimination of solvent treatments, rendering these techniques environmentally favourable. In addition, the industrial implementation of such mechanical recycling technologies faces only moderate limitations, mainly due to the high technology readiness levels (TRL) of mechanical machining technologies. However, following multiple cycles, polymer chains may undergo breakage, resulting in lower crosslink density and compromised mechanical attributes.

Considering the characteristics of the technologies applied in the two main steps of mechanical recycling, the mechanical properties of reprocessed (recycled) vitrimers significantly depend on the following parameters: particle size and its distribution of machined or broken vitrimer [34], moisture content of particles [38], hot-pressing temperature [34], hot-pressing time [34], hot-pressing pressure [30], number of recycling steps [49]. Generally, the smaller the particle size, the better the surface quality; the higher the hot-pressing temperature and the longer the hot-pressing time, the increased the interfacial chain density and modulus [34]; the larger the hot-pressing pressure, the shorter the reprocessing time of vitrimers [30,31]; (iv) the longer the recycling steps, the worst the resultant tensile modulus [34,38].

Taynton et al. [38] analysed the mechanical strength and elastic modulus of mechanically recycled polyimine vitrimer. First, prepared polyimine vitrimer films from terephthaldehyde, diethylene triamine, and triethylene tetramine in ethanol solvent. After the evaporation of the solvent, the polyimine film was cured in an oven in three stages (30 min at 75 °C, then 30 min at 85 °C, and 30 min at 105 °C). They also prepared polyimine powder by predissolving the monomers in ethyl acetate and subsequent reaction at 65 °C. Then, they hot-pressed polymer disks from these polyimine powders under a pressure of 90 kPa for 45 min at 80 °C. They ground the disks to powder using sandpaper and hot-pressed them again. This recycling process was repeated three times. They tested the specimens' mechanical properties after each hot-press stage and found no significant differences in the resulting strength; only the elastic modulus decreased slightly. This experimental work demonstrates that the mechanical strength of the vitrimer did not deteriorate significantly after multiple recycling cycles, indicating that the dynamic covalent bonds formed through the imine-exchange reaction effectively reformed an extensive crosslinked network following mechanical disruption. However, the slight decrease in elastic modulus (indicating a softer material) may suggest contamination during the grinding and hot-pressing processes, particularly from moisture in the air. The negative effects of water on rigidity and strength were also

Table 1
Classification of vitrimer resins by their dynamic mechanism with a summary of polymer components and main properties.

Dynamic mechanism	Polymer components	Properties	Reference
transesterification (TRE)	diglycidyl ether of bisphenol A (DGEBA) + sebacic acid	T_g =40–45 °C	2016 Yang [9]
	DGEBA + fatty acid mix	$T_g \approx$ 30 °C	2016 Yu [13]
	DGEBA (E44) + methylhexahydrophthalic anhydride	4 type, T_g =75–140 °C	2018 Zhu [14]
	DGEBA (E51) + methyl tetrahydrophthalic anhydride	–	2018 Yang [15]
	bisphenol A-based epoxy (DER331) + 2-ethyl-2-(hydroxymethyl)-1,3-propanediol (TMP); cured with succinic anhydride (SA) at a fixed mixing ratio (epo:anh = 2:1) = HBE (hyperbranched epoxy)	T_g =70–96 °C	2018 Han [16]
	DGEBA + blend of citric and sebacic acids	in situ-generated tertiary amines can be used as efficient catalysts of TRE. T_g =70 °C	2018 Altuna [10] 2019 Altuna [11]
	DGEBA + 4,4′- dithiodibutyric acid	T_v =31 °C (DSM); 71 °C (TRE);-6 °C (dual) T_g =35 °C (DSM); 50 °C (TRE); 30 °C (dual)	2019 Chen [17]
	epoxy prepolymer from catechol incorporated bisphenol + di-/tri acid carboxylic acid hardener	synthesis of a property-tunable epoxy prepolymer T_g =10–20 °C	2019 Zhao [18]
	various epoxidized vegetable oils (EVOs) 2,2′-dithiodibenzoic acid	T_g =17–91 °C	2020 Di Mauro [19]
	triethanolamine as a co-curing agent into a bisphenol A-based epoxy (DER331) + anhydride network	T_g =135 °C	2020 Hao [20]
	DGEBA, HBE + glycerol/succinic anhydride/glutaric anhydride/dimeric acid/glycidylamine.	T_g =70–110 °C	2020 Liu [12]
	tung oil-based triglycidyl ester + methane diamine	T_g =61–83 °C, depending on the mixing ratio	2021 Xu [21]
	DER 331 + ethylenediamine and glutaric anhydride = vitrimer with tertiary amines	n.a.	2021 Liu [22] 2019 Liu [23]
	DER331 + glutaric anhydride + glycerol		
	2-hydroxy-2-phenoxypropyl acrylate + acrylated epoxidized soybean oil	T_g =0–53 °C depending on the mixing ratio; $T_v \approx$ 72 °C	2023 Grauzeliene [24]
	DGEBA + bio-based 2,5-furandicarboxylic acid	T_g =92–147 °C depending on the mixing ratio and measurement method (DSC/DMA); T_v =65–150 °C depending on the mixing ratio	2023 Manarin [25]
	poly(butylene terephthalate) + DGEBA and aluminum diethylphosphinate as flame retardant	T_g =40–47 °C depending on the mixing ratio; UL-94 V-0 classification	2024 Meunier [26]
	bisphenol A-based epoxy + methylhexahydrophtalic anhydride and 2-(bis(2-hydroxyethyl)amino)ethyl diphanylphosphinate as flame retardant and TRE modifier	T_g =88–102 °C; LOI ≈33 V/V%; UL-94 V-0 rating	2023 Ren [27]
	DGEBA + fatty acids and glutaric anhydride	T_g =28–78 °C; tunable according to the components	2019 Zhang [34]
	lignocellulose-based diglycidyl ether of alkyl diphenolate (DGEDP) + hexahydro-4-methylphtalic anhydride (MHHPA)	T_g =110–140 °C	2021 Yue [29]
	DGEBA + fatty acids + zinc acetate catalyst	T_g =23–27 °C depending on the particle size of the grinded samples	2020 Li [30]
	DGEBA + glutaric anhydride + zinc acetylacetonate catalyst	T_g ~80 °C	2020 Zhang [31]
	DGEBA + citric acid (CA) + cashew nut shell liquid (CNSL)	T_g =24–32 °C	2021 Lorwanishpaisarn [32]
	DGEBA + trimer fatty acid	n.a.	2019 Yu [33]
imine exchange	terephthaldehyde + tris(2-aminoethyl) amine (TREN) + diethylene triamine (DETA), ethylene diamine (ED), tetraethylene tetramine (TETA)	T_g =18–145 °C depending on CL length	2014 Taynton [38] 2016 Taynton [39] 2018 Kissounko [40]
	imine-embedded bisphenol (vanillin + aminophenol) reacted with epichlorohydrin to obtain a glycidyl ether (GE-VAN-AP) + polyfunctional amine	T_g =71 °C	2018 Zhao [41]
	DER331 + 4,4′ -methylenebiscyclohexanamine and 3-methoxy-4-(oxiran-2-ylmethoxy) benzaldehyde (MB) + 4,4′-methylenebiscyclohexanamine	T_g =172 °C	2019 Wang [42]
	fructose based 2,5-furandicarboxaldehyde + Priamine 1071	T_g =−10 °C; T_v =−60 °C	2019 Dhers [43]
	60 % trifunctional epoxy resin (AFG-90H) + 40 % ethylene glycol diglycidyl ether (EGDGE) + ICH (imine containing hardener; lignin-derived vanillin + methylcyclohexanediamine)	T_g =130 °C	2020 Memon [44]
	epoxidized soybean oil + vanillin + aminophenol	T_g =28 °C	2020 Liu [45]
	vanillin-based epoxy vinyl ester resin with vinyl and imine bonds (methacrylated vanillin with diamine and triamine)	photocurable; T_g =56–80 °C	2020 Xu [35]
	vanillin + hexachloro-cyclotriphosphazene + polyetherimine	T_g =98 °C; LOI= 28.6 V/V%; V-0 self-extinguishing	2021 Liu [36]
	terephtaldehyde + diethylene triamine + tris (2-aminoethyl) amine	T_g ~75 °C; T_v > 80 °C	2021 He [52]
	vanillin-based epoxy + 4,4′-diaminodiphenylmethane (DDM) or DETA or isophoronediamine (IPDA) or polyetheramine	T_g =58–143 °C, depending on the type of the curing agent	2022 Wang [37]
	glycerol triglycidyl ether + ICH (imine containing hardener; vanillin + 4-aminophenol)	T_g =70 °C	2021 Liu [46]
	DGEBA + ICH (imine containing hardener; (m-xylylene-diamine + vanillin) or (1,6-hexane-diamine + vanillin) + 4-aminophenol	T_g =82–83 °C (based on 1,6-hexane-diamine) T_g =90–96 °C (based on m-xylylene-diamine)	2021 Liu [47]
	epoxidized menthane diamine + adipic acid	T_g =72–86 °C depending on the mixing ratio	2022 Xu [48]
	4,4-diaminophenylmethane + p-hydroxybenzaldehyde, phenol and terephtalaldehyde	$T_g \approx$ 146 °C; LOI ≈40 V/V%; UL-94 V-0 level	2022 Zhang [49]
	epoxy precursors vanillin bearing acetal, aldehyde, and oxirane functionalities) + diamines	T_g =36–63 °C	2023 Türel [50]

(continued on next page)

Table 1 (*continued*)

Dynamic mechanism	Polymer components	Properties	Reference
	divanillin acetal monomer + TREN or cyclic (IPDA or 4,4′-methylenebis (cyclohexylamine)) or diamine curing agents	T_g =0.5–42 °C	2023 Saito [53]
	biobased di-furfural monomer polymerized with various biobased multifunctional amines	T_g =8–60 °C	2024 Türel [51]
	terephthaldehyde + DETA + TREN	T_g ~60 °C; T_v ~130 °C	2021 Yu [54]
	terephthaldehyde + tris(2-aminoethyl) amine (TREN) + diethylene triamine (DETA), ethylene diamine (ED), tetraethylene tetramine (TETA); ammonium polyphosphate (APP), resorcinol bis(diphenyl phosphate) (RDP) and N,N′,N''- tris(2-aminoethyl)-phosphoric acid triamide (TEDAP) as flame retardant additives	T_g =80–136 °C, depending on the FR additive; LOI= 21–29 V/V%; above 3 % phosphorus content: V-0 self-extinguishing class.	2024 Toldy [55] 2024 Pomázi [56]
disulfide exchange	DGEBA + diethyltoluenediamine + 4-aminophenyl disulfide	T_g =130 °C	2016 Ruiz de Luzuriaga [57]
	DGEBA + aminophenyl disulfide + DOPO	T_g =120–130 °C	2021 Li [58]
	poly(lactide-co-caprolactone) (PCLA), hexamethylene diisocyanate (HDI), polytetrahydrofuran (PTMEG) + 4,4′-aminophenyl disulfide	T_g =30–40 °C, T_v =60–80 °C	2019 Jia [59]
	diglycidyl ether of vanillyl alcohol and phloroglucinol triepoxy + 4-aminophenyl disulfide	T_g =105–195 °C	2020 Genua [61]
	DGEBA + bis(4-hydrophenyl) disuplhide + 4-aminophenyl disulphide	T_g =147 °C; T_v =171 °C	2020 Si [60]
	DGEBA + hexamethylene diisocyanate + 4-aminophenyl disulfide	T_g =88–140 °C, depending on the ratio of the dynamic epoxy groups	2022 Zhou [62]
olefin metathesis	cross-linked polybutadiene with Grubbs' Ru catalyst	n.a.	2012 Lu [63] 2012 Lu [64]
transalkylation (TAM)	α-azide-ω-alkyne + 1,6-dibromohexane	n.a.	2015 Obadia [66]
	partial alkylation of the thioether functions, resulting in poly(thioether-sulfonium salt) - 1,2,3-triazolium bromide	n.a.	2017 Hendriks [65]
boronic ester exchange	1,2-diol- containing polycyclooctene + diphenylboronic ester and di(o-aminophenylboronic) ester	n.a.	2015 Cromwell [67]
	poly(hexyl methacrylate-random-2,3-dioxaborolanepropyl methacrylate) + 2,2′-(1,4-phenylene)-bis[4-methyl- 1,3,2-dioxaborolane]	T_g ≈ 0 °C	2020 Wu [68]
vinylogous urethane	pripol 2033 + Priamine 1074 (Croda) + TREN	low T_g prepolymers (≈-25 °C) T_g of the cured resin ≈80 °C	2017 Denissen [69]
	1,4-butanediol + isophorone diisocyanate + cystine and vanillin	T_g =50 °C T_v =160–180 °C	2019 Lee [70]
	1,12-dodecanebisacetoacetate + 1,5- diaminoanthracene + 1,12-dodecanediamine = vitrimer bonding with both photodimerizable and thermally exchangeable functionalities	T_g =-15-30 °C; T_v =35–80 °C	2019 Wright [71]
	DGEBA epoxy resin + vinylogous urethane curing agents (water-free and one-pot procedure)	T_g ~75 °C	2020 Speisschaert [72]
diketoneamine	three triketone dimers (6,8,10) + TREN	T_g > 120 °C	2019 Christensen [73]
transcarbamoylation (TCARB)	poly(ethyleneglycol)diol + glycerol + hexamethylene diisocyanate	n.a.	2016 Zheng [76]
	hexamethylene diisocyanate + dioxime and trioxime	n.a.	2017 Liu [74]
	carbonated soybean oil and sorbitol ether carbonate + diethylene glycol bis (3-amino-propyl) ether + Priamine (1074, biobased)	T_g =−20 to −10 °C	2019 Hu [75]
trithiocarbonate	n-butyl acrylate (BA) + trithiocarbonate = poly(n-butyl acrylate) (PBA)	T_g =−50 °C	2011 Amamoto [77]
silyl ether metathesis	poly(ethylene-covinyl alcohol) + bis-silyl ether	T_g =−125 °C; T_v =45 °C; T_m =93 °C	2019 Tretbar [78]
thiol-Michael addition	2-hydroxyethyl 3-((2-hydroxyethyl)thio)propanoate +2-((3-(2-(acryloyloxy)ethoxy)-3-oxopropyl)thio)ethyl acrylate (TMADA; thiol-Michael adduct; dynamic crosslinker) = poly(hydroxyethyl acrylate) (PHEA)	T_g =9–16 °C depending on the ratio of TMADA	2016 Zhang [79]
alkoxyamines	DGEBA + diethylenetriamine + (4-hydroxy-1-(20-hydroxy-10-phenyl- 10-methyl) ethyl-TEMPO)	T_g ≈ 60 °C	2014 Yuan [80]
	diarylbibenzofuranone + triethanolamine	n.a.	2015 Imato [81]
transcarbonation	1,4-butanediol + bis(6-membered cyclic carbonate)	T_g =20–40 °C depending on the catalyst ratio	2018 Snyder [82]

T_g (°C): glass transition temperature; T_v (°C): vitrimer transition temperature; T_m (°C): melting temperature; LOI (V/V%): limiting oxygen index; UL-94: standard flammability test (V-0: self-extinguishing classification).

investigated in the study.

Luzuriaga et al. [57] studied the recyclability of a dynamic epoxy system based on the reversible exchange of aromatic disulfides through tensile tests and visual analysis of the recycled surface. The polymer specimen was synthesised from diglycidyl ether of bisphenol A (DGEBA) epoxy component and 4-aminophenyl disulfide dynamic hardener. Diethyltoluenediamine (DETDA) was used as a reference amine-type crosslinking agent. Given the short relaxation times (20 s at 200 °C), it was expected that this dynamic system, although exhibiting similar or equal mechanical properties to the DGEBA reference, could be easily reprocessed at temperatures above T_g. The polymer specimens were ground to fine powder and hot-pressed under a pressure of 100 bar at 200 °C for 5 min. They found that recycling does not have a significant influence on the tensile strength and strain, as well as that the recycled polymer surface has a perfect visual appearance. They explained this beneficial effect by the fact that the disulfide bonds broke during the grinding process and the reshuffling of such bonds led to a full recovery of mechanical properties.

Zhou et al. [28] analysed the influence of the mechanical recycling steps of poly(butylene terephthalate) vitrimer on the resultant storage modulus. They developed a semi-crystalline vitrimer by the incorporation of glycerol (opened epoxy analogue) into poly(butylene terephthalate) via a solid-state(co)polymerization in the presence of zinc(II) acetylacetonate hydrate as a transesterification catalyst. During the recycling process, they cut the vitrimer specimens into small pieces and compression moulded at 250 °C for 25 min. The recycling was repeated four times. Dynamic mechanical thermal analysis (DMTA) results showed that the dynamical properties of the recycled specimens were

Table 2
Overview of recycling processes of vitrimers along with the main properties of the virgin and recycled vitrimer resins.

