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Pathways towards a circular economy for thermoset nanocomposites

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The growing adoption of thermoset nanocomposites across industries has raised concerns about their environmental impact and end-of-life management. These advanced materials, which combine nanoscale fillers with permanently crosslinked matrices, pose unique recycling challenges due to their inherent durability and resistance to conventional mechanical recycling methods. This perspective examines prospective approaches for achieving circularity in thermoset nanocomposites, focusing on two main strategies: reprocessing and nanofiller recovery. We analyse recent developments in the use of covalently adaptable networks (CANs) as reprocessable matrices for nanocomposites. We also evaluate physical, thermal, and chemical methods for recovering valuable nanofillers from thermoset nanocomposite waste. While significant progress has been made in developing recyclable matrices and recovery techniques for conventional composites, additional research is needed to address challenges specific to nanomaterials, including preventing nanomaterial agglomeration and surface chemistry preservation. Moving forward, tailored approaches considering the unique characteristics of different nanofiller types will be crucial for establishing effective recycling protocols and advancing towards a circular economy for these high-performance materials.

Introduction

Over the last several decades, nanocomposites have emerged as a critical class of materials that find use in nearly every sector. These multiphase materials combine a nanoscaled filler phase with a continuous matrix phase, giving rise to properties that are markedly different from the sum of their bulk counterparts (Fig. 1). Nanocomposites are utilized in the aerospace sector in high-performance components and flame-retardant panels;[1] in the automotive industry for panels, gas tanks, and bumpers;[2] in construction for structural panels and building sections;[3] in electronics for printed circuit boards and electrical components;[4] as well as in food packaging,[5] healthcare,[6] energy systems,[7] consumer goods,[8] and more. The nanoscale dimensions and exceptionally high surface-to-volume ratio of the nanofiller phase[9] impart nanocomposites with enhanced properties that vastly outperform conventional composites. Furthermore, by selecting filler phases with specific functional properties, nanocomposites can be designed to have enhanced electrical conductivity,[10] barrier properties,[10] fire retardancy,[11] mechanical properties[12,13] thermal stability,[14] solvent resistance,[15] optical properties[16] and catalytic capabilities.[17] All of these properties can be tailored by altering the local chemistry, mobility, distribution, surface characteristics, dispersion state,

morphology, and crystallinity of the nanofiller phase.[18–22] Nanocomposites can also be engineered to combine several of these properties, making them versatile multifunctional materials.

Nanocomposites can be broadly categorized based on their matrix type: ceramic matrix nanocomposites, metal matrix
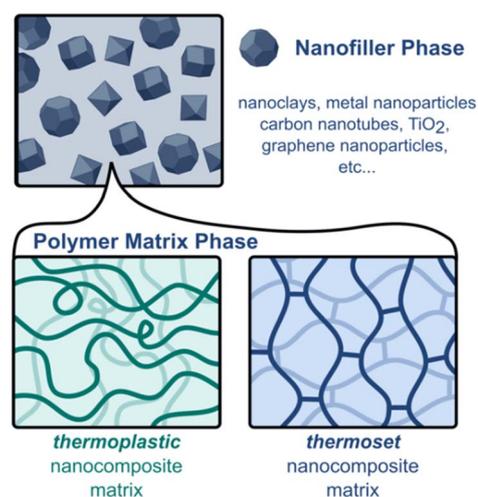

Fig. 1 Structure of polymer nanocomposites. In polymer nanocomposites, the filler phase consists of nanoparticles dispersed in a thermoset or thermoplastic polymer matrix. The nanofiller phase ranges from clay to metal nanoparticles, carbon nanotubes, titanium oxide (TiO_2), graphene nanoparticles, and more.

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nanocomposites, or polymer matrix nanocomposites.[23] Due to their cost-efficiency, lightweight, processability, and the versatile range of nanofillers that they are compatible with, polymer nanocomposites have grown into the most widely applied nanocomposites. Polymer nanocomposites can consist of either a thermoplastic or crosslinked thermoset matrix (Fig. 1) loaded with modest amounts of nanofiller (typically <5% by mass).[24] A wide range of nanofiller materials are used in polymer nanocomposites, from clay,[25–29] to silica,[30–33] titania,[34–38] carbon nanotubes (CNTs),[39–42] graphene,[43–47] and polyhedral oligomeric silsesquioxane (POSS).[48–52] Thermoplastics nanocomposites are currently the most established and widely applied polymer nanocomposites. Yet, the exceptional properties of thermoset nanocomposite matrices, including epoxy resins,[53] polyester resins,[54] phenolic resins,[55] and polyimides[56] have gained increasing attention in recent years. Their easy processing, compatibility with heterogeneous materials, considerable solvent and creep resistance, and higher operating temperatures make thermoset nanocomposites desirable for many high-performance applications in the chemical, construction, transportation, and aerospace industries. The exceptional mechanical, thermal, and electrical properties of thermoset nanocomposites may even allow them to act as lightweight and processable replacements for metals in certain applications.[57]

Critically, the excitement surrounding thermoset nanocomposites comes along with critical concerns surrounding their sustainability. The thermoset matrix is permanently crosslinked and highly resistant to degradation, rendering them environmentally persistent. Furthermore, thermosets do not melt and flow when heated and therefore cannot be recycled through conventional mechanical recycling. As a result, end-of-life thermoset waste accumulates in landfills and the natural environment. The valuable nanofiller phase is trapped in the permanent thermoset network and cannot be recovered, resulting in the loss of these valuable and resource-intensive advanced materials as waste. There is clearly a need to improve the end-of-life management of thermoset nanocomposites.

Here we provide perspective on the need for a circular economy for thermoset nanocomposites, while also highlighting the progress and challenges associated with achieving that goal. Notably, the recovery and recycling of carbon fibers from conventional composites have been investigated at length and thoroughly reviewed elsewhere.[58–60] Yet, recycling nanoparticle fillers from nanocomposites remains a relatively unexplored frontier. We start by discussing the economic and environmental motivations that necessitate a circular economy for nanocomposites. We then discuss emerging strategies for achieving a circular economy for nanocomposites, from dynamic thermoset matrices that can be reprocessed and mechanically recycled, to approaches to extract and recover the nanofiller phase from thermoset matrices, allowing these valuable materials to be recycled. We hope that this perspective will highlight the importance of this endeavor and guide innovation toward achieving a circular economy of these advanced materials.

Motivation for a circular economy for thermoset nanocomposites

The growing adoption of thermoset nanocomposites, while beneficial for numerous applications, raises concerns about both their impact on the environment and the potential economic loss of valuable resources. These concerns serve as the core motivation for developing strategies for the effective recovery and reuse of nanomaterials. The following subsections delve into the environmental and economic factors that underscore the importance of achieving a circular economy for nanocomposites.

Environmental motivation

We will start by analyzing the environmental impact of thermoset nanocomposites from two perspectives: the emissions and climate change contribution associated with their production and the ecological hazards associated with their end-of-life waste.

Notably, nanocomposites as commercial materials are relatively novel, and comprehensive information about their environmental impact is not readily accessible, particularly when focusing solely on thermoset nanocomposites. However, recent reports provide valuable analysis that we will highlight here.[61]

Environmental impact of thermoset nanocomposite production

Given the nascent nature of the market, data on the environmental impact of thermoset nanocomposite production remains very limited. In place of comprehensive data on the entire class of materials, we will focus on a representative thermoset nanocomposite as a case study to illustrate the impacts of their production: epoxy matrix with carbon nanotube (CNT) nanofiller. From 2021 to 2023, the production of thermoset nanocomposites with epoxy matrices increased from 39.7 kilotons to 51.7 kilotons, demonstrating growing commercial interest (Fig. 2). More notably, the production growth from 2021 to 2022 was measured at 11.5%, while for 2022 to 2023 it reached 16.7%, further emphasizing the positive trend. This rapid production growth underscores the importance of sustainable practices in their production. While commonly used commercial nanofillers in epoxy nanocomposites range from nanoclays to CNTs, nanofibers, and nano-oxides, here we focused on CNT nanofillers, as there is available data on their global warming potential (GWP).