Dynamic mechanism	Polymer components		Pristine resin properties	Recycling (Reprocessing)	Recycled resin properties	Reference
			T_g (°C) σ (MPa) E (GPa) ε (%)		T_g (°C) σ (MPa) E (GPa) ε (%)	
transesterification	poly(buthylene terephthalate) (PBT)	glycerol, 1,1,1-tris(hydro-methyl)propane, pentaerythritol	T_g = 59–80 °C E = 1090 MPa σ = 67 MPa	Mechanical recycling through cutting and hot-pressing (T = 250 °C, p = 100 bar, t = 25 min); recycling cycles: 4	dynamic properties of the recycled vitrimer matrix specimens were not influenced significantly by the number of recycling steps	2017 Zhou [28]
	diglycidyl ether of bisphenol A (DGEBA)	4,4′-dithiodibutyric acid (DTDA), triazobicyclodecene (TBD)	T_g = 30 °C E = 1.43 GPa	Mechanical recycling through breaking and hot-pressing (T = 100 °C, t = 60 min); recycling cycles: 4	strength of the dual dynamic vitrimer was not changed significantly	2019 Chen [17]
	vanillyl alcohol (VA)	4-methylcatechol (MC)	T_g = 11–23 °C σ = 0,18–25 MPa ε = 95–2250 %	Mechanical recycling through grinding and manual hot-pressing (T = 120 °C, t = 4 h)	tensile stress and strain of the recycled sample matched those of the original sample	2019 Zhao [18]
	diglycidyl ether of bisphenol A (DGEBA)	fatty acids and glutaric anhydride	T_g = 28–75 °C E = 3 MPa–1,15 GPa ε = 200 %–10 %	Mechanical recycling through grinding and hot-pressing (T = 200 °C, p = 16 MPa, t = 2 h); recycling cycles: 3	no significant influence on T_g and storage modulus; slight decrease of rubbery modulus; after third recycling maintained a comparable stretchability and 80 % of the original stiffness	2019 Zhang [34]
	tung oil-based triglycidyl ester	menthane diamine	T_g = 60–83 °C σ = 27–56 MPa ε = 50-12 %	Mechanical recycling through cutting and hot-pressing (T = 180 °C, p = 15 MPa, t = 1 h); Chemical recycling in ethanol amine (EA) at 90 °C for 60 min	tensile strength and the elongational break of the reprocessed vitrimer decreased by 81.63 % and 67.50 %, respectively	2021 Xu [21]
imine	terephthaldehyde	diethylene triamine, triethylene tetramine	–	Mechanical recycling through grinding and hot-pressing (T = 80 °C, p = 90 kPa, t = 45 min); recycling cycles: 3	no significant differences in the resulting tensile strength, only the elastic modulus decreased slightly	2014 Taynton [38]
	vanillin with aminophenol	polyfunctional amine	T_g = 71 °C σ = 46 MPa ε = 4 %	Chemical recycling through acidic hydrolysis by hydrochloric acid and different solvents (efficiency of solvent: water > dimethyl sulfoxide (DMSO) > dimethylformamide (DMF) > ethanol > tetrahydrofuran > benzene ≈ toluene)	no significant decrease observed in tensile or thermal properties	2018 Zhao [41]
	vanillin and epichlorohydrin	4,4′-methylenebiscyclohexanamine	T_g = 170–175 °C σ = 81–86 MPa E = 2000–2300 MPa ε = 8–15 %	Mechanical recycling through cutting and hot-pressing (T = 180 °C, p = 15 MPa, t = 20 min)	slight increase in storage modulus, other mechanical properties of the vitrimer remained unchanged or slightly improved after reprocessing	2019 Wang [42]
	vanillin	methacrylic anhydride; 4-(dimethylamino)pyridine (DMAP)	T_g = 25–31 °C (DSC) T_g = 56–75 °C (DMA) σ = 13–21 MPa E =	Mechanical recycling through grinding and hot-pressing (T = 200 °C, p = 2 MPa, t = 10 min); recycling cycles: 2 Chemical recycling in excess hexylamine/THF	elongation at break and tensile strength decreased, and the Young modulus increased slightly after the second reprocessing	2020 Xu [35]

(*continued on next page*)

Table 2 (*continued*)

Dynamic mechanism	Polymer components		Pristine resin properties	Recycling (Reprocessing)	Recycled resin properties	Reference
			T_g (°C) σ (MPa) E (GPa) ε (%)		T_g (°C) σ (MPa) E (GPa) ε (%)	
			470–830 MPa	solution at room temperature. After 24 h, complete dissolution occurred		
	lignin derived vanillin	methylcyclohexanediamine, ethylene glycoldiglycidyl ether	$T_g > 130$ °C σ = 82 MPa	Mechanical recycling through grinding and hot-pressing (T = 180 °C, p = 5 MPa, t = 40 min); recycling cycles: 3 Chemical recycling of the epoxy vitrimer through two methods. The first involved degrading epoxy networks via imine metathesis using ethylenediamine (EDA) at 100 °C, followed by recovery through EDA evaporation. The second method utilized HTDA for epoxy resin degradation	tensile strength of the recycled vitrimers decreased with the increase of recycling steps; storage moduli and the T_g exhibited a monotonous increase with the increase of recycling steps	2020 Memon [44]
	bisphenol A diglycidyl ether-based epoxy	vanillin-terminated cyclophosphazene	$T_g = 98$ °C σ = 184 MPa E = 18,7 GPa ε = 1.75 %	Mechanical recycling through cutting and hot-pressing (T = 120 °C, p = 18 MPa, t = 5 min)	no obvious decrease in the tensile strength and Young modulus	2021 Liu [36]
	glycerol triglycidyl ether	vanillin and 4-aminophenol	$T_g = 70$ °C σ = 42–62 MPa E = 1.6-2.6 GPa ε = 2.7–18.6 %	Mechanical recycling through cutting and hot-pressing (T = 140 °C, p = 20 MPa, t = 10 min)	no significant changes in mechanical and thermal properties	2021 Liu [46]
	hexacholorocyclotriphosphazene	terephthalaldehyde, diaminodiphenyl methane, p-hydroxybenzaldehyde	$T_g = 146$ °C σ = 56 MPa ε = 8.4 %	Mechanical recycling through breaking and hot-pressing (T = 180 °C, p = 15 MPa, t = 30 min); recycling cycles: 3	after the first recycling cycle, only a slight decrease was observed in the tensile strength; strain slightly decreased after the second and third recycling cycles	2022 Zhang [49]
	vanillin-based epoxy	4,4′-diamonophenylmethane, diethylenetriamine, isophoronediamine, and polyetheramine	$T_g =$ 58–143 °C σ = 33–79 MPa E = 0,85-2.9 GPa ε = 1.7–9.4 %	Chemical recycling: mixture was heated at 60 °C for 1.5 h to completely dissolve the epoxy resin, then more vanillin-based epoxy was added, and the solvent was evaporated	mechanical and thermal properties of recycling epoxy vitrimers are slightly inferior to the origins	2022 Wang [37]
	biobased di-furfural monomer	various biobased multifunctional amines	$T_g =$ 8–60 °C σ = 6.5–77.8 MPa E = 0.85-2.9 GPa ε = 1.7–9.4 %	Chemical recycling in the solution of MeTHF and 1 M HCl at 50 °C for 24 h	properties of the recycled vitrimer were nearly identical to those of the pristine polyimine	2024 Türel [51]
disulfide	diglycidyl ether of bisphenol A (DGEBA)	diethyltoluenediamine (DETDA), 4-aminophenyl disulfide	$T_g = 130$ °C σ = 88 MPa ε = 7.1 %	Mechanical recycling through grinding and hot-pressing (T = 200 °C, p = 100 bar, t = 5 min); recycling cycles: 1	no significant differences in the resulting tensile strength and strain	2016 Luzuriaga [57]
	biobased vanillin	phloroglucinol epoxy components, and 4-amino-phenyl disulfide	$T_g =$ 105–190 °C σ = 66–105	Mechanical recycling through grinding and hot-pressing (T = 40–50	T_g of vitrimers containing both vanillin and	2020 Genua [61]

(*continued on next page*)

Table 2 (*continued*)

Dynamic mechanism	Polymer components		Pristine resin properties	Recycling (Reprocessing)	Recycled resin properties	Reference
			T_g (°C) σ (MPa) E (GPa) ε (%)		T_g (°C) σ (MPa) E (GPa) ε (%)	
			MPa ε = 4–7 %	°C above T_g, p = 100 bar, t = 10 min)	phloroglucinol-based epoxy components increased slightly	
	disulfide-type epoxy vitrimer	9,10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide (DOPO)	T_g = 125 °C	Mechanical recycling through grinding and hot-pressing (T = 200 °C, p = 100 bar, t = 5 min)	same mechanical properties after reprocessing, i.e. 90 % of the initial strength is achieved	2021 Li [58]
urethane	DGEBA based epoxy	vinylogous urethane	T_g = 73–82 °C	Mechanical recycling through breaking and hot-pressing (T = 150 °C, t = 15 min); recycling cycles: 4	the recycling steps did not influence the T_g, Young modulus and tensile strength significantly	2020 Spiesschaert [72]
imine acetal	vanillin and vinyl ether precursors	acetal, aldehyde, and oxirane functionalities	T_g = 36–63 °C σ = 50–70 MPa ε = 10–20 %	Chemical recycling in the mixture of tetrahydrofurane and hydrochloric acid at 65 °C for 2 h	acidic depolymerisation produced a mixture of polyols suitable for upcycling into high-performance thermosetting polyurethane	2023 Türel [50]
	di-vanillin acetal monomers	variety of diamines (i.e. isophorone diamine (IPDA), 4,4′-methylenebis (cyclohexylamine) (MCA)	T_g = 0.5–42 °C σ = 1.6–41.8 MPa ε = 13–244 %	Mechanical recycling through cutting and hot-pressing (T = 120 °C, 50 kN, t = 30 min) Chemical recycling: full dissolution in hydrochloric acid at 50 °C within 30 min	comparable tensile stress after mechanical recycling; recovered compounds were used to generate new polymers with identical thermomechanical properties	2023 Saito [53]

T_g (°C): glass transition temperature; σ (MPa): tensile strength; E (GPa): Young's modulus; ε (%): strain.

not influenced significantly by the number of recycling steps.

Chen et al. [17] analysed the mechanical properties of three vitrimers (dual dynamic vitrimer, single disulfide vitrimer and single ester vitrimer), as well as analysed their recyclability through hot-pressing at 100 °C for 1 h. The synthesis of dual dynamic vitrimer involved reacting the diglycidyl ether of bisphenol A (DGEBA) and a disulfide-containing carboxylic acid, 4,4′-dithiodibutyric acid (DTDA) at 180 °C for 4 h, with triazobicyclodecene (TBD) as a transesterification catalyst. This vitrimer was capable of disulfide metathesis and carboxylate transesterification simultaneously. Additionally, two reference networks were synthesised: a single-disulfide vitrimer without TBD catalyst and a single-ester vitrimer with sebacic acid replacing disulfide-containing acid. They broke the cured vitrimers into pieces and remoulded them into rectangular specimens. The single disulfide vitrimer and single ester vitrimer could not be reprocessed at this low temperature, but due to the rapid stress relaxation and a moderate temperature of malleability enabled by the synergy of disulfide metathesis and carboxylate transesterification, the dual vitrimer was successfully remoulded. They repeated the recycling process up to four times. They tested the tensile properties of the vitrimers after each recycling step and found that the strength of the dual dynamic vitrimer was not changed significantly. They concluded that its mechanical strength revealed full recovery.

Zhao et al. [18] prepared a series of transesterification-based epoxy vitrimers whose reprocessing properties could be tuned on demand by adjusting the ratio of monoepoxides and triepoxides by a controlled glycidylation of a bisphenolic compound obtained from the reaction of vanillyl alcohol (VA) with 4-methylcatechol (MC). Networks with a higher epoxide content exhibited robust thermomechanical characteristics, including high crosslink density, mechanical strength, and resistance to thermal and solvent effects. Vitrimers with a higher monoepoxide content, on the other hand, demonstrated enhanced reprocessability, with features such as high strain, self-healing abilities without external pressure, and strong adhesive properties. These latter ones were recycled through hot-pressing ground vitrimer using manual pressure at 100 °C. The tensile stress and strain of the recycled sample matched those of the original sample after undergoing reprocessing via melt moulding.

Zhang et al. [34] analysed the mechanical recyclability of epoxy-based vitrimers based on transesterification, synthesised from diglycidyl ether of bisphenol A (DGEBA) fatty acids with different chain lengths and glutaric anhydride, by varying the composition of the vitrimers, and the processing time and temperature of hot-pressing. Furthermore, they developed a computational method that can predict the thermomechanical properties of recycled vitrimers based on the distribution of ground vitrimer particle size. They ground the vitrimers and then reprocessed them by hot-pressing using a pressure of 16 MPa at 200 °C for 2 h. They repeated the recycling steps three times. They found that the recyclability of vitrimers significantly depends on the particle size distribution, processing time and temperature. Generally, the hot-pressing time and temperature affect the extent of bond exchange reactions at the interfaces of vitrimer particles, i.e. longer time and higher temperature increased the interfacial chain density and modulus.

However, as the graphs in Fig. 2 show, the recycling steps were found to have an insignificant influence on the T_g and storage modulus; only a slight decrease of the rubbery modulus was observed from 8.5 MPa to 7.3 MPa, probably due to the breakage of permanent covalent bonds during the vitrimer grinding. The mechanical properties of the recycled samples were slightly degraded after each recycling cycle. Yet, compared to the 1st recycled sample, the sample after the third recycling maintained a comparable stretchability and 80 % of the original stiffness.

Wang et al. [42] developed a polyimine-type epoxy vitrimer by synthesising a monofunctional epoxy containing one formyl group from vanillin and epichlorohydrin, and consequently curing it with 4, 4′-methylenebiscyclohexanamine. The vitrimer was reprocessed by hot-pressing chopped films under a pressure of 15 MPa at 180 °C for 20 min. The FT-IR results showed that there was no obvious change in the spectrum of the reprocessed vitrimers. Mechanical testing revealed that, apart from elongation at break, other mechanical properties of the vitrimer remained unchanged or slightly improved after reprocessing. The slight increase in storage modulus suggested increased crosslink density, possibly due to oxidation or side reactions (reaction between amine and Schiff base or self-cross-linking of Schiff base) during hot-pressing.

Xu et al. [35] developed vanillin-based vinyl ester resins with imine bonds and analysed their recyclability through tensile tests. The vitrimers were synthesised by methacrylation and consequent imination by an aliphatic diamine and a cycloaliphatic triamine. The resins were further cross-linked by UV light. The vitrimers were first ground into powder, then hot-pressed using a pressure of 2 MPa at 200 °C for 10 min. The reprocessing was repeated two times. The authors reported that both vitrimers exhibited comparable mechanical properties, i.e. the Young modulus varied between 470 MPa and 830 MPa and the tensile strength between 13 MPa and 21 MPa. Although the mechanical properties after the first reprocessing step did not change significantly, the elongation at break and tensile strength decreased, and the Young modulus increased slightly after the second reprocessing. The latter is explained by the beneficial post-curing effect during hot-pressing at 200 °C, as the original thermosets had a small amount of uncured double bonds left. The vitrimer synthesised from cycloaliphatic triamine retained its mechanical and thermal properties slightly better than the one made from aliphatic diamine.

Spiesschaert et al. [72] synthesised a novel polyfunctional, oligomeric amine curing agent containing vinylogous urethane bonds that was used to prepare vitrimers by reacting it with traditional epoxy resin. They analysed the mechanical recyclability of vitrimers by FT-IR, DMA, DSC and tensile tests. The authors broke the vitrimers into small pieces, then hot-pressed at 150 °C for 15 min, and repeated this procedure four times. They reported that the recycling steps did not influence the T_g, Young modulus and tensile strength significantly. Nevertheless, the elongation of the recycled vitrimers was slightly affected, and the stress-relaxation time increased from 5.5 s to 6.5 s at 160 °C due to the oxidation of the pendant amines, leading to a lower amount of amines available for the transamination reactions.

Memon et al. [44] synthesised an imine-based epoxy hardener from lignin-derived vanillin and methylcyclohexanediamine, which was then used for the crosslinking of ethylene glycol glycidyl ether. The resulting imine-containing epoxy vitrimer was reprocessed three times, repeating the following steps: (i) grinding into fine powders and (ii) hot-pressing using a pressure of 5 MPa at 180 °C for 40 min. These recycling steps can be seen in Fig. 3a. They analysed the recyclability of such vitrimers by tensile tests (see in Fig. 3b.) and DMA (see in Fig. 3d and 3e.). They observed that the tensile strength of the recycled vitrimers decreased with the increase of recycling steps (see in Fig. 3c.). However, the tensile strength of the 3rd recycling step was still above 70 % of the virgin vitrimer. They highlighted two main issues that could negatively affect and limit their results: (i) incomplete fusion and bond exchange, and (ii) accelerated resin ageing resulting from repeated hot-pressing. These issues are recommended to be investigated in more detail in the future. Their DMA results showed that the storage moduli and the T_g exhibited a monotonous increase with the increase of recycling steps, i.e. the thermomechanical properties of the reprocessed vitrimers were even found to be better than virgin vitrimers.

Genua et al. [61] synthesised aromatic disulfide-based epoxy vitrimers from biobased vanillin and phloroglucinol epoxy components and 4-aminophenyl disulfide crosslinking agent, and analysed their recyclability through DSC, DMA and TGA. They ground the vitrimer samples into a fine powder and hot-pressed for 10 min at a temperature that is 40–50 °C higher than the T_g of the samples. They observed that the T_g of two of the vitrimers (vanillin-based and DEGBA-based reference vitrimer) did not change significantly. Nevertheless, the T_g of vitrimers containing both vanillin and phloroglucinol-based epoxy components increased slightly, probably due to the post-curing phenomena. In addition, they found no relevant variation in the storage modulus of recycled vitrimers.

Xu et al. [21] developed a catalyst-free, self-healing, chemically degradable vitrimer from renewable tung oil-based triglycidyl ester and menthane diamine curing agent. They analysed the recyclability efficiency of this vitrimer through tensile tests. They mechanically cut the vitrimer into small pieces and hot-pressed at 180 °C for 1 h at 15 MPa. The tensile strength and the elongational break of the reprocessed vitrimer decreased by 81.63 % and 67.50 %, respectively.

Liu et al. [36] synthesised a flame retardant polyimine-type vitrimer with high crosslinking density and high phosphorus and nitrogen content from vanillin-terminated cyclophosphazene, and analysed its recyclability by tensile test. They cut the vitrimer into small pieces, then hot-pressed at 120 °C for 5 min using 18 MPa. They found no obvious decrease in the tensile strength and Young modulus compared with the virgin vitrimer. Therefore, such reprocessing parameters are recommended to be used in the future to recycle cut-thin vitrimer films.

Liu et al. [46] synthesised a novel biobased epoxy vitrimer from glycerol triglycidyl ether and an imine-containing hardener. They recycled this vitrimer by cutting the samples into small pieces and hot-pressing at 140 °C for 10 min under 20 MPa. They repeated these reprocessing steps three times and measured the tensile and chemical properties after each reprocessing step. The vitrimer could be reshaped into a smooth and continuous film due to the dynamic exchange of imine bonds. Considering that neither the mechanical properties nor the chemical structure nor the T_g changed significantly, they considered the recyclability performance of the vitrimer to be excellent.

Li et al. [58] analysed the recyclability of a disulfide-type flame retardant epoxy vitrimer containing 3 % commercial flame retardant, 9, 10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide (DOPO). The tensile properties of the cured polymer are tested until its break; then, these are compared with the properties of reprocessed samples. They reprocessed the polymer by grinding it into fine powder first, then hot-pressing at 200 °C at 100 bars for 5 min. They measured almost the same mechanical properties after reprocessing, i.e. 90 % of the initial strength is achieved. They explained this slight degradation by the fact that the porosity of the reprocessed polymer was not uniform due to the imbalanced pressure along the specimen surface. In addition to the promising mechanical properties of the reprocessed polymer, their surface quality was excellent, i.e. the reprocessed laminate was defect-free and expressed relatively high transparency.

Zhang et al. [49] synthesised polyimine vitrimers from cyclophosphazene bearing three aldehyde groups, terephthalaldehyde, and diaminodiphenyl methane. They investigated the influence of the mechanical recycling cycle number on the mechanical and chemical properties of recycled vitrimers. The cured vitrimer films were broken first, then hot-pressed at 180 °C for 30 min under 15 MPa. The surface of the formed films was smooth and homogeneous without apparent macroscopic cracks and defects. The FT-IR spectra showed that the recycled samples did not change in terms of their chemical structure. However, the tensile strength decreased with the reprocessing cycles. After the first recycling cycle, only a slight decrease was observed in the

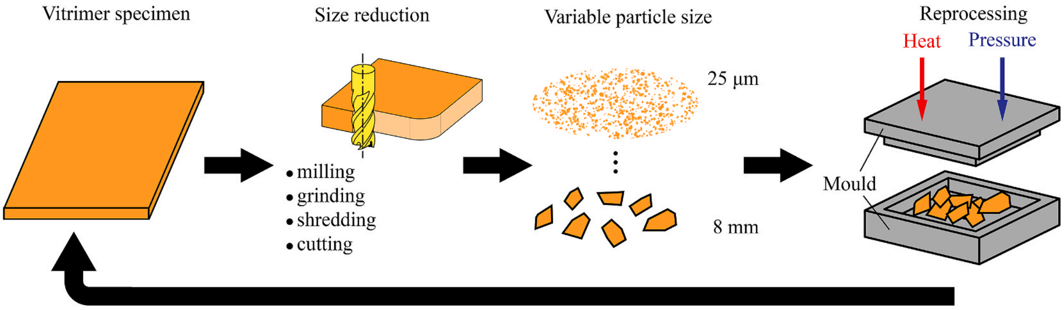

Fig. 1. Schematic overview of mechanical recycling steps of vitrimers, consisting of two major steps of mechanical size reduction via several technologies and hot-pressing the particles.

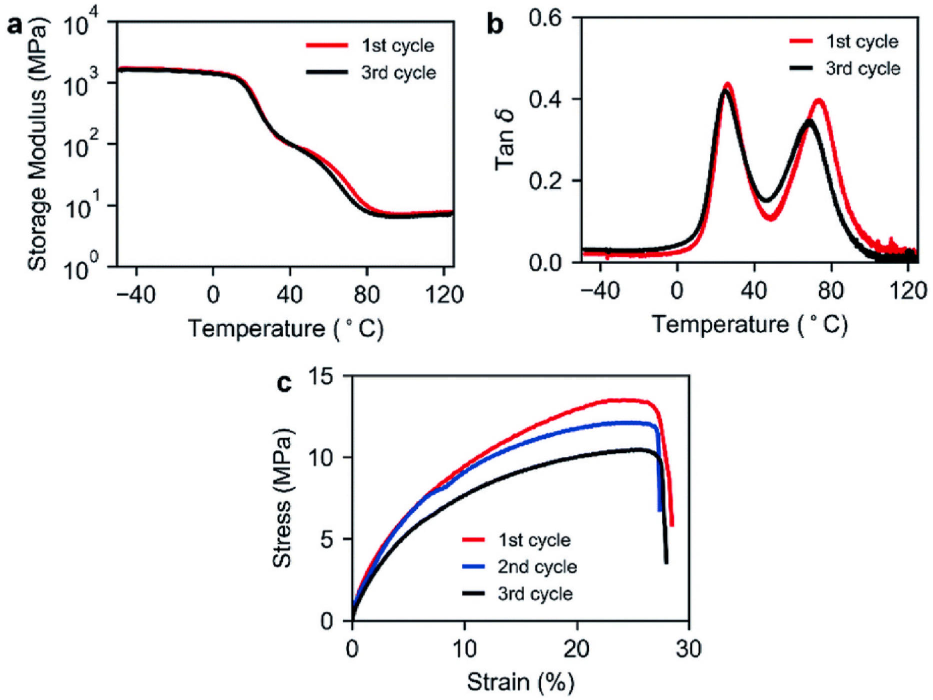

Fig. 2. Recyclability of the vitrimer blends. (a) Storage modulus vs. temperature of 1st, 3rd recycled samples. (b) Tan δ vs. temperature of 1st, 3rd recycled samples. (c) Uniaxial tensile test results of samples after being recycled for several generations [34].

tensile strength (96.6–99.2 % compared to the virgin sample), then it decreased continuously to 92.3–95.7 % after the third recycling cycle. The strain slightly decreased after the second and third recycling cycles – similar to the tensile strength – mostly because of side reactions. Nevertheless, they observed that the strain increased slightly after the first reprocessing step (up to 109.5 %).

Based on the reviewed articles, the mechanical recycling method itself did not depend significantly on the type of dynamic covalent chemistry used. Rather, the applied recycling temperatures were determined primarily by the specific chemical structure of the vitrimer and the resulting vitrimer transition temperature (T_v). For transesterification-based vitrimers, the reported recycling temperatures typically ranged from 100 to 250 °C, while for imine-based vitrimers, they varied between approximately 80 and 200 °C.

As for the properties of the recyclates, the mechanical recycling of vitrimers generally preserved key properties like mechanical strength and T_g, even across multiple cycles. Based on the reviewed articles, mechanical recycling of vitrimers generally preserves key properties like mechanical strength and T_g, even across multiple cycles. Studies by Taynton et al. [38] and Luzuriaga et al. [57] found minimal changes in strength and modulus after recycling, with effective bond reshuffling maintaining integrity. Zhou et al. [28] and Chen et al. [17] also reported stable dynamic properties and full recovery of mechanical strength after several recycling steps. This may be attributed to the fundamental nature of dynamic covalent bonds in CANs, which allow bonds between polymer chains to reform within the crosslinked vitrimer network, leading to a homogeneous structure with favourable mechanical properties. However, in some cases, minor reductions in properties have been observed. Zhang et al. [34] noted a slight decrease in rubbery modulus, while Memon et al. [44] reported a 30 % drop in tensile strength after three cycles, attributed to incomplete fusion and ageing. Zhang et al. [49] reported a slight decrease in tensile strength after repeated recycling. The reduction in mechanical properties may be caused by extensive damage to the polymer chains, as well as by contaminants trapped between the vitrimer particles during the recycling process. Despite this, T_g and storage modulus often improve due to post-curing during hot-pressing, as shown by Xu et al. [35] and Wang et al. [42]. Overall, vitrimer networks demonstrate resilience, though further optimisation is needed to limit property degradation over repeated recycling. It has to be noted that although the reviewed studies analyse the impact of mechanical recycling on the properties of reprocessed vitrimers, the reprocessing parameters are often not varied despite their expected

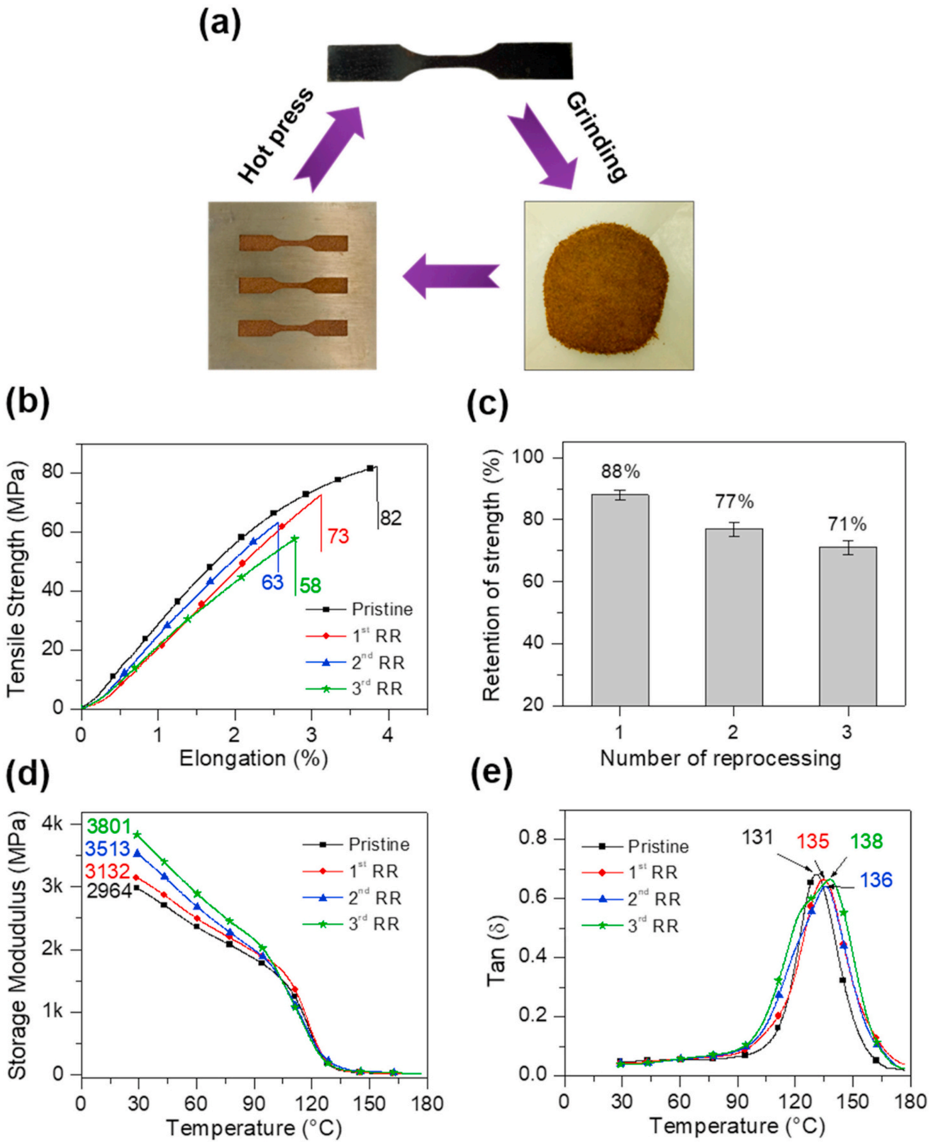

Fig. 3. (a) Illustration of the reprocessing of cured epoxy resins; (b) Representative tensile strength-elongation curves of pristine and reprocessed epoxy resins; (c) Retention of tensile strength as a function of reprocessing cycles; (d) Storage modulus and (e) tan δ versus temperature curves of pristine and reprocessed epoxy resins [44].

influence being significant. Therefore, special attention is recommended in the future on the detailed influential analysis of the (i) particle size, form and quality, (ii) size-reduction technology (whether milling, grinding, braking or shredding) and its process parameters (cutting speed, feed rate, tool geometry, application of coolants etc.), (iii) reprocessed film thickness and quantitative surface quality. Besides the essential vitrimeric material development efforts, future research should also focus on developing recycling technologies to support industrial implementation and get closer to the effective mechanical recycling of epoxy-like technical polymers encouraged by the circular economy.

2.2.2. Chemical recycling

Chemical recycling methods are more sophisticated and highly effective techniques to recover vitrimer resins. The ultimate aim of chemical recycling is to recover the vitrimer monomers in an unchanged chemical structure; thus, they can be reused as monomers again for the synthesis of recycled vitrimers. The main steps of the vitrimer resin's chemical recycling can be seen in Fig. 4. This section summarises the latest results in the chemical recycling of vitrimer systems with different exchange mechanisms.

In 2015, Denissen et al. [7] reviewed covalent adaptable network (CAN) polymers, categorising them based on their chemical structure and reactivity, which govern the dynamic exchange reactions within the polymer network. More recently, in 2023, Türel et al. [83] provided an excellent review of the working principles, properties, potential, and limitations of chemically recyclable dynamic epoxy systems, also organised by functionality. Following this approach, the subsequent sections will discuss chemical recycling methods using a similar classification framework.

Chemical recycling of vitrimers based on imine exchange reactions often involves mild acidic hydrolysis of imine linkages by hydrochloric acid solutions or reaction with an excess amine that initiate imine metathesis. The recycling process is usually carried out at temperatures ranging from room temperature to 100 °C [21,37,44,54], with reaction times varying from 1 to 24 h [37,45,56], largely influenced by the reaction temperature.

Zhao et al. [41] synthesised a polyimine-based epoxy vitrimer by initially reacting vanillin with aminophenol, followed by glycidylation and subsequent crosslinking with a commercially available polyfunctional amine. This vitrimer exhibited properties similar to traditional

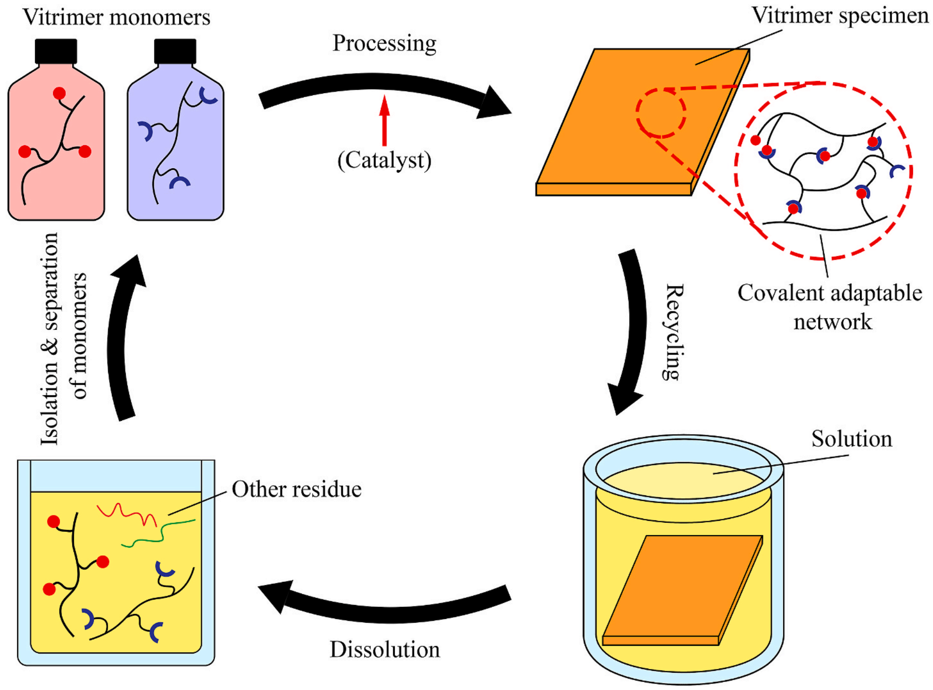

Fig. 4. Schematic overview of chemical recycling steps of vitrimers.

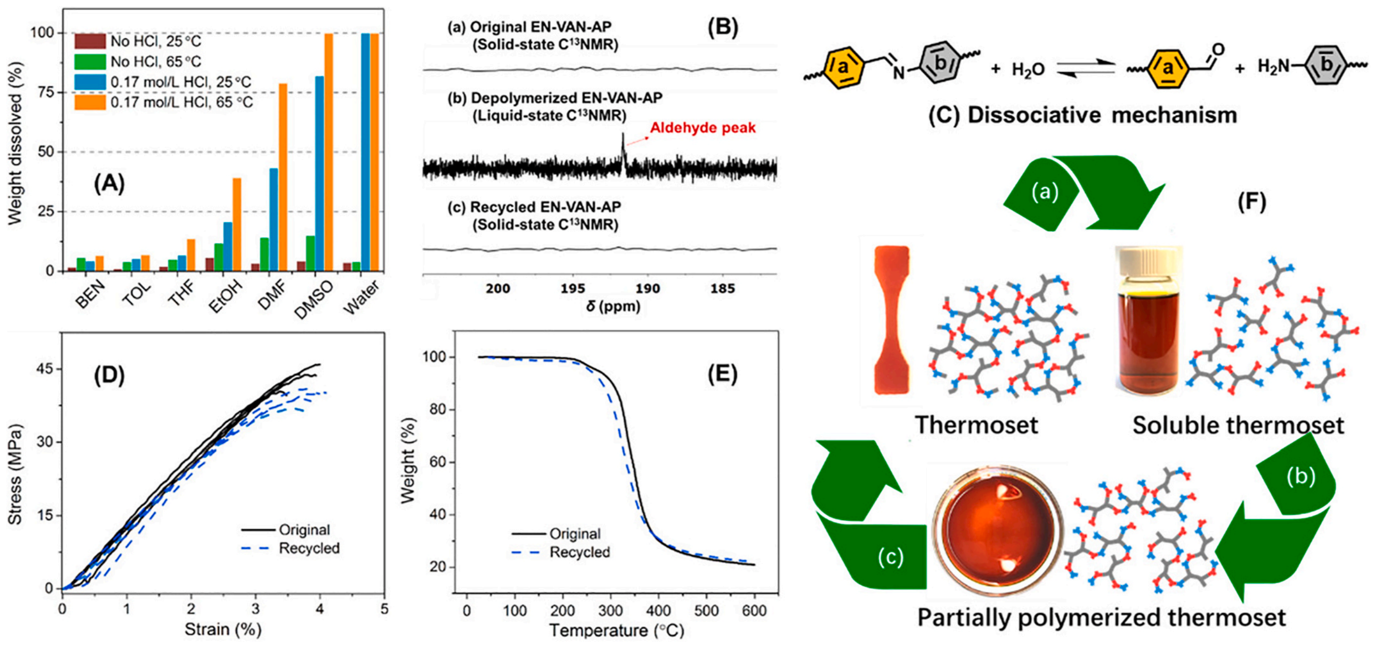

Fig. 5. Depolymerisation and recycling of EN-VAN-AP through a dissociative mechanism. (A) Effects of solvent, HCl concentration, and temperature on the depolymerisation and solubility of EN-VAN-AP. (B) NMR spectra of solid-state (a) original and (c) recycled EN-VAN-AP and (b) liquid-state depolymerised EN-VAN-AP in acidified DMSO-d_6. (C) Dissociative mechanism of the imine bond. (D) Tensile and (E) thermal properties of original and recycled EN-VAN-AP. (F) Recycling steps of EN-VAN-AP. (a) Thermoset was dissolved in DMF with the aid of HCl. (b) DMF and HCl were removed from the solution, leaving the partially polymerised thermoset as a viscous gel. (c) Polymer gel was heated to restore the thermoset [41].

bisphenol-A-based thermosets, although it had a lower onset degradation temperature due to the heightened dissociating tendency of imine bonds at elevated temperatures. The study showcased the recycling of imine-based vitrimers through a dissociative mechanism involving acidic hydrolysis of imine linkages by hydrochloric acid solutions (depicted in Fig. 5C.). The efficiency of solvent recycling followed a specific order (as Fig. 5A shows): water > dimethyl sulfoxide (DMSO) > dimethylformamide (DMF) > ethanol > tetrahydrofuran > benzene ≈ toluene, aligning with the polarity of the solvents. The recycling procedure (whose main steps are shown in Fig. 5F.) entailed dissolving the vitrimer in acidified solvents, followed by drying to eliminate solvent and HCl, and subsequently re-crosslinking at an elevated temperature to regain the thermoset. The recycled thermoset maintained a comparable chemical structure and properties to the original thermoset, with no significant decrease observed in tensile (see in Fig. 5D.) or thermal properties (see in Fig. 5E.). However, challenges arose with solvent