In CNT/epoxy nanocomposites, the loading of CNTs varies from 0.1%[62] to 68%.[63] Life cycle analyses of carbon nanotubes reveal that their global warming potential (GWP) can exceed 7×10^5 kg CO_2-eq. per kg.[64,65] Notably, this value is highly dependent on the production method and application of the CNTs (Fig. 3a). This translates to a GWP contribution from CNT nanofiller production alone ranging from approximately 480 to 10^6 kg CO_2-eq. per kg of CNT/epoxy nanocomposite production (Fig. 3a). In contrast, the epoxy matrix has a GWP of less than 6.1 kg CO_2-eq. per kg.[66] Furthermore, more than 99.1% of the GWP
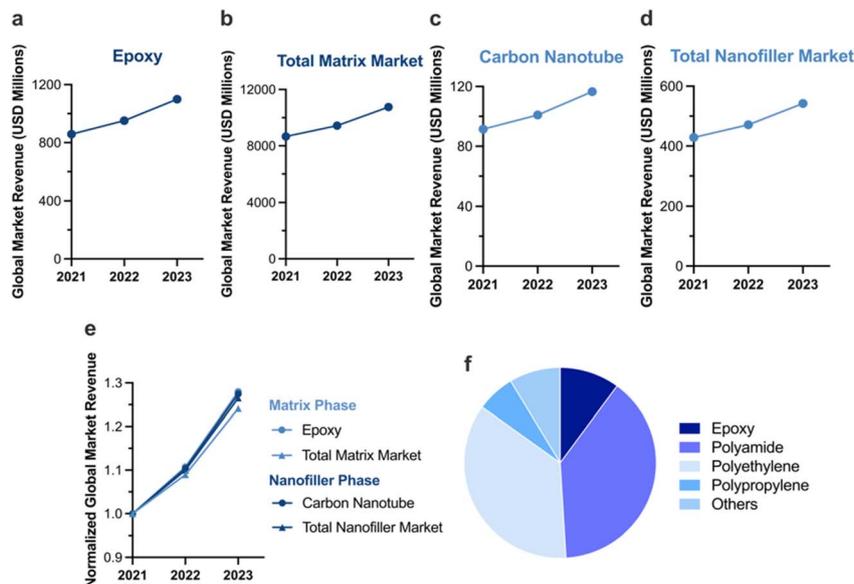
Fig. 2 Global nanocomposite market trends. (a–d) Trend in global market revenue in USD millions from 2021–2023 for the epoxy thermoset matrices (a), the total matrix market (b), carbon nanotube nanofillers (c), and the total nanofiller market. (e) Normalized increase in global market revenue from 2021–2023 for the matrix and nanofiller markets. Data in (e) is normalized to 2021 data. (f) The relative share of different matrix types of the global market revenue of nanocomposite matrices. Data in (a–f) is from ref. 61.
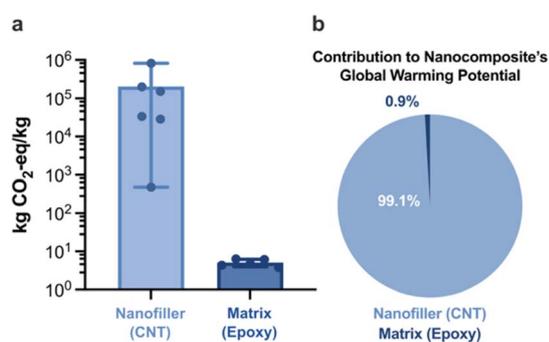

Fig. 3 Global warming potential of nanocomposite production. (a) Estimated kg CO_2-eq. per kg associated with the production of a representative nanocomposite: carbon nanotube (CNT) filler and epoxy matrix. Error bars and individual points represent the range of reported data for CNTs produced through different methods.[64–66] (b) The estimated relative contribution of the nanofiller and matrix phases to the overall global warming potential (kg CO_2-eq. per kg) of nanocomposites, based on the case study of CNT-epoxy composites. Data in (a and b) from ref. 64–66.

of epoxy/CNT nanocomposites comes from the CNT nanofiller phase (Fig. 3b). Therefore, recovery and recycling of nanofillers should be a central focus of efforts to reduce the environmental impact of thermoset nanocomposites like CNT/epoxy.

Environmental impact of thermoset nanocomposite end-of-life waste

While incineration and landfilling are the predominant disposal methods for thermoset nanocomposites, primarily due to their relative simplicity and cost-effectiveness, they come along with significant environmental concerns.[67] For example,

landfilled nanocomposites can leak into the natural environment and pose a significant ecological hazard. Thermosetting resins are highly resistant to biodegradation and can persist in the environment for decades to centuries.[58] Moreover, the small molecules resulting from the polymer's carbon skeleton decomposition are often biotoxic, posing considerable environmental risks when directly discharged into the soil.[68] Furthermore, many of the commonly utilized nanomaterials in the nanofiller phase, particularly carbon nanotubes, are associated with risks to both human and ecological health. For example, certain types of carbon nanotubes are cytotoxic and have been implicated in several human cancers. Recognizing these potential hazards, governmental agencies, including those in the United States and the European Union, have enacted policies and regulations to manage some thermoset nanocomposite waste disposal, including carbon nanofiber thermoset composites.[58,69,70] This necessitates a shift toward recycling and other innovative approaches that limit the accumulation of nanocomposite waste.

Incineration can eliminate concerns surrounding nanocomposite waste accumulation. Furthermore, the energy associated with incineration can be captured for electricity generation. Yet, from an economic perspective, the incineration of nanocomposite waste for electricity generation is imprudent, given the energy-intensive nature of the incineration process and the significantly higher value of nanomaterials compared to ordinary plastics. Furthermore, temperature-resistant nanomaterials can persist in fly and bottom ash products, creating secondary waste streams that still contain engineered nanomaterials. These nanoparticles can enter organisms through ingestion or inhalation, potentially causing toxic effects in organs and tissues.[71,72] Combustion also produces numerous

pollutants, including volatile organic compounds (VOCs), sulfur oxides, nitrogen oxides, carbon monoxide, and carbon dioxide, exacerbating environmental concerns.[73] These factors collectively emphasize the importance of recovering nanomaterials from thermoset nanocomposites.

While mechanical recycling nanocomposites by reprocessing presents a potentially superior approach to incineration and landfilling, it still faces significant obstacles. As will be discussed below, one of these drawbacks is the tendency of nanomaterials to aggregate under mechanical reprocessing conditions. This aggregation leads to poor dispersion of nanomaterials in the matrix of the recycled and reprocessed thermoset nanocomposite. Furthermore, exposure to these harsh conditions degrades the matrix and can result in the release of nanomaterials from the nanocomposite, potentially causing environmental and health hazards.[74–76]

In light of these considerations, nanomaterial recovery emerges as the most promising approach to mitigate the environmental impact of thermoset nanocomposite production and end-of-life management. This strategy not only addresses the challenges associated with disposal but also offers an opportunity to reclaim valuable materials for reuse in nanocomposite production, thereby moving towards a more circular and sustainable materials economy.

Economic motivation

Next, we will discuss the economic motivation for recovering and recycling nanofillers from thermoset nanocomposites. As discussed above, the nanocomposite market has experienced significant growth over the last several years, with the global market revenue for the matrix and nanofiller phases increasing by 24.1 and 26.5%, respectively, between 2021 to 2023 (Fig. 2a–e).[61] Notably, thermoset matrices account for a significant portion of this growth, as 10.1% of the global market revenue for nanocomposite matrices comes from epoxy resins alone (Fig. 2c). These epoxy matrices, in addition to the carbon nanotube fillers in the case study discussed above, have also shown market growth in recent years (Fig. 2a and e). Increasing the recycling of nanofillers can mitigate production costs and allow for the high value of the nanofiller phase to be recaptured in the economy, rather than disposed of. This approach follows the principles of the circular economy, which is an economic model designed to eliminate waste and maximize resource efficiency. It is based on the principles of eliminating waste and pollution, circulating products and materials at their highest value, and regenerating natural systems.[77] In contrast to the traditional linear economy model of "take-make-dispose", the circular economy emphasizes the importance of keeping resources in use for as long as possible, extracting the maximum value from them while in use, and then recovering and regenerating products and materials at the end of their service life.[78]

These circular economy principles are particularly relevant to thermoset nanocomposites. Many nanomaterials used in these composites, such as carbon nanotubes,[79,80] graphene,[81,82] and metal nanoparticles,[83,84] are valuable and often derived from scarce raw materials. Furthermore, the production of nanomaterials is typically energy and resource-intensive. For instance, the production of carbon nanotubes often requires rare metal catalysts and high-energy processes.[85,86] Similarly, metal nanoparticles used in nanocomposites may contain precious metals or strategically important elements, including rare earth metals.[87] By recovering these nanomaterials from end-of-life thermoset nanocomposites, we can reduce the need for virgin raw material extraction, thereby conserving valuable resources. This approach also decreases the energy consumption and associated costs of producing new nanomaterials, mitigates the environmental impact of both raw material extraction and nanomaterial production, and creates a secondary market for recovered nanomaterials, potentially reducing the overall cost of nanocomposite production. Moreover, as regulations around waste management and resource conservation become more stringent, companies that can effectively recover and reuse nanomaterials may gain a competitive advantage. This could come in the form of reduced material costs and better alignment with emerging environmental policies.

The economic value that can be recovered from nanomaterials in thermoset nanocomposites is substantial. For example, incorporating carbon nanotubes into materials, which can cost usually from $20 to $2000 per gram depending on their quality and specifications, represents a significant economic investment.[88] Recovering even a fraction of these materials could result in considerable cost savings and resource conservation for nanocomposites.

In sum, the recovery and recycling of nanomaterials offers a pathway to retain the economic value of these advanced materials, reduce the environmental impact of their production, and move towards a more sustainable model of nanocomposite manufacturing and use.

Prospective pathways towards a circular economy for nanocomposites

So far, we have discussed that as production and use of thermoset nanocomposites continue to grow, so does the need to develop effective recycling methods that align with circular economy principles. The unique extreme stability that makes thermoset nanocomposites valuable in various applications also presents significant challenges for their end-of-life management.

Here we present and discuss two prospective approaches that for recycling thermoset nanocomposites: reprocessing the nanocomposite as a whole material, and extracting and recovering the valuable nanomaterials from the thermoset matrix. While neither method has been widely implemented on an industrial scale, both are being investigated as potential strategies to reduce waste, conserve resources, and minimize the environmental impact of these advanced materials. However, each approach faces its own set of challenges and limitations that currently hinder widespread adoption.

In this section, we will examine these two potential routes to circularity for nanocomposites in detail, evaluating current data on their effectiveness, limitations, and potential. By critically analyzing the current state of nanocomposite recycling, we can better understand the progress made thus far and the hurdles that remain in closing the loop for these advanced materials.

Reprocessable thermoset nanocomposites – covalent adaptable networks

Plastics can be mechanically recycled by reprocessing at elevated temperatures and mechanical force. This is achieved with widely available and established polymer processing tools, including extrusion, injection molding, compression molding, and more (Fig. 4). While reprocessing could provide a pathway to a circular economy for thermoset nanocomposites, the thermoset matrix is a permanent covalently crosslinked network that cannot flow or be reprocessed at elevated temperatures. One approach to overcoming the permanence of thermoset matrices is incorporating dynamic covalent bonds into the polymer network that are reversible under specific conditions.[89,90] A variety of stimuli can be used to induce bond exchange in these dynamic materials, including the high temperatures and mechanical forces associated with mechanical recycling and polymer reprocessing.[89] Leveraging these dynamic thermosets, which are called covalently adaptable networks (CANs), as the matrix in nanocomposites provides a pathway to preparing reprocessable and recyclable nanocomposite thermosets (Fig. 5).

A key advantage of CANs is their ability to combine the strength and stability of thermosets with the recyclability and processability of thermoplastics. Notably, the bond exchange in CANs can be either dissociative or associative (Fig. 5a). During associative bond exchange, the crosslinking density is maintained, while dissociative exchange results in a loss in crosslinking density after reprocessing. As a result, CANs that undergo associative exchange, called vitrimers, better maintain the advantageous properties of thermosets after reprocessing. As such, there has been growing interest in the potential of
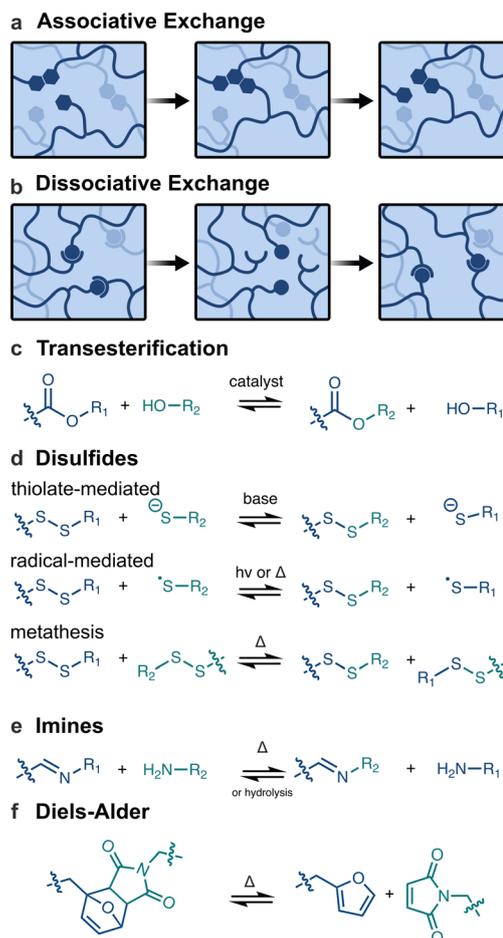

Fig. 5 Covalent adaptable networks as matrices for reprocessable nanocomposites. Covalent adaptable have dynamic covalent cross-links that can be exchanged through an (a) associative mechanism (vitrimers) or (b) dissociative mechanism. (c–f) Examples of dynamic covalent chemistries that are utilized in covalent adaptable networks include (c) transesterification, (d) disulfides exchange, (e), imine exchange and (f) Diels−Alder chemistry. Disulfide exchange in (d) can occur through thiolate-mediated, radical-mediated, or metathesis-mediated mechanisms.
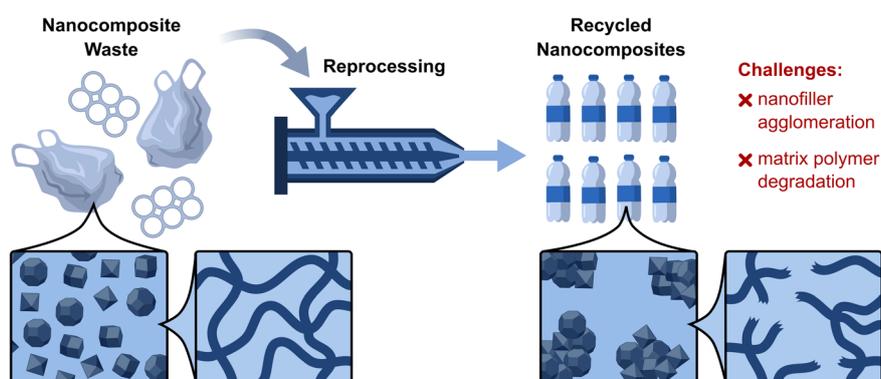
Fig. 4 Nanocomposite reprocessing. Reprocessing nanocomposite waste as a whole material through mechanical recycling (e.g. re-extrusion, as illustrated) into recycled nanocomposite materials offers one pathway toward a circular economy for nanocomposites. Challenges associated with reprocessing include the agglomeration of nanofillers during reprocessing and degradation of the matrix through chain scission, which compromises nanocomposite properties.

Table 1 Examples of reprocessable nanocomposites with covalent adaptable network matrices

	Matrix material	Nanofiller type	Reprocessing method	Ref
Dynamic chemistry				
Transesterification	EMA[a]	Nanoclay + rGO[b]	Ball milling + hot pressing + laser irradiation	138
		CNF + nanoclay	Hot pressing	139
	Epoxy vitrimer	Nanoclay + MXene	Ball milling + hot pressing	140
		Cellulose nanocrystals	Ball milling + hot pressing	141 and 142
		Graphene nanoplates	Hot pressing	143
		Silica nanoparticles	Welding/joining	144
		Silica nanoparticles	Welding/reshaping	145
		MWCNTs[c]	Hot pressing + welding	146
		MWCNTs	Photo-welding	147
	Epoxidized natural rubber	Carbon nanodots	Hot pressing	148
		MWCNTs	Hot pressing	149
Boronic ester exchange	Carboxyl-grafted styrene–butadiene rubber	Epoxy group-functionalized silica	Hot pressing	150
	Polystyrene-b-poly(ethylene-co-butylene)-b-polystyrene	Silica nanoparticles	Hot pressing	151
Transcarbamoylation	Polyurethane vitrimer	Carbon nanotubes	Compression molding	152
Trans-N-alkylation (pyridine-bromo quaternization)	Crosslinked polyacrylate thermoset	Silica nanoparticles	Hot pressing and thermal reshaping	153
Imine bonds	Styrene–butadiene rubber	Amino-functionalized graphene	Hot pressing	154
Transamination	Poly(butyl methacrylate-co-2-(acetoacetoxy)ethyl methacrylate) vitrimer	Fe_3O_4 magnetic nanoparticles	Induction heating (electromagnetic field)	155
Dual dynamic systems				
Transesterification + Fe^{3+} coordination bonds	Carboxylated styrene–butadiene rubber	Epoxy-modified tunicate cellulose nanocrystals	Hot pressing	156
Transcarbamoylation + cyclic carbonate aminolysis	Polyhydroxyurethane	Silica nanoparticles	Hot pressing	157

[a] EMA: poly(ethylene glycol methacrylamide)-co-hydroxy(ethylene glycol) methacrylate-co-N-(2-hydroxyethyl)methacrylamide. [b] rGO: reduced graphene oxide. [c] MWCNTs: multiwalled carbon nanotubes.

vitrimers, and CANs in general, to serve matrices for thermoset nanocomposites that allow them to be reprocessed and recycled without compromising their valuable properties. Yet, the maximum application temperature for CANs is limited by the temperature at which they are designed to be dynamic, not by their degradation temperature, which can limit adoption of CANs as matrices for nanocomposites. Hence, this design dilemma restricts CANs' application range in comparison with classic thermosets nanocomposite matrices, which are suitable for many high temperature uses.[91]

Many different exchangeable bond chemistries have been used in the preparation of CAN matrices for thermoset nanocomposites, from transesterifications to Diels–Alder chemistry, imines, disulphides, and more. Several of these examples are listed in Table 1. These dynamic matrices have been leverage to prepare thermoset composites with wide variety of nanomaterials as nanofillers including carbon nanotubes, graphene, nanosilica, nanoclay, MXene, nanosilver, *etc.*[92] The following sections will delve into these chemical approaches, exploring their mechanisms, advantages, and potential applications in recycling nanocomposites.