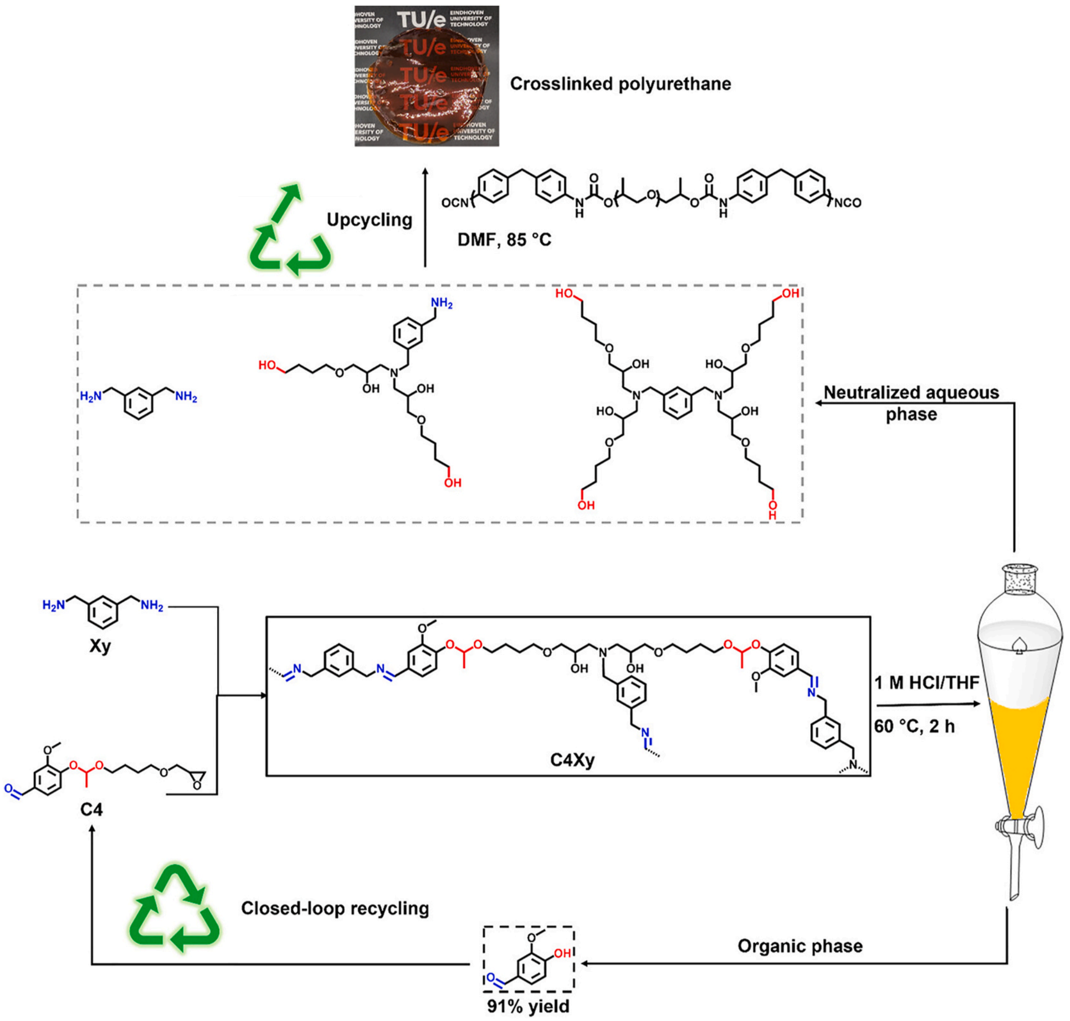

Fig. 6. Schematic representation of depolymerisation of C4Xy, partial closed-loop recycling toward vanillin, and upcycling into polyurethane [50].

removal, particularly for DMSO, which possesses a high boiling point and may affect the properties of the recycled thermoset.

In 2020, Xu et al. [35] introduced two chemically recyclable imine-type vitrimers derived from vanillin through methacrylation and imination with diamine or triamine. These photocurable vitrimers exhibited the capability of chemical recycling in excess hexylamine/THF solution at room temperature. After 24 h, complete dissolution occurred as excess hexylamine initiated an imine exchange reaction, substituting the diamine. Consequently, this process led to the degradation of the thermoset vitrimers into linear polymers.

In 2020, Memon et al. [44] developed an epoxy resin featuring dynamic imine bonds derived from lignin-derived vanillin and methylcyclohexanediamine (HTDA). They achieved chemical recycling of the epoxy vitrimer through two methods. The first involved degrading epoxy networks via imine metathesis using ethylenediamine (EDA) at 100 °C, followed by recovery through EDA evaporation. The resulting epoxy powders, obtained after EDA removal, were then hot-pressed into new epoxy resins. The second method utilized HTDA for epoxy resin degradation, followed by re-utilization of the obtained amine-functional oligomers for epoxy resin production by adding vanillin and epoxy monomers. The recycled resin from the second method retained 95 % of the tensile strength of the pristine resin, whereas the first method yielded a recycled resin with only 74 % of the original tensile strength. This reduction is likely attributed to residual EDA or potential defects introduced during the hot-pressing process. Both chemically recycled epoxy resins exhibited comparable glass transition temperatures to the pristine epoxy resin, indicating no significant changes in the crosslinking density.

Wang et al. [37] made vanillin-based epoxy vitrimers using four different curing agents: 4,4′-diamonophenylmethane, diethylenetriamine, isophoronediamine, and polyetheramine. These vitrimer materials were also used as the matrix materials for carbon fibre-reinforced composites, and the recyclability of the resins and the fibres were investigated. Typically, the vitrimers based on vanillin and isophorone diamine were placed into ethanol, and then an excess amount of isophorone diamine was added to induce the imine exchange reaction. This mixture was heated at 60 °C for 1.5 h to dissolve the epoxy resin completely, then more vanillin-based epoxy was added, and the solvent was evaporated. They stated that this method provides reusable epoxy monomer for vitrimer formulations, although the stoichiometric ratio of the epoxy and the curing agent must be fixed. According to Wang et al. the recycling process is a close-looped method, and no waste is generated during recycling. However, the recycled materials showed lower T_g and weaker mechanical properties than pristine vitrimer systems.

Türel et al. [51] synthesised a biobased di-furfural monomer and polymerized it with various biobased multifunctional amines to create a library of polyimines. These polyimine thermosets exhibited tailored thermal and mechanical properties, with glass transition temperatures ranging from 8 °C to 60 °C and tensile strengths between 6.5 and 77.8 MPa. The polyimines demonstrated high char yields of up to 57 % at 800 °C and a significant bio-content of 78 %–90 %. They featured closed-loop recyclability under mildly acidic conditions, allowing for the

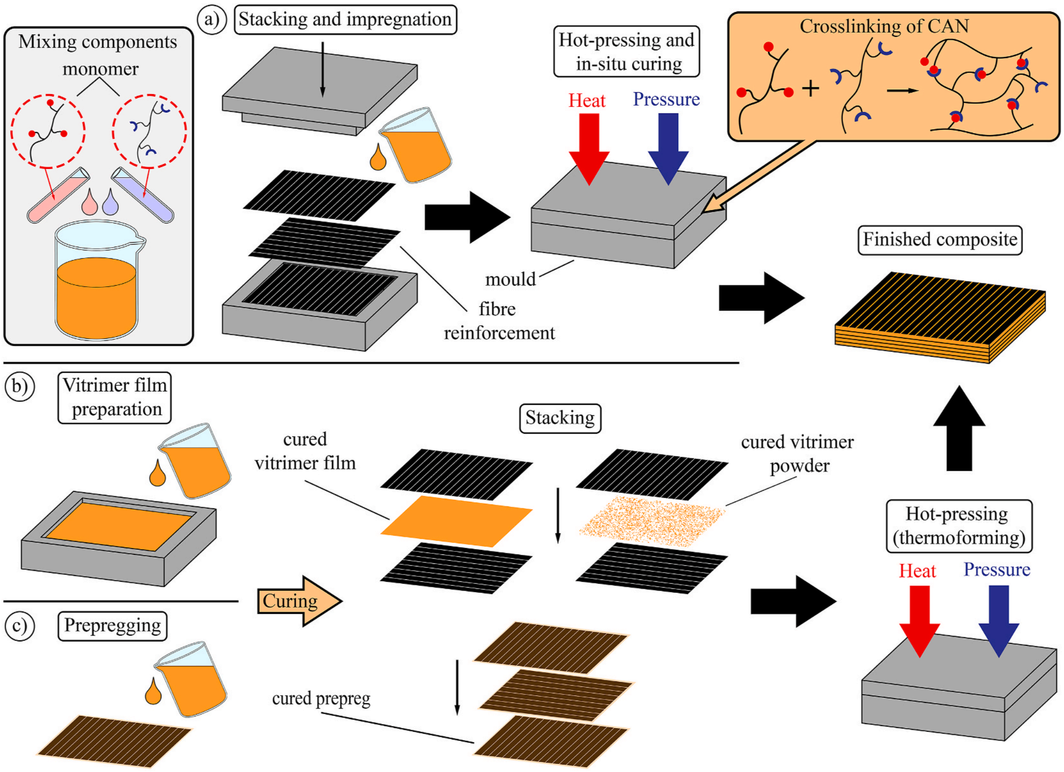

Fig. 7. Schematic figure of the main steps of vitrimer composite fabrication techniques: (a) in-situ polymerisation during lamination process, (b) hot-pressing of cured vitrimer film or powder with dry fibre reinforcement, (c) stacking and thermoforming of cured vitrimer prepregs.

efficient recovery of monomers. Depolymerisation in a MeTHF/1 M HCl mixture at 50 °C caused the organic monomer (OBMF) to precipitate as a fine powder, which was filtered, washed, and recovered in 97 % yield. The crosslinker TREN·HCl remained in the aqueous phase and was regenerated (55 % yield) via neutralisation with an anion-exchange resin. The recovered monomers retained high chemical purity, as confirmed by ^{1}H NMR, and were successfully reused to synthesise a recycled vitrimer (rP(FIm)-TREN) with properties comparable to the original, demonstrating the feasibility of full material recovery and reuse.

Chemical recycling of disulfide vitrimers is commonly performed through disulfide metathesis, utilizing excess thiol group-containing solutions. This process can occur at moderately elevated temperatures of up to 90 °C with relatively short reaction times or at room temperature with extended reaction durations of up to 24 h.

Si et al. [60] investigated degradable, reprocessable, and recyclable epoxy vitrimers and their composites, where both the epoxy and the amine components of the matrix were based on aromatic disulfide bonds, which form an aromatic exchangeable disulfide crosslink network (dual disulfide vitrimer resin). Due to the high content of these disulfide crosslinks, the dual disulfide vitrimers can easily become malleable via disulfide metathesis. This chemical reaction required thiol-based solutions (e.g., dithiothreitol (DTT) with dimethyl formamide (DMF)) and a moderately high temperature of 90 °C.

Di Mauro et al. [19] developed epoxidized vegetable oil (EVO)-based vitrimer systems containing disulfide aromatic dicarboxylic acid. According to the source of the EVOs, the vitrimer resins showed a wide range of thermomechanical properties and T_g approx. 90–110 °C. They investigated the solubility of the materials in methanol, ethanol, acetone, toluene, NaOH and HCl solutions, although the materials did not solve but rather swelled in these solutions.

Chemical recycling of vitrimers containing ester functionalities can be performed through hydrolysis under acidic or basic conditions, as well as through alcoholysis employing an excess of alcohol or aminolysis. Although these are established methods, they usually require high temperatures and/or high pressures, and the separation of oligomers obtained from depolymerisation is not yet efficient [83]. Notably, the ability to enable chemical recycling through solvolysis, such as hydrolysis, while also maintaining high stability and low water uptake during the vitrimer's service life presents a challenge in balancing these competing properties for practical applications.

The chemical recycling of vitrimers can be enhanced by incorporating dual or multiple exchange mechanisms into their structure. The applicable recycling methods align with those suitable for single exchange mechanisms. Ideally, the tailored synthesis should integrate exchange mechanisms that can be recycled using the same techniques.

In 2023, Türel et al. [50] synthesised two liquid epoxy precursors bearing acetal, aldehyde, and oxirane functionalities via catalyst-free, click-type addition reaction between vanillin and vinyl ether precursors, which could be cured with commercial diamines under solvent-free conditions. The vanillin-based recyclable epoxy resins exhibited tensile strength of 50–70 MPa, elongation at break of 10–20 % and glass transition temperature of 36–63 °C (determined by differential scanning calorimetry). The incorporation of reversible imine and acetal bonds into the epoxy network enabled efficient chemical depolymerisation under mild acidic conditions in the mixture of THF/1 M HCl (2/8 by volume) at 65 °C in 2 h, yielding high-purity vanillin in a more efficient pathway than with traditional methods from starting from lignin. Additionally, the acidic depolymerisation produced a mixture of polyols suitable for upcycling into high-performance thermosetting polyurethane. The main steps of the chemical procedure of recycling C4Xy toward vanillin and upcycling into polyurethane are depicted in Fig. 6.

In 2023, Saito et al. [53] combined imine and acetal bonds to create polymer networks based on vanillin, enabling chemical recycling to primary building blocks owing to the presence of these dual-cleavable bonds. Their approach involved the synthesis of di-vanillin acetal monomers through a catalyst-free, click-type addition reaction between vanillin and di-vinyl ether. Subsequently, they prepared crosslinked

Table 3
Technology overview of processing, reprocessing and recycling of vitrimer composites along with the main properties of the virgin and recycled vitrimer composites.

Dynamic mechanism	Polymer components	Processing	Recycling and reprocessing	Properties		Reference
				Pristine	Recycled	
transesterification (TRE)	DGEBA + fatty acid mix	Mould casting of 13.7 m/m % woven carbon fibres and epoxy at 130 °C for 6 h.	Dissolving composite in ethylene glycol at 180 °C for 4 h. Evaporating ethylene glycol at 180 °C for 3 h. Mould casting of reclaimed fibres and epoxy at 180 °C for 10 h.	$T_{g,\ resin} = 30$ °C $\sigma_{fibre} = 4200 \pm 190$ MPa $E_{fibre} = 239 \pm 11.7$ GPa	$\sigma_{fibre} = 4400 \pm 530$ MPa $E_{fibre} = 223 \pm 16.3$ GPa	2016 Yu [13]
	DER331 + ethylenediamine + glutaric anhydride	Hand lay-up 43 m/m% woven carbon fibres.	Decomposing composite in Parr reactor (water) at 180 °C for 5 h.	$T_{g,\ resin} = 70.7$ °C $\sigma_{composite} = 356 \pm 28.7$ MPa $E_{composite} = 17.1 \pm 2.5$ GPa $\sigma_{fibre} = 3100 \pm 540$ MPa	$\sigma_{fibre} = 2870 \pm 350$ MPa	2021 Liu [22]
	DGEBA + cashew nut shell liquid + citric acid	Prepregging of plain weave carbon fibre fabric (BN CC200P) with a fibre areal weight of 200 g/m^2 laminates. Curing the laminates individually for 1 h at 60 °C, 80 °C, 100 °C, 120 °C and for 2 h at 200 °C. Hot-pressing of cured laminates at 150 °C for 20 min under 690 kPa.	Dissolving composite in dimethylformamide at 100 °C for 2 h	$T_{g,\ resin} =$ 24–32 °C $\sigma_{composite} \approx 250$ MPa	$\sigma_{composite} \approx 240$ MPa	2021 Lorwanishpaisarn [32]
	tung oil-based triglycidyl ester + methane diamine	Hot-pressing of 65 m/m% woven carbon fibre layers with in-situ polymerization at 100 °C for 2 h, at 140 °C for 3 h, at 160 °C for 2 h under 5 MPa.	Dissolving composite in ethanolamine at 90 °C for 1 h.	$T_{g,\ resin} = 89.8$ °C $\sigma_{composite} = 541 \pm 8$ Mpa $\varepsilon_{composite} = 6.9 \pm 0.6$ % σ_{fibre} and ε_{fibre} are close to the recycled values.	$\sigma_{fibre} = 2880 \pm 235$ MPa $\varepsilon_{fibre} = 1.8 \pm 0.23$ %	2021 Xu [21]
	epoxidized menthane diamine + adipic acid	Hot-pressing of vitrimer coated plain weave filament carbon fibre fabric (3 K, 200 g/m^2) at 100 °C for 2 h, at 130 °C for 2 h, at 150 °C for 2 h under 15 MPa.	Dissolving composite in ethanolamine at 60 °C for 30 min.	$T_{g,\ resin} =$ 72.1–86.4 °C $\sigma_{composite} = 465$ MPa $\varepsilon_{composite} = 7.7$ % σ_{fibre} and ε_{fibre} are close to the recycled values.	$\sigma_{fibre} = 2795$ MPa $\varepsilon_{fibre} = 4.3$ %	2022 Xu [48]
imine exchange	terephthaldehyde + tris(2-aminoethyl) amine + diethylenetriamine	Hot-pressing of impregnated 35 m/m% woven carbon fibres at 78 °C for 3 h, at 95 °C for 1 h and at 105 °C for 1 h.	Dissolving composite in diethylenetriamine for 24 h.	$T_{g,\ resin} = 56$ °C $\sigma_{composite} = 309 \pm 57$ MPa $E_{composite} = 12.2 \pm 1.7$ GPa $\varepsilon_{composite} = 3.8 \pm 1.2$ %	–	2016 Taynton [39]
	60 % trifunctional epoxy resin (AFG-90H) + 40 % ethylene glycol diglycidyl ether (EGDGE) ICH (imine containing hardener) = lignin-derived vanillin and methylcyclohexanediamine	Prepregging of 58–63 m/m % plain weave carbon fibre fabric (HS700) laminates. Curing the laminates individually for 15 min at 80 °C. Hot-pressing of cured laminates at 80 °C for 1 h, 140 °C for 2 h, 170 °C for 1 h under 0.2 MPa.	Hot-pressing of damaged composite parts for 1 h at 180 °C under 5 MPa. Recovering fibres by dissolving with ethylenediamine.	$T_{g,\ resin} = 131$ °C $\sigma_{flexural,\ composite} = 1028$ MPa $E_{flexural,\ composite} = 56$ GPa ILSS = 52 MPa	$\sigma_{flexural,\ composite} = 764$ MPa $E_{flexural,\ composite} = 46$ GPa ILSS = 48 MPa	2020 Memon [44]
	epoxidized soybean oil + vanillin + aminophenol	Prepregging of 28.5 m/m% carbon fibres (at 40 °C for 12 h and at 80 °C for 3 h). Pre-curing prepregs at 150 °C for 2 h. Hot-pressing at	Cutting composite into pieces. Hot-pressing at 180 °C under 20 MPa for 10 min.	$T_{g,\ resin} = 28$ °C $\sigma_{composite} = 145.4 \pm 17$ MPa $E_{composite} =$	$\sigma_{composite} = 11.21 \pm 0.35$ MPa $E_{composite} = 0.76 \pm 0.13$ GPa	2020 Liu [45]

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Table 3 (*continued*)