Transesterification

Chemistry. Transesterification is a classic vitrimer exchange reaction that involves the exchange esters with free alcohols (Fig. 5). In the context of recyclable thermoset systems, this process is typically applied by grafting reversible β-hydroxyl esters onto an epoxy matrix. At elevated temperatures and in the presence of a catalyst, dynamic associative exchange occurs between esters and hydroxyl groups through the transesterification reaction (TER).[93] The incorporation of TER exchange into thermoset systems provides a mechanism for network rearrangement, allowing for the reprocessing and recycling of these otherwise permanent structures. Reprocessing of these vitrimers involves the application of external force at high temperatures to initiate TER. This thermal activation triggers a rearrangement of the crosslinked network, allowing the material to be reprocessed.[94]

Several key factors influence the rearrangement and exchange processes in these TER-vitrimer systems. These include the stiffness of the polymer backbone, the density of crosslinks, the operating temperature, and the nature and concentration of the catalyst. For instance, insufficient catalyst loading, or the use of a less effective catalyst can result in a TER that is too slow to enable reprocessing. Conversely, a decrease in backbone stiffness enhances the mobility of polymer segments, which can accelerate the TER. Temperature also plays a crucial role: at temperatures below the glass transition temperature (T_g) of the material, the crosslinked network remains rigid and immobile, hindering TER exchange. As the temperature increases above T_g, it activates and accelerates the TER, allowing for more efficient network rearrangement. Understanding and optimizing these factors is essential for designing effective recyclable thermoset nanocomposites with desired properties and processing characteristics.[95]

While the TER offers significant advantages in thermoset recycling, it also presents certain challenges. Typically, metal-based catalysts such as zinc acetate and zinc acetylacetonate are employed to accelerate the TER process. However, these metal catalysts can introduce undesirable effects, including potential toxicity and material instability in the final product. These concerns can be mitigated to an extent by using of hyperbranched thermoset polymers rich in free hydroxyl groups which can enhance both the curing process and the transesterification reaction without the need for metal catalysts.[96] Vitrimers based on TER also face other limitations, including poor thermal stability. Additionally, the reprocessing conditions for these materials often remain stringent, requiring precise control of temperature and other parameters.[97]

Use as nanocomposite matrices. There is growing literature specifically addressing the application of TER in nanocomposite materials. For example, Chen *et al.* demonstrated a particle fusion recycling approach in which a nanocomposite was ground into small particles and subsequently thermally compressed to enable particle integration and reformation into the desired shape through TER.[98] In that study, epoxide-modified silica nanoparticles in hard epoxy vitrimer matrices were used, allowing interfacial covalent binding between functionalized silica and the vitrimer network to improve nanoparticle dispersion. Furthermore, the surface of the nanoparticles was able to participate directly in transesterification reactions. At higher silica loadings, the nanocomposites exhibited unique microphase separation with distinct hard and soft phases, achieving enhanced mechanical properties while preserving reprocessability.

Critically, their recycling approach revealed significant limitations inherent to transesterification-based hard nanocomposite vitrimer systems. The particle fusion recycling method required harsh conditions including high pressure and extended processing times, making it both energy-intensive and time-consuming. The authors acknowledged that recycling hard vitrimer materials with high modulus presents substantial challenges, as good particle contact is essential for TER-enabled effective welding of the reprocessed material, necessitating high pressures. Furthermore, interfacial chain diffusion and bond exchange reactions remain slow even at elevated temperatures in highly crosslinked materials. Consequently, visible flaws were evident in the recycled samples under their processing conditions, indicating incomplete recovery of material properties. The study highlighted that surface modification of ground vitrimer particles could potentially accelerate interfacial reactions, but this approach requires further development.[98]

Overall, when applying TER to nanocomposite matrices, consideration needs to be given to the unique challenges posed by different nanomaterials, including nanoparticle agglomeration and potential interactions between metal TER catalysts and exchangeable bonds in the nanofiller phase. These limitations demonstrate that while transesterification-based vitrimer nanocomposites show promise, significant advances in processing methods and material design are needed to achieve efficient and practical recycling of these systems. Table 1 summarizes some of the works in this field.

Disulfides

Chemistry. Another dynamic covalent bond widely used in vitrimers is disulfide bonds. Disulfides undergo three general

types of exchange reactions: thiolate-disulfide exchange, radical-mediated disulfide exchange, or concerted metathesis-mediated disulfide exchange (Fig. 5d).[99] A key advantage of utilizing disulfide bonds in vitrimers is the lower activation energy of the exchange reaction, particularly when compared with other processes such as TER. As a result, materials containing disulfide bonds typically require less harsh conditions for reprocessing. This characteristic not only presents opportunities for energy conservation but also minimizes the risk of heat-induced degradation of the matrix and nanofiller phase during nanocomposite recycling. Furthermore, the activation energy and relaxation phenomena can be modulated by adjusting the chemistry of the disulfides. Specifically, aromatic and aliphatic disulfides have different energetic requirements for cleavage, which leads to diverse relaxation phenomena in disulfide-crosslinked materials.[99]

Use as nanocomposite matrices. Disulphide-based vitrimers ability to serve as reprocessable matrices for thermoset nanocomposites has been demonstrated in several recent studies. They have been used to develop reprocessable and mechanically recyclable epoxy composites with filler phases ranging from fibers[100] to Fe_3O_4 nanoparticles,[101,102] graphene and graphene oxide-based nanocomposites,[103,104] and CNTs.[105,106]

A notable example by Lorero *et al.* demonstrated that epoxy vitrimer nanocomposites reinforced with CNTs can be mechanically recycled through milling and hot-pressing, with the resulting recycled nanocomposites maintaining 77–100% of their original mechanical strength. Importantly, CNT addition appeared to catalytically enhance disulfide bond exchange, improving recyclability compared to neat vitrimers, though the milling process affected CNT dispersion quality leading to variable electrical properties in recycled materials. These materials also demonstrated electro-thermal welding capabilities, where electrical heating through the Joule effect enabled joining of components without external heat sources.[105]

Another approach to disulfide-based vitrimer nanocomposites involves the use of polymeric hardeners rather than small molecule crosslinkers. Zheng *et al.* developed epoxy/Fe_3O_4 nanocomposites using poly(α-lipoic acid) as a polymeric crosslinker with disulfide bonds along the main chain. They demonstrated fine dispersion and excellent reprocessability through hot pressing at 130 °C. The nanocomposites exhibited multifunctional properties including shape memory behaviour, photothermal heating capabilities, and magnetic responsiveness, while maintaining mechanical properties after reprocessing cycles. Notably, the incorporation of Fe_3O_4 nanoparticles appeared to enhance disulfide bond exchange, as evidenced by reduced activation energies compared to the control epoxy, potentially due to enhanced thermal conductivity from the inorganic fillers. This study illustrated how the combination of functional nanofillers with disulfide-based vitrimers can create smart materials with multiple responsive behaviors while retaining recyclability, expanding the potential applications of these sustainable composite systems.[101]

Extending beyond traditional composite applications, Zheng *et al.* demonstrated the potential of disulfide-based CANs for sustainable manufacturing through 3D printing. They developed a disulfide-crosslinked polyurethane composite incorporating hydroxylated multi-walled carbon nanotubes (MWCNTs-OH) for direct ink writing (DIW) 3D printing. The dynamic disulfide bonds enabled the material to be printable at elevated temperature, while providing excellent recyclability through simple crushing and reprinting without complex treatment. The incorporation of MWCNTs-OH served dual purposes: enhancing mechanical properties and enabling photothermal conversion for precise *in situ* repair using near-infrared laser irradiation. Remarkably, the printed parts maintained 86.3% of their original mechanical properties after three repair cycles, demonstrating the practical viability of self-healing capabilities. The materials could be recycled multiple times with consistent performance, showing tensile strengths of 9.07 MPa after five recycling cycles compared to 9.12 MPa for the original material. This work exemplifies how dynamic disulfide bond exchange can enable truly circular manufacturing processes for nanocomposites, addressing both end-of-life recyclability and extending service life through repairability, while also demonstrating the practical implementation of CAN-based nanocomposites in advanced manufacturing applications.[107]

Critically, the widespread use of these materials is currently limited by their high production costs, specifically the production cost of the specialized disulfide matrices.[99] Furthermore, the recycling process may affect the dispersion quality of non-functionalized nanofillers, decreasing the quality of the nanocomposite as a whole.