Dynamic mechanism	Polymer components	Processing	Recycling and reprocessing	Properties		Reference
				Pristine	Recycled	
		210 °C under 10 MPa for 50 min.		1.18 ± 0.14 GPa $\varepsilon_{composite}$ = 19 ± 1 %	$\varepsilon_{composite}$ = 8 ± 2 %	
	polyethereimide (D230) + vanillin terminated cyclotriphosphazene	Prepregging of 65 m/m% plain weave carbon fibres. Hot-pressing of prepregs under 18 MPa at 120 °C for 15 min.	Recovering fibres by dissolving with 0.1 M HCl/DMF at room temperature for 20 h	Recovering fibres by dissolving with 1 M HCl/THF at room temperature for 12 h	$T_{g,\ composite}$ = 98.4 °C $\sigma_{flexural,\ composite}$ = 271 MPa $E_{flexural,\ composite}$ = 46.3 GPa	$\sigma_{flexural,\ composite}$ and $E_{flexural,\ composite}$ values are close to prestine.
	2021 Liu [36]					
	terephthaldehyde + diethylenetriamine + tris(2-aminoethyl) amine	Prepregging 57 m/m% woven carbon fibres and curing by hot-pressing at 75 °C for 2 h, at 85 °C for 2 h, at 105 °C for 2 h.	Recovering fibres and vitrimer monomers by dissolving in DETA and ethanol solution for 24 h. Preparing composite from reclaimed components.	$T_{g,\ resin}$ = 59.8 °C $\sigma_{composite}$ = 170.45 ± 14.41 MPa $E_{composite}$ = 18.26 ± 3.68 GPa	$\sigma_{composite}$ = 190.68 ± 27.05 MPa $E_{composite}$ = 13.72 ± 3.11 GPa	2021 Yu [54]
		Stacking the cured vitrimer in powder form with 57 m/m% woven carbon fibre layers and hot-pressing at 140 °C for 4 min under 1.25 MPa.	Recovering fibres and vitrimer monomers by dissolving in DETA and ethanol solution for 24 h. Preparing recycled vitrimer powder and processing composite from reclaimed components.	$\sigma_{composite}$ = 184.68 ± 28.58 MPa $E_{composite}$ = 13.10 ± 2.27 GPa	$\sigma_{composite}$ = 145.03 ± 51.61 MPa $E_{composite}$ = 12.19 ± 1.56 GPa	
	terephthaldehyde + diethylenetriamine + tris(2-aminoethyl) amine	Preparing vitrimer films. Hot-pressing vitrimer films and 57 m/m% carbon fibres at 120 °C under 3 MPa for 6 h.	Dissolving composite in propylamine at 120 °C. Preparing vitrimer films from solution. Hot-pressing vitrimer films and reclaimed fibres	$T_{g,\ resin}$ = 75 °C $\sigma_{flexural,\ composite}$ ≈250 MPa	–	2021 He [52]
	glycerol triglycidyl ether + ICH = Vanillin and 4-aminophenol-based imine	Prepregging 62 m/m% plain weave filament carbon fibre fabric. Hot-pressing of prepregs at 140 °C for 25 min.	Dissolving composite in 0.1 mol/L ethylenediamine (solvent DMF) at 50 °C for 2 h. Impregnating reclaimed fibres with reclaimed matrix.	$T_{g,\ resin}$ = 70 °C $\sigma_{composite}$ = 449 ± 12 MPa $E_{composite}$ = 12.9 ± 0.9 GPa $\varepsilon_{composite}$ = 4.6 ± 0.6 % σ_{fibre} = 2783 ± 159 MPa E_{fibre} = 166.5 ± 11.0 GPa ε_{fibre} = 1.8 ± 0.1 %	$\sigma_{composite}$ = 574 ± 9 MPa $E_{composite}$ = 13.7 ± 0.1 GPa $\varepsilon_{composite}$ = 5.1 ± 0.1 % σ_{fibre} = 2584 ± 278 MPa E_{fibre} = 172.5 ± 8.1 GPa ε_{fibre} = 1.6 ± 0.2 %	2021 Liu [46]
	DGEBA + lignin-derived vanillin + m- xylylene-diamine	Prepregging carbon fibres. Hot-pressing of prepregs at 120 °C for 6 h, at 150 °C for 3 h, at 180 °C for 2 h under 15 kPa.	Reclaiming fibres by dissolving composite in o.2 mol/L tetrahydrofuran/H_2O (8:2) solution at room temperature for 12 h.	$T_{g,\ resin}$ = 96 °C $\sigma_{composite}$ = 622 ± 17 MPa $E_{composite}$ = 19.3 ± 2.4 GPa $\varepsilon_{composite}$ = 4.01 ± 0.19 % σ_{fibre} = 3645 ± 192 MPa E_{fibre} = 309.4 ± 7.6 GPa ε_{fibre} = 1.17 ± 0.11 %	σ_{fibre} = 3582 ± 207 MPa E_{fibre} = 308.1 ± 5.3 GPa ε_{fibre} = 1.22 ± 0.12 %	2021 Liu [47]
	DGEBA + lignin-derived vanillin +1,6 hexane-diamine			$T_{g,\ resin}$ = 83 °C $\sigma_{composite}$ = 584 ± 11 MPa $E_{composite}$ = 16.8 ± 1.4	σ_{fibre} = 3533 ± 146 MPa E_{fibre} = 304.6 ± 13.8 GPa ε_{fibre} = 1.20 ± 0.08 %	

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Table 3 (*continued*)

Dynamic mechanism	Polymer components	Processing	Recycling and reprocessing	Properties		Reference
				Pristine	Recycled	
				GPa $\varepsilon_{composite}$ = 4.16 ± 0.30 % σ_{fibre} = 3645 ± 192 MPa E_{fibre} = 309.4 ± 7.6 GPa ε_{fibre} = 1.17 ± 0.11 %		
	vanillin-based epoxy + diethylenetriamine (DETA)	Prepregging woven carbon fibres (198 g/m^2) and curing at 80 °C for 2 h, at 100 °C for 2 h. Hot-pressing layers at 130 °C for 20 min under 10 MPa.	Dissolving composites to reclaim carbon fibres in 0.1 M HCl/DMF solution at 25 °C for 2 h. Washing reclaimed fibres in CH_2Cl_2.	$T_{g,\ resin}$ = 58 °C $\sigma_{composite}$ ≈ 350 MPa $E_{composite}$ ≈5.8 GPa	–	2022 Wang [37]
	vanillin-based epoxy + isophoronediamine (IPDA)	Prepregging woven carbon fibres (198 g/m^2) and curing at 80 °C for 2 h, 120 °C for 2 h, at 140 °C for 1 h. Hot-pressing layers at 130 °C for 20 min under 10 MPa.		$T_{g,\ resin}$ = 132 °C $\sigma_{composite}$ ≈ 571 MPa $E_{composite}$ ≈11.7 GPa		
	vanillin-based epoxy +4,4′-Diaminodiphenylmethane (DDM)	Prepregging woven carbon fibres (198 g/m^2) and curing at 80 °C for 2 h, at 120 °C for 2 h, at 150 °C for 3 h. Hot-pressing layers at 150 °C for 20 min under 10 MPa.		$T_{g,\ resin}$ = 143 °C $\sigma_{composite}$ ≈ 510 MPa $E_{composite}$ ≈3.4 GPa		
	vanillin-based epoxy + polyetheramine D230 (IPDA-D230)	Prepregging woven carbon fibres (198 g/m^2) and curing at 80 °C for 2 h, at 120 °C for 2 h, at 140 °C for 1 h. Hot-pressing layers at 130 °C for 20 min under 10 MPa.		$T_{g,\ resin}$ = 84 °C $\sigma_{composite}$ ≈ 509 MPa $E_{composite}$ ≈6.5 GPa		
	polyimine-based VITRIMAX T130 system	Prepregging 22 m/m% nonwoven and 42 m/m% UD carbon fibres and curing at 150 °C for 1 h, at 180 °C for 1 h. Hot-pressing prepregs at 160 °C for 15 min under 15 bar.	Reclaiming fibres by dissolving composite in diethylenetriamine/xylene (20:80) solution at 80 °C for 8 h. Prepregging reclaimed fibres with pristine resin.	$T_{g,\ nonwoven\ composite}$ = 82.52 °C $T_{g,\ UD\ composite}$ = 70.39 °C ILSS = 37.45 ± 1.74 MPa	$T_{g,\ nonwoven\ composite}$ = 47.66 °C $T_{g,\ UD\ composite}$ = 75.76 °C ILSS = 27.09 ± 2.5 MPa	2024 Pomázi [56] 2024 Toldy [55]
biobased di-furfural monomer + various biobased multifunctional amines	Prepregging woven carbon fibres and curing at 80 °C for 24 h. Hot-pressing at 130 °C for 30 min under 50 kN.	Dissolving composite in 1 M HCl solution for 24 h at 50 °C.	$T_{g,\ resin}$ = 8–60 °C $\sigma_{composite}$ = 295.1 MPa $E_{composite}$ = 10.7 GPa	–	2024 Türel [51]	
disulfide metathesis (DSM)	DGEBA + 4-aminophenyl disulfide	Prepregging 50 m/m% glass fibre reinforcement. Hot-pressing at 200 °C under 100 bar for 5 min	Dissolving composite in 2-mercaptoethanol/DMF solution for 24 h at room temperature for reclaiming reinforcement. Grinding composite into powder. Hot-pressing at 200 °C under 100 bar for 5 min.	$T_{g,\ resin}$ = 130 °C $\sigma_{flexural,\ composite}$ = 595 ± 39 MPa ILSS = 37 ± 3 MPa $\sigma_{compression,\ composite}$ = 292 ± 16 MPa IS = 194 ± 18 kJ/m^2	–	2016 Luzuriaga [57]
		Manual hand lay-up 50 m/m% glass fibres.	–	$T_{g,\ resin}$ = 130 °C $\sigma_{flexural,\ composite}$ = 557 ± 7 MPa ILSS = 29 ± 1 MPa $\sigma_{compression,\ composite}$ = 242 ± 18 MPa	–	

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Table 3 (*continued*)

Dynamic mechanism	Polymer components	Processing	Recycling and reprocessing	Properties		Reference
				Pristine	Recycled	
				IS = 159 ± 18 kJ/m^2		
	DGEBA + diethyltoluenediamine + 4-aminophenyl disulfide	Prepregging plain weave carbon fibres. Hot-pressing at 170 °C for 10 min. Post-curing at 150 °C for 2 h, 170 °C for 2 h.	Dissolving composite in HCl solution at room temperature.	$T_{g,\ resin}$ = 172 °C $\sigma_{composite}$ = 763 ± 71 MPa $E_{composite}$ = 35.3 ± 2.4 GPa $\varepsilon_{composite}$ = 3.02 ± 0.31 % σ_{fibre} = 3020 ± 390 MPa E_{fibre} = 222.9 ± 5.0 GPa ε_{fibre} = 1.62 ± 0.21 %	$T_{g,\ resin}$ = 175 °C σ_{fibre} = 3160 ± 440 MPa E_{fibre} = 231.1 ± 13.8 GPa ε_{fibre} = 1.55 ± 0.19 %	2019 Wang [42]
	bis(4-glycidyloxyphenyl) disulfide + 4-aminophenyl disulfide DGEBA + 4-aminophenyl disulfide	Mould casting of woven carbon fibres and resin at 90 °C for 30 min. Pre-curing at 110C for 30 min and post-curing at 160 °C for 3 h	Dissolving composite in 0.1 mg/ml DTT/DMF solution at 90 °C. Reprocessing reclaimed reinforcement with pristine resin.	$T_{g,\ composite}$ = 131 °C $\sigma_{composite}$ = 334.5 ± 87.7 MPa	$T_{g,\ composite}$ = 126 °C $\sigma_{composite}$ = 320.9 ± 64.6 MPa	2020 Si [60]
	DGEBA + hexamethylene diisocyanate) + 4-aminophenyl disulfide	Prepregging of 64 m/m% unidirectional carbon fibre tows. Curing the laminates individually for 2.5 h at 120 °C. Hot-pressing of cured laminates at 160 °C for 1 min under 500 psi.	Dissolving composite in DDT/DMF solution at room temperature for 24 h. Reprocessing the reclaimed reinforcement with pristine resin.	$T_{g,\ resin}$ = 88–100 °C $\sigma_{composite}$ ≈ 413 MPa $\sigma_{flexural,\ composite}$ ≈ 903 MPa ILSS ≈50 MPa	$\sigma_{composite}$ ≈ 415 MPa	2022 Zhou [62]
	DGEBA + 4-aminophenyl disulfide			$T_{g,\ resin}$ = 140 °C $\sigma_{flexural,\ composite}$ ≈1086 Mpa ILSS ≈ 71 MPa	–	

T_g (°C): glass transition temperature; σ (MPa): tensile strength; E (GPa): Young's modulus; ε (%): strain; $\sigma_{flexural}$ (MPa): flexural strength; $\sigma_{compression}$ (MPa): compressive strength; ILSS (MPa): interlaminar shear strength; IS (kJ/m^2): impact strength.

poly(imine-acetal)s with adjustable thermal and mechanical properties by condensing di-vanillin acetal monomers with selected amines. The use of rigid, cyclic amines resulted in higher glass transition temperature and superior mechanical properties, while the application of flexible amines led to lower glass transition temperature and elastomer-like behaviour. These polymers can be completely depolymerized into pure vanillin, diethylene glycol, respective amines, and acetaldehyde upon hydrolysis of imine and acetal bonds with 0.1 M HCl solution under continuous stirring at 50 °C within 30 min. The authors also demonstrated the selective depolymerisation of these vitrimers under the same conditions within a mixed plastic waste stream (PET, HDPE, PP and PC), which remained intact due to its chemical stability. The recovered compounds were used to generate new polymers with identical thermomechanical properties, establishing an efficient closed-loop recycling process.

3. Vitrimer composites

Vitrimer composites represent a significant advancement in the field of materials science, integrating the advantageous properties of dynamic covalent networks with traditional composite reinforcement strategies. This section provides an overview of the processing and properties of vitrimer composites, emphasising their distinct manufacturing methods compared to conventional thermoset and thermoplastic polymer composites. Due to their unique chemistry, vitrimer composites offer enhanced reprocessability and recyclability, which are vital for tackling sustainability challenges in the materials sector. This section also examines the recycling methodologies for vitrimer composites, emphasising approaches that facilitate the simultaneous recycling of both the fibre reinforcement and the vitrimer matrix.

3.1. *Processing and properties of vitrimer composites*

Vitrimer matrix fibre-reinforced polymer (FRP) composites are typically processed using established thermoset and thermoplastic composite manufacturing techniques. However, due to the dynamic covalent bonds within vitrimers, these materials also enable novel processing approaches that combine the benefits of both thermoset and thermoplastic systems, offering greater flexibility in FRP composite fabrication. The fabrication of vitrimer matrix FRP composites can be divided into two categories based on the crosslinking state of the resin during processing. FRP composites can be produced using either: (i) a process in which the resin crosslinks in situ during fabrication [21,22,36,48,57,60,72,84] (see in Fig. 7a.), or (ii) a process that uses an already cured, crosslinked matrix or composite prepregs. In the latter case, the covalent adaptable network enables the reformation of dynamic covalent bonds between polymer chains after the initial curing process [32,33,37,40,47,52,54,62] (see in Fig. 7b and 7c.). The main technological steps of the different vitrimer composite fabrication techniques can be seen in the schematic Fig. 7.

The liquid, uncured vitrimer resin can be used for various lamination techniques, such as hand lay-up [22], vacuum-assisted hand lay-up [72], prepregging [47,62], mould casting [47] and pultrusion [84], which impregnation and lamination techniques are followed by a curing process, usually by hot pressing [21,48] or in a drying oven. Spiesschaert et al. [72] fabricated unidirectional glass fibre (UD-GF)-reinforced composites with vitrimer matrix (commercially available EPIKOTE resin MGS LR 135 with the synthesised vinylogous urethane curing agent). After analysing the viscosity profiles of the vitrimer, they found that at increased temperatures, their system is suitable for resin transfer technologies; however, this low viscosity is hard to maintain. They hand-laminated the UD-GF layers and sealed the composite in a vacuum bag for the curing process. The vitrimer system exhibited a good adhesion to the glass fibres; however, porosity of the matrix was observed due to the trapped air during the fabrication process. This can be solved with a degassing process step, although the rapidly increasing viscosity of the cooling resin makes this process hard to carry out. The crosslinked vitrimer resin can also be processed using techniques similar to those employed in thermoplastic FRP fabrication. In these techniques usually the reinforcement and the solidified vitrimer resin either in a powder form [33,37,54] or in a thin film form [52] is stacked in a mould and then hot-pressed; also, prepregs with cured matrix can hot-pressed together creating a FRP composite sheet [32,40,47,62]. He et al. [52] manufactured polyimine-based CFRP composite workpieces by making crosslinked polyimine thin films via hot-pressing, which were stacked in a sandwich structure with carbon fibre sheets, then hot-pressed together. Their CFRP composite sheets could be further thermoformed. They highlighted that the void content both of the polyimine film and the CFRP composite could be decreased with pressure applied and heating above the bond exchange reaction temperature; however, the pressure should not exceed the materials' strength to avoid fractures. Toldy et al. [55] and Pomázi et al. [56] fabricated CFRP composites with VITRIMAX T130 polyimine-based vitrimer matrix. First, they prepregged the reinforcement layers individually and then cured them individually in a drying oven for 1 h at 150 °C and for 1 h at 180 °C. The cured prepregs were hot-pressed at 160 °C for 15 min under 15 bar pressure. They examined that due to the technology steps, also the high viscosity and tackiness of the vitrimer, the fibre content of composites is difficult to influence and could result in low fibre content. Moreover, prepregging and hot-pressing the layers may destroy the structure of the reinforcement, which can decrease the quality and mechanical properties of the reclaimed reinforcement. Yu et al. [54] fabricated polyimine-based CFRP composites with two different methods: they used (i) crosslinked polyimine powder for compression moulding and a (ii) liquid-based impregnation method with an in-situ polymerisation process. Their experimental work showed that the powder-based manufacturing technique can result in a similar macro- and microstructure of the composite as the liquid-based technique, but the powder-based method takes significantly less time (4 min instead of 24 h). They also found out that the process parameters (*i.e.* time, temperature and pressure) affect the porosity, impregnation quality and mechanical properties both of the composites and polyimines.

At this point, it has to be also noted that although low processing, repairing and recycling temperatures are beneficial in terms of energy consumption; these vitrimer resins are supposed to replace traditional thermoset matrices with high thermal stability and glass transition temperature. In order to fulfil the requirements of structural application during their service phase, the transition temperature of the vitrimers should be carefully designed.

One of the biggest challenges in vitrimer matrix composite fabrication is the often high viscosity of the resin. This is often due to the presence of dissociative side chains in the monomers [85]. Viscosity plays a key role in the handling of the resin in the case of certain fabrication technologies (e.g. vacuum infusion, pultrusion, etc.); therefore, for high viscosity vitrimers the prepregging and hot-pressing technologies are more suitable. The void content of the composite is highly dependent on the viscosity because air bubbles can become trapped in the resin during the mixing process. In high-viscosity vitrimers, achieving good impregnation is more difficult, and controlling the fibre content precisely is more challenging [55,56] because the resin cannot penetrate efficiently into the fibre rovings. For these reasons, viscosity should be maintained at a low value by applying solvents or higher process temperatures to enable highly automated and high-precision manufacturing technologies for vitrimer composites. However, there is significant industrial potential in technologies where the composite is processed using an already crosslinked matrix. Thanks to this approach, handling and storage of the vitrimer component become much easier compared to prepregs, which can be tacky, may require refrigerated storage, and have a limited shelf life.