Diels–Alder chemistry

Chemistry. The Diels–Alder (DA) reaction is another commonly applied dynamic covalent bond in CANs that used to prepare reprocessable matrices for nanocomposites. This $[4 + 2]$ cycloaddition reaction occurs between a conjugated diene and a dienophile, forming a cyclic "DA-adduct" (Fig. 5e). At elevated temperatures, the adduct fragments back to the diene and dienophile in a reverse retro-Diels–Alder (rDA) reaction. When incorporated into thermosets, these DA-adducts can undergo dynamic exchange through DA and rDA reactions at elevated temperatures, rendering the materials reprocessable and recyclable.[108–110] Maleimides and furans are the most commonly employed diene–dienophile pair in materials (Fig. 5e). Their widespread use is primarily attributed to the relatively low temperature (approximately 110 °C) required for the rDA reaction to occur. This temperature is significantly lower than typical polymer degradation temperatures, allowing the DA mechanism to serve as a controlled chemical trigger for recyclable macromolecular networks.[111–114]

While DA chemistry enables reprocessing under relatively mild conditions without catalysts or solvents, several limitations exist that may restrict its practical applications in composite sectors. For example, thermoreversible DA resins are unsuitable for applications requiring continuous-use service temperatures above the rDA reaction temperature (around 110–120 °C), due to significant rDA-induced matrix softening.[111,115] Furthermore, the DA/rDA process is dissociative and not fully reversible under practical conditions due to side reactions, which can compromise material's properties post-

recycling.[108,111,115] Overall, further investigation is required to ensure the long-term stability of DA-chemistry-based recycled materials to achieve industrial-scale implementation.

Use as nanocomposite matrices. A number of proof-of-concept studies have recently demonstrated that DA-chemistry-based CANs can serve as reprocessable matrices for nanocomposites. For instance, self-healing and recyclable nanocomposites have been prepared with a DA-adduct containing polyurethane or epoxy thermoset matrices and with nanofillers ranging from halloysite nanotubes (HNTs) to graphene nanoplatelets as the filler phase.[116,117] One of these studies, revealed that, while addition of bismaleimide crosslinkers creates aggregates that hinder CNT dispersion and reduce electrical conductivity compared to conventional epoxy systems, the resulting nanocomposites still maintain predictable Joule heating capabilities. Notably, they demonstrated effective self-healing through electrical stimulation, reducing crack volumes by over 50% when materials were heated to 150–170 °C *via* the Joule effect. The study also suggested that CNT incorporation enhances crosslink density of the network after reprocessing. This was evidenced by the increased enthalpies of the rDA reaction, suggesting that carbon nanotubes participate directly in the dynamic bonding through their DA reaction of the unsaturated carbon–carbon bonds. However, the addition of CNTs reduced chain mobility at elevated temperatures, highlighting the ongoing challenge of balancing mechanical reinforcement with dynamic network properties. These findings demonstrate that while electroactive DA nanocomposites show promise for autonomous repair applications, optimization of filler dispersion and processing methods remains crucial for achieving the full potential of these recyclable materials.[118]

A notable implementation of DA chemistry in nanocomposites was demonstrated by Oh *et al.*, who developed thermally healable graphene-nanoplate/epoxy (GNP/EP) nanocomposites through an *in situ* DA reaction between GNPs and furfurylamine during the epoxy curing process. This approach utilized GNPs as both reinforcement and crosslinking platforms, eliminating the need for traditional dienophiles like maleimide. Importantly, the materials exhibited thermal self-healing properties and mechanical recyclability. This study also revealed that DA-based CANs showed significantly lower glass transition temperatures compared to conventional cross-linked thermosets due to rDA reactions at elevated temperatures, highlighting both the advantages and temperature limitations inherent to DA-based recyclable nanocomposites.[117]

Cantamessa *et al.* extended DA chemistry applications to elastomer nanocomposites, developing a thermoreversibly crosslinked rubber nanocomposites through solvent-free melt processing. Using furan-functionalized ethylene–propylene rubber cross-linked with bismaleimide, they incorporated reduced graphene oxide (rGO) to create mechanically reinforced and thermally conductive elastomer nanocomposites. The rGO nanocomposites demonstrated remarkable property enhancements, with stiffness increasing dramatically from 1.4 MPa for the neat elastomer to 8.3 MPa with 10 wt% rGO (+465% increase), while tensile strength improved by 51% and thermal conductivity reached 0.47 W (m^{-1} K^{-1}) (+124% increase). SEM analysis revealed good rGO dispersion at 5 wt% loading, though some aggregation occurred at higher loadings due to processing limitations. Importantly, solubility tests indicated that rGO played an active role in crosslinking the network, with direct participation of rGO in DA reactions with bismaleimide. Crucially, these nanocomposites retained full thermoreversibility and recyclability through multiple melt reprocessing cycles, demonstrating that sustainable elastomer nanocomposites with significantly enhanced properties can be achieved while maintaining the circular economy benefits of dynamic covalent networks.[119]

Imines

Chemistry. Imine bonds, also known as Schiff bases, have emerged as another promising approach in the design of dynamic matrices for recyclable thermoset nanocomposites.[120] Imines are formed through condensation between carbonyls (particularly aldehydes and ketones) and amines.[121] Imine bonds are reversible and dynamic, enabling the development of recyclable thermosets.[122] Notably, imines can undergo exchange through a versatile range of mechanisms, including hydrolysis and condensation reactions, exchange with primary amines, and metathesis between imines.[96] A key advantage of imine bond-based CANs is their responsiveness to external stimuli such as solvent changes, pH fluctuations, and temperature variations, which can influence the equilibrium of these dynamic exchange reactions. This responsiveness occurs under relatively mild conditions, making imine bonds particularly attractive for designing recyclable thermoset nanocomposites.[60,123] Indeed, polyimines have recently emerged as useful recyclable thermosets that can be reprocessed using either water or heat, without the need for catalysts. The versatility of imine bond chemistry extends to epoxy vitrimers as well. Imine bond-containing epoxy thermosets exhibit good ductility, reworkability, and mechanical properties.[124] These materials can be deconstructed and recycled through the hydrolysis and reformation of the imine bond and remolded and repaired through imine exchange reactions.[125] Notably, these processes do not require metal catalysts, pressure heating, or additional monomers, significantly simplifying the thermoset systems reprocessing.[96]

Use as nanocomposite matrices. Most studies on dynamic imine-based matrices have focused on conventional composites. For example, polyimine-based thermosets have been applied as recyclable matrices for composites, including as binders for carbon fiber-reinforced composites (CFRCs). These composites can be directly recycled and reused by soaking in an amine solution, while simple heat-press processes can repair defects in the CFRCs.[122,126,127] Yet, with appropriate modifications to account for the unique properties of nanomaterials, similar imine-based approaches could potentially be applied to develop recyclable thermoset nanocomposites.

For example, Wang *et al.* explored imine-based epoxy vitrimers as matrices for recyclable 'smart' nanocomposites designed for sensor applications, using carbon black (CB, particle size ~95 nm) as the conductive nanofiller.[128] Their work demonstrated that incorporating imines into the matrix enabled nanocomposites to be both mechanically and

chemically recycled while maintaining critical sensing properties. The vitrimer nanocomposites exhibited superior positive temperature coefficient (PTC) effects, exhibiting a sharp increase in electrical resistivity with increasing temperature due to thermal expansion coefficient mismatch between the polymer matrix and conductive filler particles. They also exhibited high thermal expansion rates above the T_g, making them particularly suitable for thermo-sensitive applications. The materials showed excellent repeatability in PTC behavior with negligible negative temperature coefficient effects, attributed to the cross-linked vitrimer network's ability to hold conductive particles in place and prevent aggregation at elevated temperatures. Importantly, these smart nanocomposites could be recycled through both reprocessing *via* hot pressing and closed-loop chemical recycling *via* imine base hydrolysis and reformation.[128]

In another study, imine-based CANs were applied as matrices for conductive nanocomposites. Chen *et al.* developed bio-based poly(imine-amide) networks using vanillin-derived dialdehydes, citrate ester, and diamine monomers *via* catalyst-free condensation reactions. The resulting poly(imine-amide) networks exhibited excellent mechanical properties (tensile strength up to 70 MPa) while maintaining rapid stress relaxation capabilities, confirming their dynamic covalent nature. When incorporated with multiwalled carbon nanotubes (MWCNTs), these nanocomposites demonstrated high electrical conductivity with a low percolation threshold of 0.43%. Critically, the nanocomposites retained full reprocessability through thermal remolding.[129]

These studies highlight the potential of imine bond chemistry to contribute to achieving a circular economy for nanocomposites by developing thermoset nanocomposites with both enhanced properties and recyclability. However, it is important to note that the moisture sensitivity of imines raises concerns about the long-term stability of these materials.[130] This sensitivity, while advantageous for recyclability, may limit the applicability of these materials in certain environments.