Besides recycling, the reuse and repair of composite parts also play an important role in sustainability. However, the repair of most conventional fibre-reinforced thermoset composite structures—such as wind turbine blades or aircraft fuselages—is both time-consuming and expensive, as damaged regions must be removed and fully replaced. In contrast, vitrimer composites represent a significant advancement in repairable materials. Vitrimers, defined by their dynamic covalent networks, enable self-healing in fibre-reinforced architectures through reversible bond exchange reactions. Repairing technologies for vitrimer composites are similar to composite fabrication techniques, while also utilising the advances of dynamic covalent bonds that enable the formation of new connections between crosslinked networks. A growing body of work has demonstrated that diverse vitrimer systems can restore mechanical performance under a variety of loading and environmental conditions.

Yu et al. [54] developed polyimine-based vitrimer composites via a rapid powder compression moulding process. These composites exhibit low-temperature, mould-free repair—even on curved surfaces—using only a handheld iron. Their moisture- and heat-responsive imine bonds allow the material to recover and maintain mechanical integrity, highlighting the potential for adaptive, in-situ repair.

Amfilochiou et al. [86] investigated a siloxane-based vitrimer reinforced with glass fibres for wind-energy applications. By hot-pressing for just 40 min, the vitrimer composite achieved over 89 % recovery of both Mode I and Mode II fracture toughness, outperforming conventional thermosets and suggesting a promising replacement for irreversible epoxy systems in large-scale structural components.

Messmer et al. [87] focused on sustainability by combining natural basalt fibre with a vitrimeric resin. Their vitrimer–basalt composites demonstrated a 25 % increase in stiffness and a 4 % increase in strength (normalised for fibre volume fraction) compared to basalt-epoxy analogues. Low-temperature compression patching restored a substantial fraction of the original tensile strength, underscoring the viability of these composites as durable, eco-friendly alternatives to synthetic fibre–reinforced materials.

Cong et al. [88] explored catalyst-free oxidised carbon black (OCB)/epoxy vitrimer coatings. OCB incorporation enhanced stress-relaxation rates and enabled efficient photothermal-stimulated healing—achieving up to 100 % macroscopic scratch recovery—without catalysts or preheating. Additionally, these coatings exhibit high solar absorptance, shape-memory behaviour, and ion-transfer barrier properties, marking them as multifunctional, contactless-repair systems for outdoor applications.

Lorwanishpaisarn et al. [32] reported a bio-based vitrimer system synthesised from cashew nut shell liquid (CNSL) and citric acid, reinforced with carbon fibre. When applied as a repair patch on a damaged alloy substrate, this composite restored up to 98 % of the original tensile strength. Furthermore, the vitrimer matrix could be dissolved away to recover the carbon fibres, which retained comparable tensile properties to pristine fibres.

Barnett et al. [89] examined epoxy-based vitrimer composites reinforced with recycled carbon fibre. By adjusting consolidation time, they demonstrated a trade-off between increased interlaminar stiffness and reduced ductility, while still maintaining favourable repairability. The ability to process and store these organosheet laminates at room temperature offers a practical advantage for scalable manufacturing.

Collectively, these studies illustrate a rapidly evolving field in which vitrimer composites can deliver scalable, sustainable, and effective repair solutions for structural, coating, and recyclable composite applications. Nevertheless, industrial implementation still faces challenges, including high production costs and processing complexity. Further research is needed to optimise vitrimer formulations and processing routes to facilitate widespread adoption in industry.

3.2. *Recycling of vitrimer composites*

The recycling of fibre-reinforced vitrimer composites has gained increasing attention due to the growing demand for sustainable and reprocessable materials in advanced applications. Owing to their dynamic covalent bonds, vitrimers offer unique recyclability through both mechanical and chemical methods. This section explores key developments in composite recycling, highlighting advances that enable the recovery and reuse of fibre reinforcements and vitrimer matrices. As with vitrimer resins, the reviewed studies are categorised according to the underlying exchange mechanisms (see Table 3).

Imine-based vitrimers demonstrate versatile recycling solutions through dynamic exchange reactions, effectively breaking down polymer networks and allowing for the recovery of valuable fibre reinforcements. Mild acidic conditions combined with moderate temperatures are reported to facilitate the degradation of imine networks while preserving the mechanical properties and structural integrity of the recycled fibres, ensuring their performance remains comparable to virgin counterparts.

In 2016, Taynton et al. [39] synthesised polyimine vitrimers using terephthaldehyde and amines with various chain lengths and functionalities alongside tris(2-aminoethyl)amine (TREN). They showcased the chemical recycling of polyimine networks in carbon fibre-reinforced composites by introducing excess free primary amine groups, such as excess diamine monomer. Through transimination reactions between the excess diamine monomers and the imine-linked network, the number of end groups within the matrix increased, leading to a reduction in molecular mass and solubilization of the network. Simple immersion of carbon fibre-reinforced composite samples in neat diethylenetriamine (DETA) resulted in the complete dissolution of the polyimine vitrimer and recovery of carbon fibre reinforcement.

In 2020, Liu et al. [45] developed a high-performance epoxy vitrimer utilising dynamic Schiff-base chemistry from epoxidised soybean oil, vanillin, and 4-aminophenol. They successfully extracted the carbon fibre reinforcements from the vitrimer matrix via acid-induced dissociation in a 0.1 M HCl DMF solution at room temperature (see in Fig. 8a.), achieving complete dissolution of the matrix within 20 h. Notably, the morphology and stress-strain behaviour of the recycled carbon fibre monofilaments closely mirrored those of the originals, as Fig. 8d shows, indicating the feasibility of recycling the composite without compromising fibre structure (which also did not deteriorate visibly, as the SEM images show in Fig. 8c.) or mechanical properties. In addition to chemical recycling, the authors explored mechanical recycling through crushing and subsequent hot-pressing of the vitrimer composite (Fig. 8b.). As illustrated in Fig. 8e., the mechanical properties of the reprocessed composite were significantly reduced due to the transition from continuous to short carbon fibres. The tensile strength, Young's modulus, and elongation at break dropped to 11.2 ± 0.4 MPa, 0.76 ± 0.13 GPa, and 8 ± 2 %, respectively—compared to 145.4 ± 17.1 MPa, 1.18 ± 0.14 GPa, and 19.2 ± 0.9 % in the original ESO-VA-CF composite. Despite this decline, the vitrimer matrix retained its reprocessability, allowing for consolidation into a new part. While the mechanical performance no longer meets the requirements for structural applications, the reprocessed material remains suitable for non-structural or secondary applications.

Liu et al. [46] investigated the chemical recycling of a novel biobased epoxy vitrimer, synthesised from glycerol triglycidyl ether (Gte) and an imine-containing hardener derived from vanillin and 4-aminophenol (VA). The complete dissolution of the matrix was carried out in ethylene diamine (EDA) solution. After washing and drying, scanning electron microscopy (SEM) analysis confirmed that the recycled CF retained its integrity, showing no fibre damage or visible resin residues. Furthermore, Raman spectroscopy indicated that the chemical structure of the recycled fibres closely resembled that of the original fibres, with similar peak intensities and ratios. The mechanical properties of the

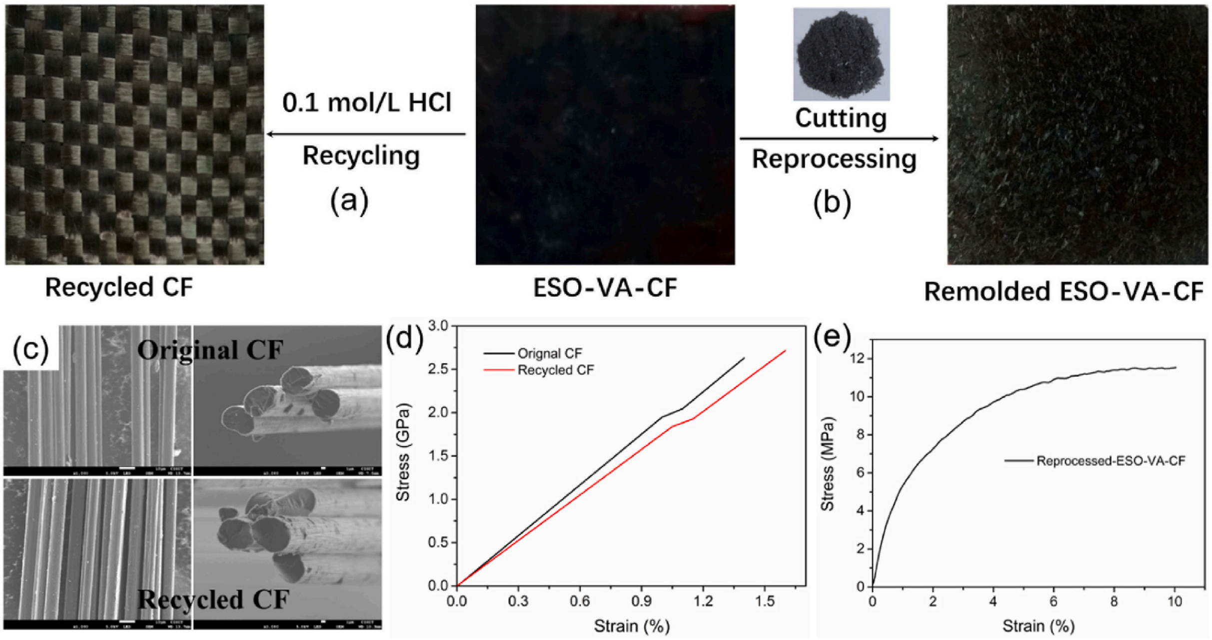

Fig. 8. (a) The recycling process of ESO-VA-CF, (b) the reprocessing of ESO-VA-CF, (c) the SEM of original and recycled CF, (d) the stress-strain curves of original and recycled CF monofilaments, and (e) the stress-strain curve of reprocessed ESO-VA-CF [45].

recycled carbon fibres were assessed through tensile tests, revealing that the Young's modulus and tensile strength of the recycled fibres were nearly identical to those of the original fibres, indicating effective recycling without loss of performance. The recycled carbon fibres were then re-infiltrated with the degraded matrix solution, resulting in recycled carbon fibre-reinforced composites (Gte-VA-CF) that maintained mechanical properties comparable to the original composites, and potentially enhanced due to further curing of the matrix.

Liu et al. [36] reported the closed-loop chemical recycling of carbon fibre-reinforced biobased composite featuring high flame retardancy, achieved by incorporating a vanillin-terminated phosphazene monomer (HVP) into a polyimine binder. The composites were completely degraded in a mixed solution of 1 M hydrochloric acid and tetrahydrofuran (THF) within 12 h, facilitating the near-total retrieval of the plain carbon weaves and HVP monomer. The vitrimer monomer (HVP) was recovered by precipitation in deionised water, followed by washing and vacuum drying, yielding a purified solid product. Scanning electron microscopy (SEM) analysis revealed that the recycled carbon fibres maintained a clean and smooth surface with no visible damage or residual polyimine matrix, indicating effective recovery. The tensile properties of the recycled carbon fibre multifilaments were found to be comparable to those of the virgin fibres. In addition, the flexural properties of the regenerated composites prepared from the recycled fibres and monomer were comparable to those of the original laminates, confirming the efficiency of the closed-loop recycling process. Additionally, the dynamic imine crosslinking provides the composites with excellent reparability, allowing for recovery of more than 90 % of flexural strength after hot-pressing.

In 2021, Xu et al. [21] developed a chemically degradable bio-based vitrimer using tung oil-derived triglycidyl ester and menthane diamine. Carbon fibre-reinforced composites formulated with this imine-based vitrimer system showcased remarkable reprocessing capabilities, self-adhesion, and shape memory attributes. Moreover, they could be rapidly degraded chemically by ethanolamine at 90 °C. The recycled carbon fibres retained comparable surface morphology, chemical structure, and tensile properties to those of the pristine fibres.

Liu et al. [47] synthesised bio-based curing agents for epoxy vitrimer composites from lignin-based vanillin and 1,6-hexylenediamine and

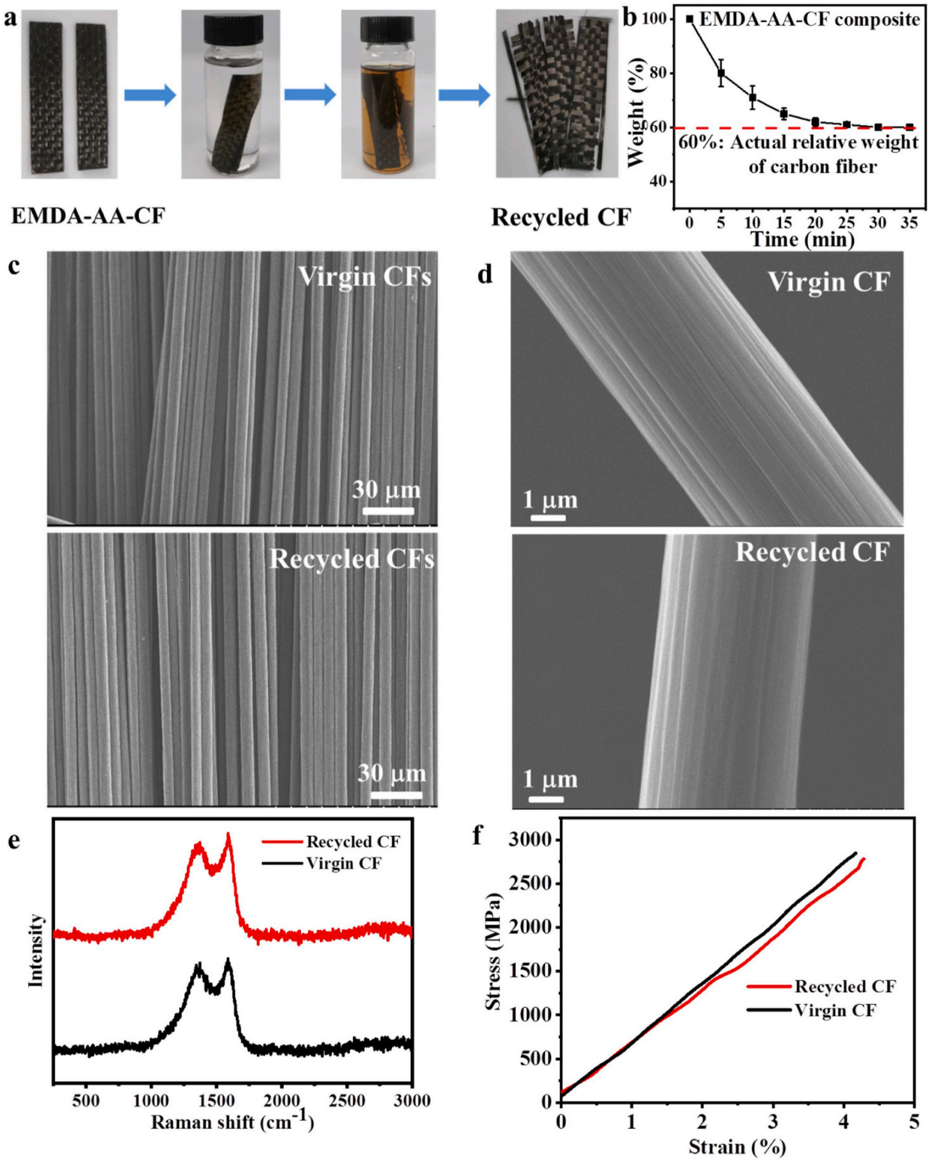

Fig. 9. (a) Photographs of the recycling process of the EMDA–AA–CF composites in ethanolamine at 60 °C. (b) The relative weight of the EMDA–AA–CF composite in the degradation process. (c) SEM images of the virgin and recycled CFs. (d) SEM images of the virgin and recycled CF monofilament. (e) Raman spectra of the virgin and recycled CFs. (f) Stress-strain curves of the virgin and recycled CF monofilaments [48].

m-xylylene diamine. These two curing agents were mixed with DGEBA epoxy and used as the matrix in carbon fibre-reinforced composites. The vitrimer composite samples were placed into 0.2 mol/L HCl solution and stirred with a magnetic stirrer at room temperature. After the dissolution of the matrix (up to 24 h at room temperature), the carbon fibre sheets were neutralised with saturated sodium bicarbonate, then washed with tetrahydrofuran and deionised water, and dried in a vacuum at 80 °C for 6 h. They found that the recycled carbon fibres had the same chemical structure and very similar mechanical properties as the original ones, and they were not damaged during the degradation of the matrix. They stated that both vitrimer resins (with different curing agents) had the ability to degrade under weakly acidic conditions, and their degradation depends on temperature, solvent and acidity.

He et al. [52] developed a sustainable manufacturing method of a polyimine-based vitrimer system containing terephtaldehyde, diethylene triamine and tris(2-aminoethyl) amine as a hardener. They prepared thin films from the polyimine-based vitrimer using composite scraps. According to them, the main advantage is that a fully cured thin thermoset film can be used to make a compact composite structure based on the malleability and the dynamic chemistry of covalent adaptive networks (CANs): the material can fuse into the fabric reinforcement layers and weld them together. The developed process consists of four steps, including the preparation of polyimine films, preparation of the composite laminates, reshaping of composite structures and recycling using propylamine as a solvent. Propylamine seemed to be a proper solvent to dissolve this vitrimer system at elevated temperatures (approx. 50–90 °C) as the amine diffuses into the network and participates in the imine exchange reaction and breaks the polymer chains, resulting in soluble oligomers. This process is reversible. Thus, repolymerisation occurs above the boiling point of the propylamine (above 49 °C, at about 90 °C). They compared the recycled and remanufactured composites to the pristine composites with the same fibre content (approx. 49 %). They found that the shear bonding strength, the flexural strength and flexural stiffness are in the same range. They also stated that their novel process can be repeated multiple times and can be extended to other polymer systems (e.g., anhydride-cured epoxy resins using an alcohol solvent and a bond exchange reaction catalyst; other ester-containing thermosets).

In 2022, Xu et al. [48] developed an epoxidised menthane diamine–adipic acid vitrimer matrix. This vitrimer matrix could be rapidly degraded into polyols via an amination reaction with ethanolamine under mild conditions, enabling efficient recycling of carbon fibres. The recycling process and the weight loss of the composite during the dissolution procedure can be seen in Fig. 9a and 9b. Scanning electron microscopy (see in Fig. 9c and 9d.), Raman spectroscopy (see in 9e.), and tensile tests (see in Fig. 9f.) revealed that the recycled fibres maintained nearly identical chemical structure and mechanical properties compared to the virgin carbon fibres, the chemical recycling did not deteriorate the chemical structure of the fibres.