Nanocomposite matrices with multiple dynamic covalent chemistries. Other CANs and vitrimers with multiple dynamic covalent chemistries have been developed, leading to materials with hierarchical responses to different stimuli.[131–133] These multi-dynamic systems offer enhanced control over material properties and recycling processes, though their increased complexity presents challenges in terms of processing and property optimization.[134]

The complexity of incorporating multiple dynamic chemistries into nanocomposites while maintaining both mechanical performance and processability has been explored by Dodo *et al.*, who developed a triple-dynamic system using hydrogen bonds, thiol-Michael exchange, and Diels–Alder chemistry with carbon nanotube reinforcement.[135] Their work specifically addressed a fundamental challenge in dynamic polymer nanocomposites: the typical trade-off between enhanced mechanical performance and material reprocessability that occurs when nanofillers are added. They noted that direct reinforcement using nanocomposites enhances mechanical properties in resulting composites, but also hinders dynamic characteristics

such as self-healing and reprocessability. While their orthogonal multi-dynamic approach showed promise in maintaining reprocessability despite mechanical reinforcement, achieving over 90% stress relaxation and rapid healing within one hour, the system still faced limitations. The complexity of managing three different dynamic chemistries simultaneously, the need for careful control of reaction conditions to ensure proper bond hierarchy (with thiol-Michael adducts forming preferentially over Diels–Alder), and the requirement for multiple processing steps highlight the ongoing challenges in developing truly practical multi-dynamic nanocomposite systems. Although their approach represented progress toward solving the trade-off between mechanical reinforcement and reprocessability, the increased synthetic complexity and processing requirements demonstrate that simpler, more robust solutions are still needed for widespread implementation.[135]

In another example of multi dynamic nanocomposite matrices, Zheng *et al.* developed dual dynamic polyhydroxyurethane (PHU) nanocomposites that address many of the complexity issues associated with other multi-dynamic networks.[136,137] These systems achieve reprocessability through two synergistic mechanisms: transcarbamoylation exchange reactions between hydroxyl and carbamate groups, and reversible cyclic carbonate aminolysis specific to five-membered cyclic carbonate systems. The dual chemistry approach has proven compatible with various nanofillers, including polyhedral oligomeric silsesquioxanes (POSS) microdomains and Fe_3O_4 nanoparticles, while maintaining recovery rates of 90.6% and 80.5% for neat PHU systems after multiple reprocessing cycles. Notably, the incorporation of Fe_3O_4 nanoparticles enables remote, contactless reprocessing through photothermal heating *via* near-infrared laser irradiation, demonstrating how strategic nanofiller selection can add processing advantages beyond mechanical reinforcement. While higher nanofiller loadings increase activation energies for dynamic exchange, the dual chemistry approach remains viable, requiring lower temperatures and shorter processing times than some single-mechanism systems. The work illustrates that simpler multi-dynamic systems with strategically chosen nanofillers may offer a more viable pathway to commercial implementation than highly complex multi-dynamic chemistry approaches, achieving enhanced performance without the synthetic complexity and processing challenges associated with triple-dynamic systems.[136,137]

Another successful dual-dynamic approach was demonstrated by Bai *et al.*, who developed mechanically strong, healable, and reprocessable conductive carbon black/polysiloxane nanocomposites by incorporating both imine bonds and Zn(II)-amino coordination bonds. The coordination bonds functioned as sacrificial elements that ruptured progressively during deformation, providing energy dissipation while the imine bonds-maintained network integrity, allowing this system to achieve simultaneous improvements in strength, stretchability, and toughness without compromising reprocessability. Notably, the carbon black nanofiller promoted the rupture of sacrificial bonds, improving energy-dissipating efficiency and enabling excellent electrical conductivity. The materials

demonstrated superior cyclability with minimal property degradation over multiple heating–cooling cycles, showcasing how carefully designed dual-dynamic systems can overcome typical nanocomposite-vitrimer compatibility issues.[158]

In general, reprocessable-nanocomposite matrices approaches often suffer from poor filler dispersion, weak interfacial interactions, and the tendency for nanoparticles to interfere with dynamic bond exchange reactions. To address these issues, Bai *et al.* developed a dual-dynamic system combining vinylogous urethane crosslinks with sacrificial Zn(II)–amine coordination bonds in silica-filled PDMS composites, incorporating both chemistries at the filler-matrix interface through surface modification. While their approach showed improvements in mechanical properties and maintained reprocessability, the system still faced limitations typical of nanocomposite vitrimers. The complexity of managing multiple dynamic chemistries simultaneously, potential interference between different bond exchange mechanisms, and the challenge of achieving uniform dispersion at higher filler loadings remained as obstacles. Although their dual-chemistry approach represented progress toward addressing fundamental nanocomposite-vitrimer compatibility and dispersion issues, complete resolution of the trade-offs between mechanical enhancement, processability, and long-term stability in multi-dynamic systems requires further development.[159]

The future of reprocessable matrices for nanocomposites. The discussion above revealed several challenges and design considerations that are common across the different chemical approaches for developing reprocessable thermoset nanocomposites. For example, good dispersion of the nanofiller phase must be maintained, and aggregation must be avoided. Therefore, the dynamic covalent chemistry introduced to the matrix must not compromise the combability between the matrix and nanofiller phases. Furthermore, the heat and mechanical forces involved in reprocessing can lead to agglomeration of the nanomaterials in the filler phase, compromising the material's properties. To avoid agglomeration, harsh stimuli for inducing bond exchange should be avoided (*e.g.* very high temperatures). Another key consideration is the relationship between the temperature required to induce bond exchange and the other key thermal properties of the matrix material. For example, the matrix will be rigid and glassy at temperatures below its T_g, hindering bond exchange. Furthermore, the temperature to induce bond exchange and processability must be below the thermal decomposition temperatures of the matrix and the nanofiller phases. Notably, thermal decomposition during reprocessing can lead to the shedding of nanofiller and nanocomposite micro- or nanoplastics after recycling, posing an environmental hazard.[160] Finally, it is critical to consider the stability of the material at the temperatures associated with the intended application of the nanocomposite. As such, many vitrimers and CANs are not suitable for application in high-temperature settings where significant bond exchange, stress relaxation, and softening will occur. These constraints can serve as design parameters to guide innovation in the field moving forward.

The range of dynamic covalent chemistries continues to expand, offering new routes to address these challenges and optimize the performance of dynamic reprocessable matrices for thermoset nanocomposites. The chemistries highlighted above represent just a limited range of the dynamic bonds that can be used to prepare dynamic thermoset matrices for nanocomposites. Other emerging dynamic covalent chemistries that could be utilized and are being utilized include dynamic siloxane reactions,[161,162] phenoxies[163,164] quaternary anilinium salts,[165,166] aromatic diselenide-based systems,[167] olefin metathesis chemistry,[168] transcarbamoylation chemistry,[169] and boronic esters.[170]

The development of these alternative chemistries continues to expand the toolbox available for designing recyclable thermoset systems. Each approach offers distinct advantages and limitations, providing options for tailoring material properties and recycling conditions to specific application requirements. The ongoing exploration of new dynamic chemistries will continue to advance the recycling of thermoset nanocomposites.

Recovery of nanomaterials

As an alternative to reprocessing, circularity for thermoset nanocomposites can also be achieved by separating nanofillers from the matrix phase, thereby extracting valuable nanomaterials from nanocomposite waste. In this 'nanomaterial recovery approach' illustrated in Fig. 6, the nanofiller phase is separated from the matrix and collected in a purified state, allowing the valuable nanomaterials to be reused. While promising, research specifically focused on nanomaterial recovery from thermoset matrices remains limited. In contrast, there is extensive data and established techniques for recovering and recycling the filler phase in conventional fiber-reinforced composites, particularly those involving carbon fibers (CF) and glass fibers (GF).[171–173] While the scale and specific challenges differ between nanomaterials and conventional fiber fillers, many of the fundamental principles and approaches developed for CF and GF recovery can inform our understanding of nanomaterial recovery processes.

The methodologies for recovering materials from thermoset composites can be broadly categorized into physical, thermal, or chemical, in addition to other methods that use of deconstructable-by-design thermoset matrices. Each method presents unique advantages and challenges when applied to nanomaterial recovery.[174] While direct data on nanomaterial recovery is scarce, examining the established processes for CF and GF recovery provides valuable insights into the potential pathways and challenges for nanomaterial recovery. These insights are particularly relevant as many of the fundamental challenges – such as matrix degradation, filler damage prevention, and process optimization – remain consistent across different scales of reinforcement materials.