Pomázi et al. [56] investigated the recycling of carbon fibre-reinforced polyimine vitrimer composites, comparing those made from virgin fibres to those from recycled fibres. The vitrimer matrix was composed of a two-component polyimine vitrimer system, utilising nonwoven felt and unidirectional carbon fibre as reinforcement. Various

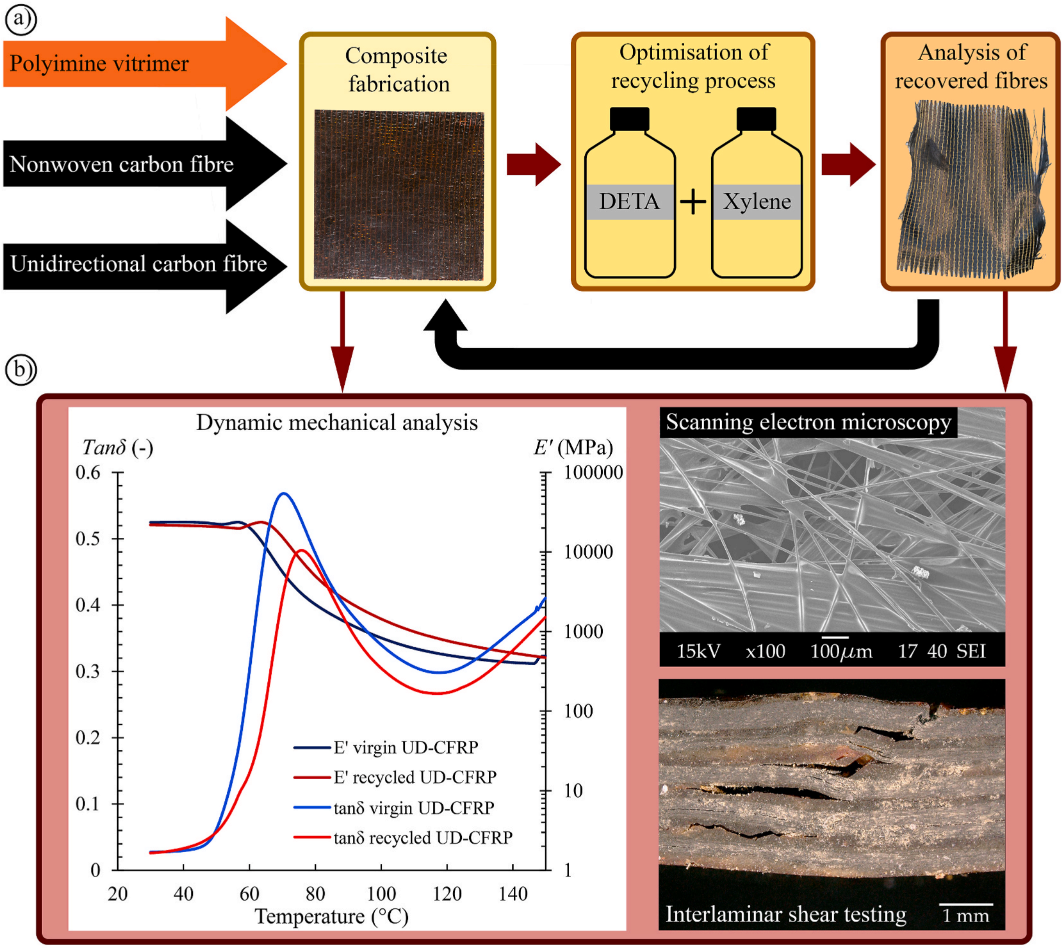

Fig. 10. Chemical recycling of carbon fibre-reinforced polyimine vitrimer composites by dissolution in DETA-xylene solution. (a) Steps of processing, recycling and reprocessing of polyimine-based CFRP composites. (b) Analysis of the effectiveness of the recycling process: dynamic mechanical analysis of virgin and recycled composites; scanning electron microscopic image of reclaimed reinforcement; interlaminar shear testing of virgin and recycled composites [56].

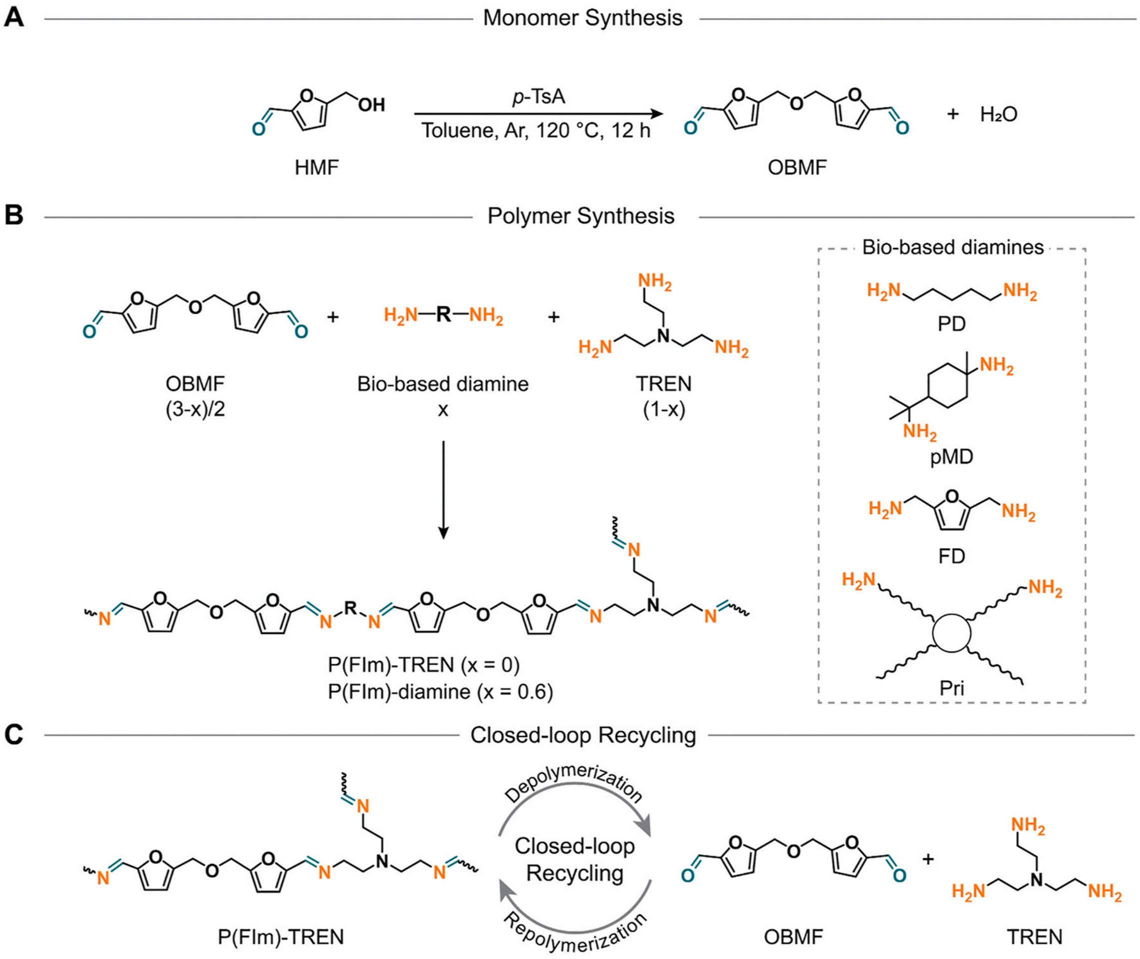

Fig. 11. (A) Synthetic scheme for OBMF and (B) resultant furfural-based polyimine networks. (C) Schematic illustration of the closed-loop recycling of P(FIm)-TREN [51].

ratios of diethylenetriamine (DETA) to xylene were tested to determine the optimal dissolution conditions, with a 20:80 DETA:xylene ratio proving to be the most effective. Elevated temperatures (80 °C) significantly enhanced the dissolution process, and the feasibility of scaling up to larger composite structures was demonstrated. The steps of the recycling experiment can be seen in Fig. 10a. Scanning electron microscopy (SEM) confirmed the effective removal of the matrix with minimal residue on the carbon fibre surfaces, ensuring good adhesion in the recycled composites. The recycled nonwoven composite exhibited a decreased glass transition temperature due to residual solvents in the matrix, while the unidirectional composite exhibited a slight increase. Dynamic mechanical analysis revealed an increased storage modulus for nonwoven composites at room temperature and greater resistance to deformation at elevated temperatures for the unidirectional composites (as Fig. 10b shows). Interlaminar shear tests indicated a slight reduction in adhesion strength in the reprocessed composites.

In 2024, Türel et al. [51] synthesised a biobased di-furfural monomer (see in Fig. 11A.) and polymerised it with mixtures of various biobased di- and triamines to construct a library of polyimines (see in Fig. 11B.). These polyimine thermosets displayed tailor-made thermal and mechanical properties, featuring a wide range of glass transition temperatures from 8 °C to 60 °C and tensile strength spanning from 6.5 to 77.8 MPa. These novel polyimines exhibited high bio-content (in the range of 78 %–90 %) and closed-loop recyclability under mildly acidic and energy-efficient conditions (solution of MeTHF and 1 M HCl (v: v = 2: 8) at 50 °C for 24 h with continuous stirring). The chemical process of closed-loop recycling can be seen in Fig. 11C. The recovered monomers were then reused to synthesise recycled vitrimer, which had almost identical properties to the pristine polymer (see in Fig. 12A and 12B.). Chemical recycling was also demonstrated on 2-ply carbon-fibre reinforced vitrimer composites in 1 M HCl solution at 50 °C for 24 h. The quality of the reclaimed carbon reinforcement compared to the virgin reinforcement can be seen in Fig. 12C. Scanning electron microscopy of recycled carbon fibres revealed that the surface of the fibres was similar to the virgin carbon fibres (see in Fig. 12D.).

Vitrimer composites based on disulfide mechanism can be chemically recycled by dissolution in excess of thiols even at room temperature.

Zhou et al. [62] synthesised dynamic polyurea/epoxy vitrimers using aliphatic hexamethylene diisocyanate prepolymer and DGEBA with 4-aminophenyl disulfide as a dynamic crosslinking agent. Their strategy was to form dynamic bonds between the ductile polyurea segment and the rigid epoxy network based on disulfide metathesis. They found that immersion of the polyurea segment not only improved the mechanical characteristics of the material (for example, toughness, ductility, etc.) but also lowered the processing temperature compared to neat DGEBA epoxy resin. This vitrimer resin was used as the matrix of carbon fibre-reinforced composites, and the recyclability was investigated. Although these vitrimers have good solvent resistance, they can be dissolved in excess thiol group-containing solutions or solution mixtures based on the exchange reaction of thiol and disulfide groups. Zhou et al. used 1-dodecanethiol (DDT) or 3-mercaptopropionic acid for this purpose in DMF or DMAc solvents. They found that each binary solvent formulation fully dissolved the developed polyurea/epoxy vitrimer. The

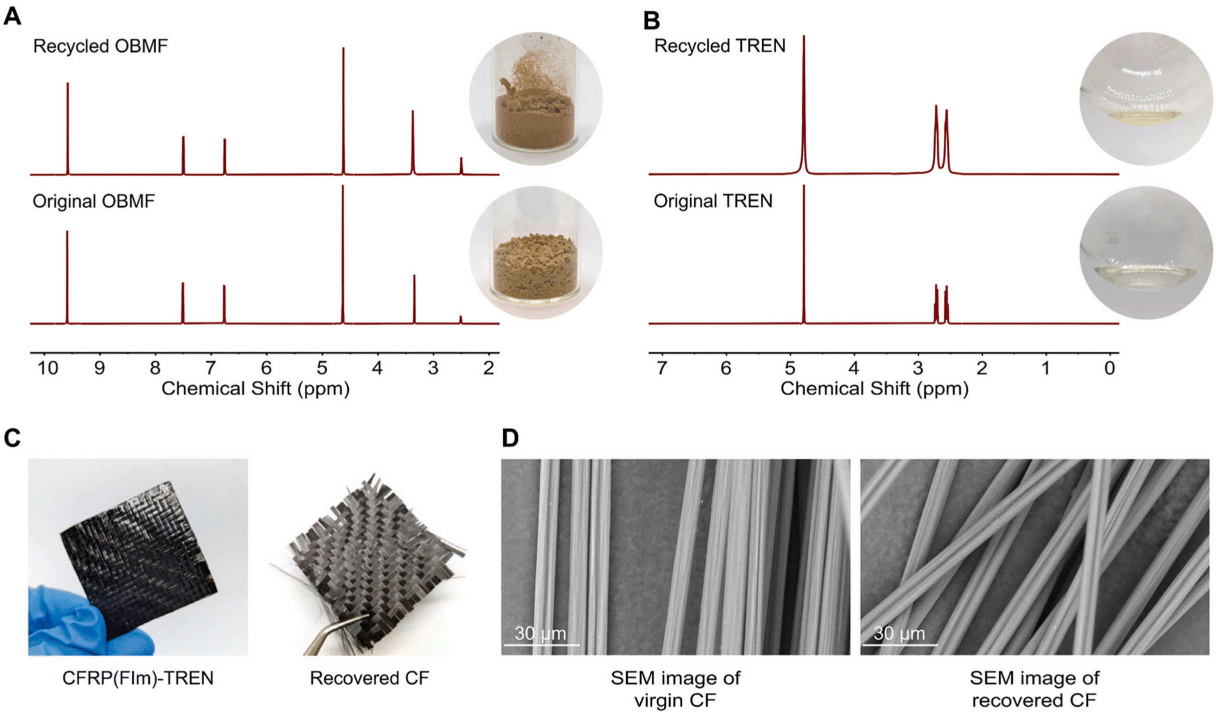

Fig. 12. (A) Chemical recycling of P(FIm)-TREN: physical appearance and ^{1}H NMR spectra of OBMF in DMSO-d_6 and (B) TREN in D2O after depolymerisation and subsequent purification, (C) visual appearance of CFRP(FIm-TREN) and recovered carbon fibre cloth upon depolymerisation of the composite, (D) SEM images of virgin and recovered carbon fibres. Scale bar is 30 μm [51].

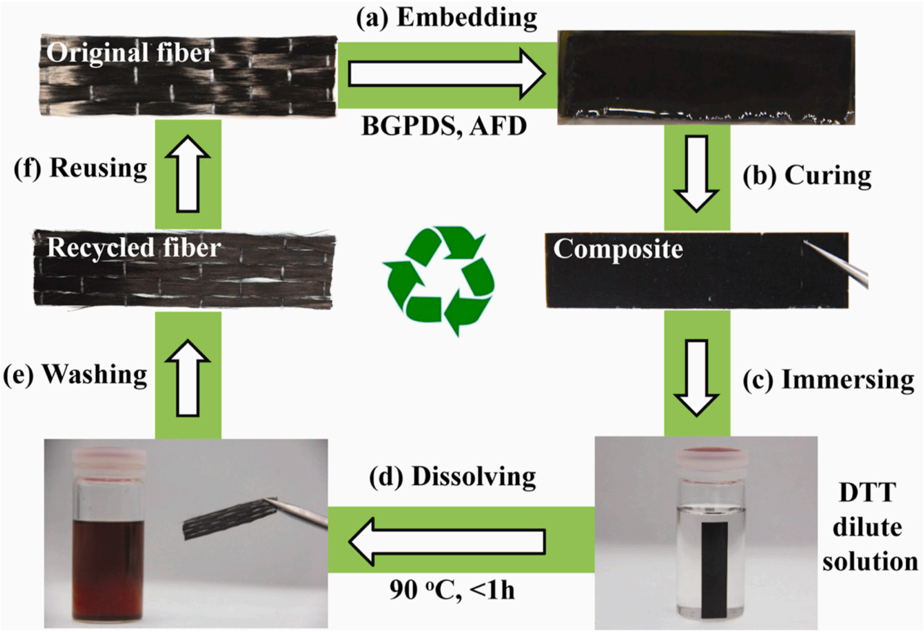

Fig. 13. The recycling process of CFRP composites based on dual disulfide vitrimer: (a) embedding the carbon fibres in epoxy matrix; (b) obtaining CFRP composites by curing; (c) immersing the CFRP composites in DTT dilute solution (0.1 mg/ml); (d) dissolving the epoxy matrix at 90 °C in 1 h; (e) washing and drying the recovered carbon fibres; (f) reusing the recycled carbon fibres to form new composites [60].

composites were soaked in DDT/DMF solution at room temperature for 24 h while the mixture was constantly stirred. After the dissolution of the matrix, the fibres were reclaimed and reused in vitrimer composites. According to Zhou et al., the main advantage of this material compared to “classical” epoxy vitrimers is the higher reprocessing cycle times.

Si et al. [60] investigated the chemical recycling of dual disulfide vitrimer carbon fibre-reinforced composites. They used DTT/DMF at 90 °C for 1 h and further washing, rinsing, and drying cycles for carbon fibre-reinforced composites to recover the fibrous media fully (see the main steps of the procedure in Fig. 13.). They found that after recovering and using the fibres in epoxy composites again, the mechanical properties of these composites reduced only by a small amount (E’ from 10.5 GPa to 8.2 GPa; tensile strength from 334.5 MPa to 320.9 MPa, shown in graph Fig. 14a).

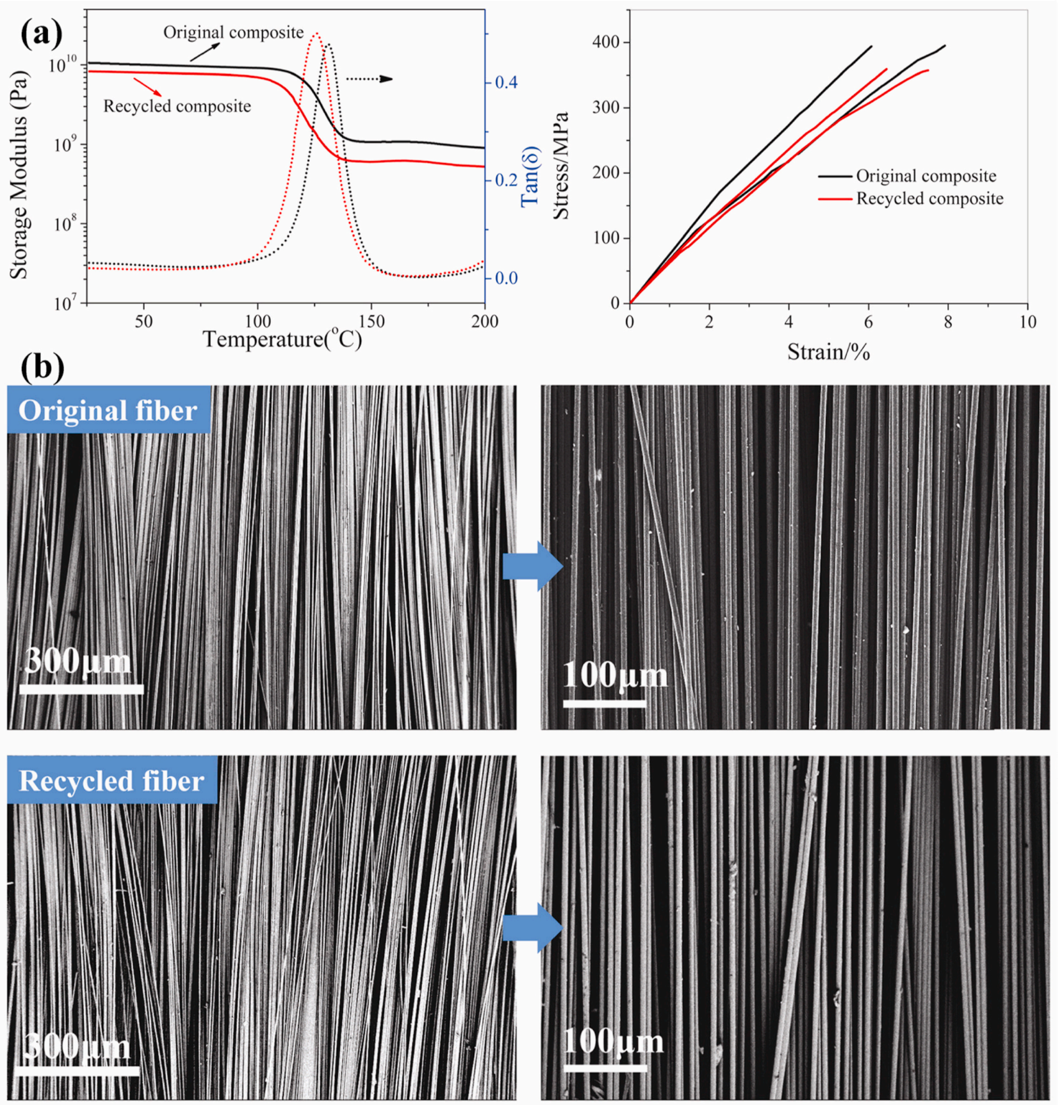

Fig. 14. (a) DMA (left) and tensile tests (right) of the original (black) and recycled (red) CFRP composites; (b) Morphology of the fresh (top) and recycled (bottom) carbon fibres by SEM [60]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Ester-based vitrimer composites can be efficiently recycled through transesterification and hydrolysis, typically involving the use of alcohols or water under elevated temperatures (ranging from 150 °C to 180 °C) and optimised reaction times (from a few hours up to several hours), leading to the successful recovery of high-quality carbon fibres that retain their mechanical performance.

Yu et al. [13] investigated a potentially environmentally and economically friendly method for carbon fibre-reinforced epoxy composites based on covalent adaptive networks (see the main steps of the procedure in the schematic Fig. 15a and the experimental pictures in Fig. 15b.). The epoxy matrix was composed of bisphenol-A diglycidyl ether (DGEBA) and fatty acids, while for the chemical recycling, ethylene glycol (EG) was used at elevated temperature. They stated that the recycling method is based on the dissolution and repolymerisation of the CAN-containing epoxy resin via transesterification. While the transesterification is a reverse reaction, excess EG should be used. After soaking and dissolving the vitrimer at elevated temperatures (in this case, approx. 150 °C above T_g), the EG solvent was evaporated, pushing the reverse reaction to the repolymerisation way, therefore recovering the epoxy resin. Their results showed that the thermomechanical performance of the repolymerised epoxy was close to that of the original, confirming partial retention of matrix quality. They also confirmed that the process works not only in the case of neat epoxy resin samples but also in carbon fibre-reinforced composites. The epoxy matrix was dissolved using the optimised parameters (180 °C, 4 h), and an excess amount of EG solvent and the carbon fibres were reclaimed. The dissolved polymer solution was further heated to evaporate the excess EG, and this polymer solution was used as a matrix bath for the recovered carbon fibres. After another 10 h of heating at 180 °C, the EG was fully evaporated, and recycled CFRP composite was made.

Liu et al. [22] made carbon fibre-reinforced polymer composites based on epoxy vitrimers. First, they designed a self-catalytic epoxy prepolymer with built-in hydroxy and tertiary amine groups. Then, anhydride was added to form a catalyst-free epoxy vitrimer. The prepolymer was based on excess DGEBA and a diamine, and the hydroxyls and tertiary amine groups catalysed the curing and the dynamic transesterification in the crosslinked resin; thus, no further catalyst was needed. They used this vitrimer system as the matrix material for carbon fibre-reinforced composites and investigated their mechanical properties. They stated that the matrix can be efficiently hydrolised in pure water at 160–180 °C in a reactor in 5 h.

Memon et al. [44] fabricated carbon fibre-reinforced composites with an imine-based vitrimer matrix, with (i) the hot-pressing of uncured prepregs and (ii) hot-pressing cured prepregs. In the carbon

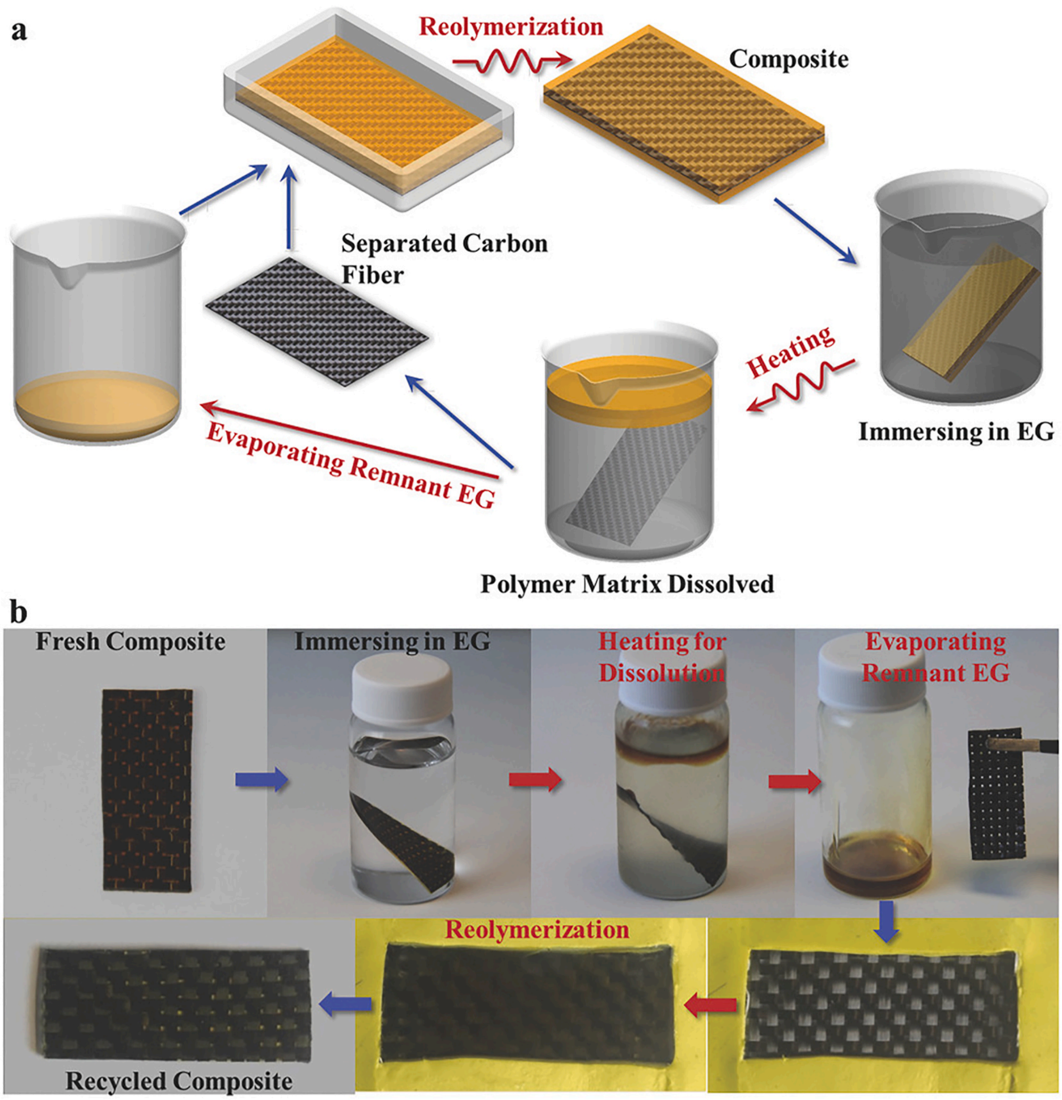

Fig. 15. The closed-loop recycling paradigm for CFRP composites. (a) A schematic view. (b) The experimental pictures [13].

fibre-reinforced composites, the matrix could be fully dissolved in EDA through amine–imine exchange reactions. Subsequent washing with ethanol and drying resulted in soft reclaimed carbon fibre fabrics with no visible epoxy residues or fibre damage based on their scanning electron microscopy analysis. They also stated that not only the reinforcement and the vitrimer could be reclaimed by their process, but also the solvents (EDA and ethanol) could be recycled via distillation and condensation. This separation of components and recovery of not only the monomers but also the solvents by distillation can likewise be applied to other types of materials, making the chemical recycling of vitrimer composites more sustainable.

Yu et al. [54] investigated the repairability, reprocessability and recyclability of carbon fibre-reinforced composites based on a polyimine vitrimer matrix. They used two methods to prepare the composites: a liquid-based and a solid-based method according to the state of matter of the polyimine. The polyimine matrix was soaked in a solution containing diamine and ethanol to initiate the depolymerisation; then, the matrix was fully dissolved after the composite was placed into DETA and ethanol for 24 h at room temperature. They could retrieve the carbon fibres with approx. 98 % original fibre's weight. In the case of the liquid-based method, the recycled composite was made directly from the polyimine oligomer solution, while in the case of the solid-based process, the solution was cured into polyimine and ground into a powder. This powder was later used during compression moulding to prepare recycled composites. Mechanical tests showed that Young's moduli and the tensile strength of the recycled samples were comparable to the original composites, both in the case of the liquid and the solid-based methods.

Lorwanishpaisarn et al. [32] developed a novel, bio-based vitrimer system from DGEBA epoxy, citric acid (CA) and cashew nut shell liquid (CNSL), where the major ester linkages were produced from the curing reaction between the epoxy and CA, while the reaction between epoxy and CNSL was responsible for ether and secondary ester linkages. They also changed the acid-to-epoxy ratio in the system from 0.30 to 0.40; the increase of CA resulted in higher storage modulus, glass transition temperature, crosslink density, and activation energy for transesterification. They found that the vitrimer transition temperature (T_v) was lower than T_g in each case; therefore, the topology rearrangement is controlled by T_g. They also prepared carbon fibre-reinforced vitrimer composites and chemically recycled them using DMF. The recovered carbon fibres were used as fibre reinforcement in vitrimer composites and compared to the samples made with the pristine composites. They found that the composites containing reused carbon fibres had comparable tensile strength and thermomechanical properties to the vitrimer composites cured by a petroleum-based curing agent.

Wang et al. [37] prepared imine-based epoxy vitrimer composites

Table 4

Potential defects, along with their causes and effects, arising during mechanical and chemical recycling of vitrimers and their composites.

Recycling Method	Defect	Cause	Effect
Mechanical	Incomplete network reformation	Insufficient bond exchange during reprocessing (e.g. low temperature or short time)	Weak interfaces, inhomogeneous crosslink density, reduced strength
	Chain scission or backbone degradation	Excessive thermal or mechanical stress (e.g. high grinding energy)	Irreversible molecular weight reduction, impaired strength and elasticity
	Contamination (moisture, dust, residues)	Exposure to ambient conditions during grinding or hot-pressing	Plasticisation, poor crosslinking, especially in imine/ transesterification systems
	Void formation, poor consolidation	Inadequate pressure or degassing during remoulding	Pores, weak bonding, reduced mechanical integrity
Chemical	Incomplete matrix depolymerisation	Mild conditions, insufficient reaction time or or suboptimal temperature	Contaminated fibres, incomplete monomer recovery
	Side reactions or degradation	Oxidation, hydrolysis, or heat effects	Altered monomer structure, limits re-synthesis or reduced final quality
	Loss of monomer purity	Difficulty in selectively recovering only the original monomers	Variation in chemical composition, altered properties of reprocessed material
	Fibre contamination or surface degradation	Harsh solvents damaging sizing or fibre surface	Reduced fibre–matrix adhesion, especially with glass or natural fibres
	Residual solvent retention	Inadequate rinsing/drying of fibres or oligomers	Porosity, weakened mechanical performance during reprocessing

with carbon fibres and they analysed the possibilities of reclaiming carbon fibres during chemical recycling. Carbon fibres were retrieved using 0.1 M HCl/DMF solution at room temperature and then washed with dichloromethane. The SEM images showed that the recycled fibres had a smooth and clean surface; therefore, no residual matrix was attached to the fibres after the recycling process. They also conducted Raman spectroscopy on the virgin and recycled fibres, and they found out that the chemical structure of the carbon fibres did not alter.

Despite the promising research reviewed in this section, a breakthrough in industrial implementation has yet to be achieved. To support progress towards more scalable and robust recycling strategies, we have compiled and analysed the key technical challenges encountered during vitrimer recycling. These challenges—summarised in Table 4—highlight the possible defects, their causes, and their effects observed during both mechanical and chemical reprocessing, including closed-loop recycling as well.

Several studies [44,45,57] reviewed in this section have investigated the mechanical reprocessing of vitrimer composites, often facing challenges such as fibre shortening, loss of structural integrity, reduced fibre orientation, and ultimately diminished mechanical properties—leading to a form of quasi downcycling. However, in some cases [45], short-fibre composites have shown improved performance after multiple recycling cycles, as repeated curing can enhance their properties to some extent. Importantly, hot-pressing enabled by covalent adaptable networks allows vitrimer composites to be repurposed for applications closer to their original use, whereas mechanical recycling of conventional thermosets typically results in downcycling into low-value fillers, such as those used in concrete [90]. During mechanical reprocessing, several potential defects may arise (Table 4). Incomplete network reformation due to insufficient exchange reactions (e.g., due to suboptimal temperature or time) can lead to inhomogeneous crosslink density and weak interfaces, and consequent lower mechanical performance. Excessive heat or mechanical stress may cause irreversible chain scission, reducing strength and elasticity. Contamination by moisture or dust during grinding or hot-pressing can interfere with crosslinking, particularly in imine- or transesterification-based systems. Furthermore, inadequate consolidation may result in voids that weaken the material.

Other studies [13,21,22,32,36,37,39,42,44–48,51,52,54–57,60,62] have demonstrated the efficient chemical dissolution of the vitrimer matrix, successfully preserving the structural integrity and mechanical performance of the reinforcement fibres. Chemical recycling offers substantially greater potential, as it enables the separation and reuse of both matrix and fibre components. The reclaimed fibre reinforcement largely retains its original structure, allowing for true recycling where new composites of comparable quality can be produced. One challenge, however, is the potential presence of residual solvents in the reinforcement, which may negatively affect reprocessing and can also lead to porosity, resulting in compromised mechanical properties (Table 4). This highlights the importance of thorough rinsing and solvent removal. Incomplete depolymerisation of the vitrimer matrix can leave residues, which may reduce interlaminar shear strength and lower adhesion between the fibres and the matrix. Nevertheless, residues from the vitrimer matrix are generally less problematic due to the presence of dynamic covalent bonds, which allow the new vitrimer matrix to form bonds with any remaining material. However, matrix residue can cause the reinforcement to become stiffer and more brittle, making it more difficult to process. Scanning electron microscopy (SEM) analyses have shown no significant differences in surface quality between virgin and reclaimed fibres—particularly carbon fibres—suggesting that the solvents typically used in vitrimer FRP recycling do not structurally damage the fibres. These solvents may damage or remove the fibre sizing, potentially reducing interfacial adhesion between the recycled fibres and the vitrimer matrix during reprocessing. Consequently, re-sizing of the fibres may therefore be necessary, representing a future industrial challenge, especially in the case of mixed reclaimed fibres, where assessing the extent of sizing loss can be difficult.

While most research has focused on the quality of the recycled fibres, a few pioneering studies have achieved full closed-loop recycling, in which the original vitrimer matrix is re-synthesised from monomers recovered during chemical recycling [13,36,51]. These examples are critical in highlighting the advantages of chemical recycling over conventional methods, such as pyrolysis, which is commonly used to produce high-quality carbon fibres but does not allow matrix reuse. In the case of closed-loop recycling, several problems may arise as well (Table 4). Incomplete depolymerisation limits monomer recovery, while side reactions may alter the chemical structure of the recovered components, reducing their suitability for reuse. Additionally, loss of monomer purity and fibre surface degradation—particularly in harsh chemical environments—can hinder the regeneration of high-performance composites. Solving these problems and demonstrating closed-loop recycling routes will be crucial to the development of truly sustainable composite recycling systems.

4. Summary and outlook

This review provides a comprehensive overview of the recycling methods for vitrimer resins and their fibre-reinforced composites, with a focus on the dynamic covalent mechanisms that enable such recyclability. This section focuses on the critical findings, knowledge gaps, and future directions emerging from the reviewed literature.

Mechanical recycling of vitrimers, while relatively simple and energy-efficient, remains under-optimised. Although many studies report retained mechanical properties across multiple cycles, few provide detailed insight into the effects of particle morphology, processing conditions, or moisture sensitivity. Grinding-induced defects such as localised chain scission or surface degradation are rarely investigated in depth. To support reproducibility and industrial applicability, future work should develop standardised reprocessing protocols and clarify the role of reprocessing parameters in crosslink density and performance recovery.

Chemical recycling offers the potential for true circularity, especially when monomers or oligomers are recovered in high yield and reused. Imine-, disulfide-, and ester-based vitrimers have shown promising results, yet side reactions, incomplete depolymerisation, or solvent retention remain practical obstacles. Additionally, the trade-off between recyclability and durability poses a design challenge: vitrimers must maintain resistance to hydrolysis or thermal degradation in use, while still enabling controlled depolymerisation at end-of-life. Incorporating multiple exchange mechanisms or designing smart, switchable systems could help navigate this compromise. Additionally, while high thermal stability and elevated glass transition temperatures are essential for structural applications, they pose challenges in terms of energy demands and sustainability when it comes to repairing and chemically recycling vitrimers with higher transition temperatures.

Regarding the recycling of fibre-reinforced composites, mechanical recycling offers simplicity but often leads to reduced fibre lengths and diminished mechanical properties over successive cycles. In contrast, chemical recycling has shown greater potential for material recovery, particularly in preserving the integrity of carbon fibres and recovering monomers. However, the limited demonstration of true closed-loop recycling, where the recovered matrix is reconstituted into new vitrimer composites with equivalent performance, remains a key barrier. Future efforts should focus on advancing scalable, cost-effective, and energy-efficient closed-loop recycling techniques that integrate seamlessly into existing composite manufacturing workflows and significantly enhance the sustainability of vitrimer composites, reduce the amount of composite waste and the demand for virgin materials. With continued innovation, vitrimer composites could play a crucial role in the development of fully sustainable, high-performance materials for various industrial applications.

CRediT authorship contribution statement

Andrea Toldy: Writing – review & editing, Writing – original draft, Visualization, Supervision, Resources, Project administration, Methodology, Conceptualization. **Dániel István Poór:** Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Norbert Geier:** Writing – original draft. **Ákos Pomázi:** Writing – review & editing, Writing – original draft.

Statement

During the preparation of this work the authors used ChatGPT and Grammarly in order to improve the language and readability of the paper. After using this tool, the author(s) reviewed and edited the content as needed and take full responsibility for the content of the published article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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