In the following sections, we examine these recovery methods mostly through the lens of recycling conventional composites, particularly carbon fiber-based composites, which are referred to as carbon fiber-reinforced polymer (CFRP). At the
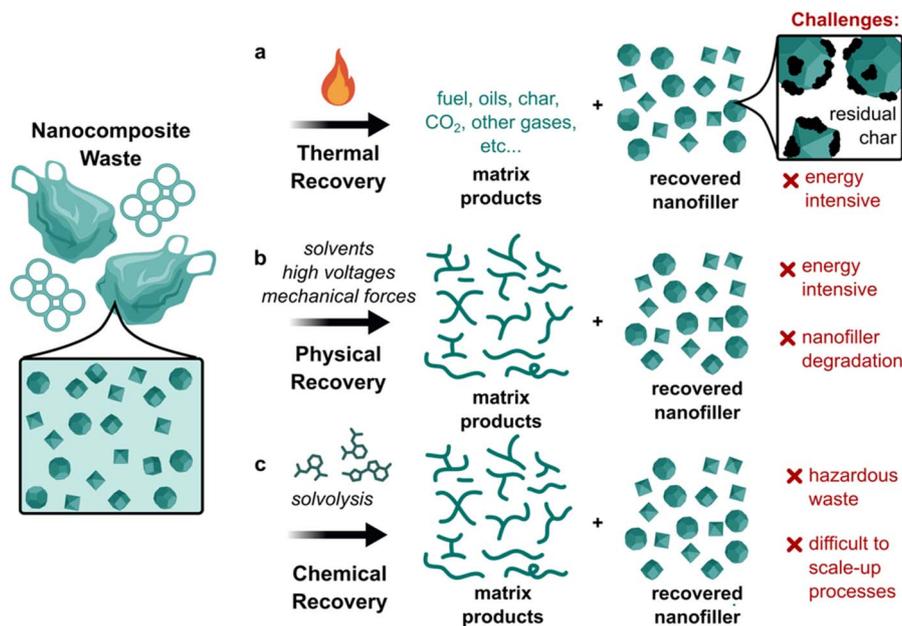
Fig. 6 Recovering nanofillers from thermoset nanocomposites. The nanofiller phase can be recovered from thermoset nanocomposites through (a) thermal, (b) physical, or (c) chemical recovery. (a) Thermal recovery produces recovered filler and the products of matrix pyrolysis, which range from fuels to oils, char, CO_2, and other gases. (b) Physical recovery (b) and chemical recovery (c) produce the recovered nanofiller and the oligomeric, polymer, or monomeric fragments of matrix degradation. The challenges of each approach are highlighted on the right.

end, we highlight the implications and potential adaptations necessary for nanomaterial recovery.

Physical recovery. Physical recovery encompasses techniques to extract valuable nanomaterials from thermoset nanocomposite without using chemical reactions to deconstruct or degrade the matrix.[96] These methods can be broadly categorized into mechanical processes, solvent-based approaches, and high-voltage techniques. In general, physical recovery approaches must consider that the presence of nanomaterials can affect processing parameters. The high surface area and reactivity of nanomaterials may influence optimal processing conditions. Furthermore, contained processing methods may be necessary to minimize nanoparticle release during recovery operations.[175]

Mechanical recovery. Mechanical recovery represents the most straightforward and widely implemented approach. This process begins with the concentration of filler waste, which undergoes crushing and grinding to produce millimeter-scale chips. These chips are further refined into finer powder, yielding recyclates of varying particle sizes that can be segregated into fibrous and powdered forms through sieving.[176] While this method offers advantages such as ease of implementation, high recovery rates, and minimal environmental impact due to the absence of additional pollutants, it comes with significant drawbacks. The aggressive mechanical processing substantially degrades the surface morphology of recovered fillers and can cause irreversible damage to high-value materials, compromising their performance.[58] Additionally, the process is challenged by the high variability in waste material composition, necessitating energy-intensive grinding.[171]

When focusing on nanocomposite materials, mechanical processing approaches should consider granulation or powder generation as intermediate steps, though these typically focus on reducing particle size for subsequent processing rather than achieving nanofiller-matrix separation. Additionally, mechanical recovery operations must account for potential nanoparticle release during processing, necessitating contained processing environments to minimize occupational and environmental exposure.[175]

The challenges of mechanical recovery are further complicated by the nature of thermoset matrices. The permanently crosslinked nature of thermosets makes it exceptionally difficult to remove resins adhering to filler surfaces, hampering effective separation.[177] The presence of nanomaterials can further complicate processing parameters, as the high surface area and reactivity of nanoscale fillers may influence optimal processing conditions. In particular, for carbon fibers recovered through mechanical methods, recovered fillers typically exhibit inconsistent sizes and reduced strength. These limitations, combined with poor bonding between the recovered filler fraction and new matrices, significantly restrict the reusability and economic value of mechanically recovered filler materials.[178] Consequently, mechanically recovered fillers are often limited to low-grade applications with minimal compositional requirements, such as concrete mixes or asphalt for construction.[179] These limitations have kept the field of mechanical recycling of thermoset nanocomposites relatively underdeveloped, necessitating the development of alternative physical recovery methods.

Solvent-based approaches. Solvent-based approaches, particularly swelling techniques, have shown promise in preserving

fiber length and strength. For example, hot acidic pretreatment can induce swelling in carbon fiber-reinforced composites, enabling the recovery of carbon fibers with superior length, tensile strength, and enhanced torsional properties through the subsequent hot pressing of the swelled waste composite.[180] However, the applicaton of solvent-based approaches to thermoset nanocomposites remains under investigated.

High-voltage techniques. High-voltage techniques offer a different approach to separating and isolating the filler phase from composites. Electrodynamic fragmentation (EDF), for instance, employs high voltage pulses between 50 and 200 kV in ionized water to break down composite waste into smaller pieces.[171,181] A related technique, high voltage fragmentation (HVF), utilizes 160 kV pulses to disintegrate materials through repetitive electrical discharge between electrodes. During HVF, the generated spark channel produces intense shockwaves at extreme temperatures ($>10^4$ K) and pressures (approximately 10^9–10^{10} Pa), creating internal stresses that exceed the material's tensile strength and lead to controlled disintegration. While these high-voltage methods can produce relatively clean and long recovered fibers, they require significantly higher energy input, approximately 2.6 times more than mechanical recovery.[182]

Looking ahead, there are many opportunities to improve the physical recovery of fillers and advance this field. For mechanical recovery, processing equipment needs to be improved to better preserve the original fiber structure. Additionally, optimizing solvent-based and high-voltage techniques could help balance the trade-offs between the quality of the recovered materials and energy consumption, potentially leading to more efficient and effective recovery methods for valuable nanomaterials from thermoset composites.

Thermal recovery. Thermal recovery represents one of the primary approaches for reclaiming fillers from conventional thermoset composites and can also be implemented for recovering valuable nanomaterials from thermoset nanocomposites. This method leverages heat to break down the composite matrix while preserving the embedded fillers. Operating at temperatures between 400–1000 °C, thermal recovery processes decompose, burn, or evaporate the matrix, allowing for the extraction of pure fillers.[171] While these methods effectively produce clean fillers, and in some cases can convert matrices into potentially useful fuel products, they face several challenges, including high energy requirements, potential degradation of filler strength and other properties, and the generation of potentially harmful gases from matrix decomposition.[183,184]

The effectiveness of thermal recovery heavily depends on the careful control of several key parameters including the operation temperature, oxygen flow rate, and residence time. The optimal processing temperature varies based on the specific resin type and filler used in the composite.[185] Using temperatures that are too low can leave residual char on the filler surface (often referred to as "undercooking"), while excessive temperatures may reduce the size of recovered filler ("overcooking").[183] Additionally, the presence or absence of oxygen significantly impacts the decomposition process and the quality of recovered

fillers. The duration of thermal exposure must also be carefully managed to achieve optimal filler regeneration without causing excessive degradation.[186,187] When processed at optimal processing parameters, some fillers can be recovered with minimal surface damage and retain 50–95% of their original characteristics, particularly tensile strength. Carbon fiber is an example of such a filler.[183,188] Furthermore, the presence of nanomaterials can significantly influence the thermal recovery process itself. Mineral nanoparticles such as clay, SiO_2, and TiO_2 can considerably change the degradation patterns of polymers during pyrolysis, potentially altering the composition of gases produced, affecting monomer yields, or increasing char generation. This suggests that thermal recovery parameters may need to be optimized differently for different nanofiller types to achieve effective separation.[175] Critically, thermal recovery proves less effective for many fillers, as high-heat conditions can compromise filler properties, significantly limiting their potential for reuse, and post-chemical treatments are often necessary to restore their properties.[184]

Three main approaches have emerged for the thermal recovery of fillers from composites: pyrolysis, microwave heating, and the fluidized bed process. Pyrolysis refers to the thermochemical decomposition of the composite's matrix in an oxygen-free atmosphere at temperatures ranging from 400–1000 °C to draw the fillers.[58] While pyrolysis can produce clean fillers, it has high energy requirements, which must be balanced against the value of recovered nanomaterials. Beyond pyrolysis, alternative thermal approaches such as gasification and hydrocracking have been suggested for nanocomposite processing, though their specific application to nanomaterial recovery from thermoset matrices requires further investigation.[175]

Three of the commonly used atmospheric conditions during pyrolysis are vacuum, nitrogen, and superheated steam. During this process, the decomposed matrix produces low molecular weight products in the form of gases, oils, and char that can to the recovered filler.[171] Notably, in addition to the recovered and reusable filler phase, these products of matrix decomposition can be re-used as fuel or as new resources for chemical processes. However, the removal of toxic compounds may be necessary before pyrolysis to ensure the produced oils are safe for use as fuels or petrochemical industry feedstock.[188]

The most significant challenge of pyrolysis is the char that can adhere to the fillers' surface, as it can substantially reduce the mechanical properties of recovered fillers (Fig. 6). The quality of the recovered filler must meet requirements for effective use.[173] Pyrolysis parameters can be optimized to improve recovered filler quality by reducing the growth rate of pre-existing surface damage. Furthermore, other solutions include post-pyrolysis oxidative treatment to remove carbon residues,[189] use of CO_2 and water vapour to reduce char formation,[190] two-temperature step processes to optimize filler quality,[191] and chemical treatments and post-heating to minimize char formation and improve the recovered filler's quality.[192]

Microwave heating represents a significant advancement over conventional pyrolysis, offering faster processing speeds

(only 12% of conventional heating time), lower energy requirements, and higher recovery rates. This method also provides uniform internal heating and better process control.[192,193] However, microwave heating is currently limited to the laboratory scale, requires precise control of feedstock dielectric properties, and sophisticated equipment and operational expertise.[194] Currently, microwave heating often serves as a complement to conventional pyrolysis, accelerating the decomposition process.[58] Future developments in microwave control and energy efficiency could expand its applications.

The fluidized bed technique represents a distinct approach to thermal recovery, using air as both a heating medium and transport mechanism. This process employs a bed of fine silica sand (0.85 mm particle size) with airflow at 0.4–1.0 m s^{-1} to create fluidization. The airflow helps oxidize the polymer matrix, while a secondary chamber at 1000 °C handles volatile compound oxidation, ensuring clean recovered fillers.[195] This method shows particular strength in handling mixed and contaminated materials, accommodating various polymer types, painted surfaces, and foam cores without requiring separation or purification before processing.[195] It proves especially suitable for end-of-life waste composites. However, it is worth noting that when applied to carbon fibre composites, the process struggles with degradation in the length and mechanical properties of recovered filler fibers, produces fluffy fiber structures, consumes high energy, and results in discontinuous recovered materials.[173,178]

While thermal recovery methods show promise, several challenges remain for industrial-scale implementation. Issues persist regarding precise temperature control to prevent undesirable degradation, energy efficiency improvements, better solutions for char prevention, methods to maintain filler quality during processing, and strategies for handling mixed material streams.

Chemical recovery – solvolysis. Chemical approaches for filler recovery use chemical reactions to deconstruct the crosslinked matrix of thermoset nanocomposites, facilitating the separation of fillers from matrix fragments. Solvolysis is the most common approach to deconstructing the thermoset composite matrices. The primary advantage of solvolysis over thermal and physical recovery is its ability to recover high-quality fillers while preserving their original structure and avoiding the high energy consumption associated with thermal methods. Under optimal conditions, chemical processes can reclaim fillers with minimal degradation – for instance, achieving carbon fiber recovery with only 1.1% tensile strength loss, while simultaneously recovering the polymer matrix.[58,196] However, chemical recovery methods face several challenges, particularly the generation of hazardous waste and the harsh conditions required. Additionally, many chemical processes require sophisticated equipment and tightly controlled conditions, leading to difficulties in commercial-scale implementation.[197]

Solvolysis involves using solvents to degrade the polymeric matrix. During solvolysis, hydrolyzable linkages in the matrix backbone (*e.g.* amides, esters, ethers, urethanes, or carbonates) are chemically cleaved through chemical reactions with the solvent. Solvolysis typically begins with mechanical grinding of the composite to increase surface area, followed by chemical treatment and washing of the recovered fillers to remove surface residue.[172] Supercritical fluids have emerged as particularly effective solvents for solvolysis due to their unique physicochemical properties, combining liquid-like density and solvating power with gas-like viscosity and high diffusivity. A fluid reaches its supercritical state when both its temperature and pressure exceed critical points, at which point it can achieve resin removal efficiencies of 95–100% on filler surfaces.[198]

Under supercritical conditions (500 °C and 23 MPa), matrix degradation rates can reach >95% after 90 minutes.[171] The addition of catalysts, such as potassium hydroxide, sodium hydroxide, or phenol, can significantly improve degradation efficiency,[171] though this may result in reduction in filler tensile strength.[171] The process can be further enhanced by adding carbon dioxide or organic solvents to improve water's fluidity and mass transfer properties. However, many thermoset matrices are hydrophobic in nature, which can hinder efficient hydrolysis.[171]

Alcohols (methanol, ethanol, propanol) in their supercritical state offer an alternative to hydrolysis, as they generally require lower critical temperatures and pressures than water. In particular, propanol has shown superior performance due to its high solvation capacity for organic matrices. The addition of catalysts can further improve degradation efficiency, though this may impact filler properties.[199]

Oxidative degradation, particularly with nitric acid and/or hydrogen peroxide (H_2O_2) has also been demonstrated to effectively break down thermoset matrices in composites. The process typically operates at lower temperatures (60–90 °C) compared to supercritical treatments.[200] The addition of H_2O_2 to various solvents (such as acetic acid, acetone, or DMF) has shown promising results, achieving over 90% resin decomposition while maintaining filler properties.[96]

Notably, the filler recovery quality and yield can be significantly improved by combining the chemical processes discussed above with sonication or irradiation. When combined with chemical treatments, sonication can achieve matrix degradation rates exceeding 95% in reduced timeframes.[201] Microwave-enhanced depolymerization can lead to high filler yield in a short process time.[178]

In conclusion, chemical recovery methods offer several advantages over mechanical and thermal approaches, particularly in preserving filler properties and enabling matrix recovery. However, the widespread implementation of these techniques faces challenges related to cost, equipment requirements, and environmental considerations. The emergence of green solvents, such as ionic liquids and deep eutectic solvents, offers promising directions for addressing these limitations, potentially enabling more sustainable and economically viable recycling processes. But all of these requires studies focused specifically on nanocomposites.

Deconstructable by design thermoset matrices. Notably, thermoset matrices for composites are not designed to be readily degraded through chemical reactions. Given that thermoset composites are used in demanding high-
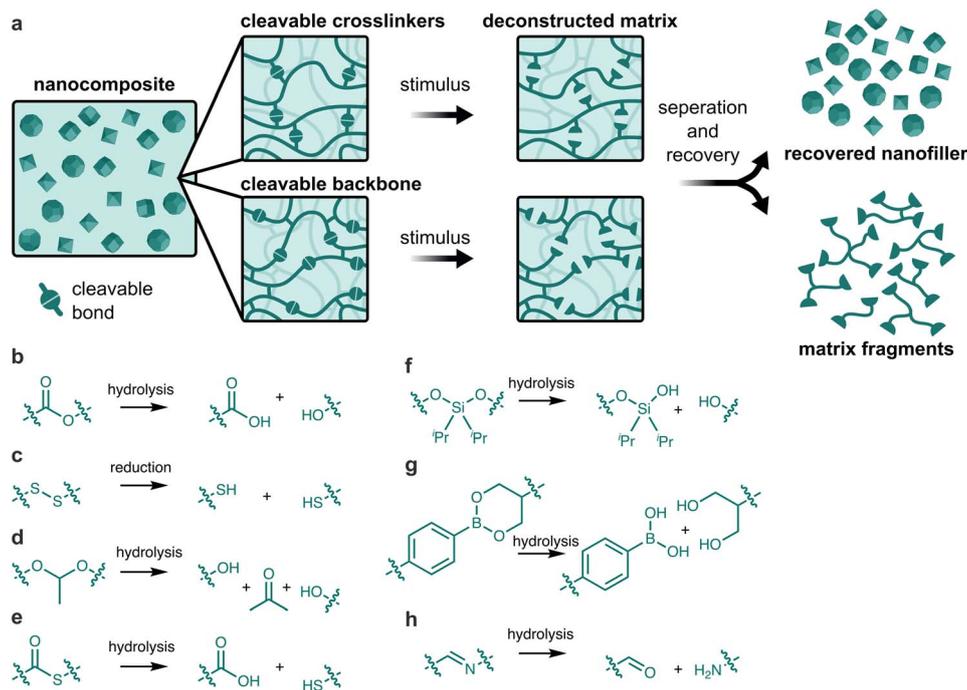
Fig. 7 Deconstructable by design matrices for thermoset nanocomposites. (a) Cleavable bonds can be added to the matrix of thermoset nanocomposites into either the crosslinker or polymer backbone. These matrices can be deconstructed, allowing the nanofillers to be separated and recovered. (b–h) Representative chemistries used as cleavable bonds include (b), esters, (c) disulfides, (d) acetals, (e) thioesters, (f) silyl ethers, (g) boronic esters, and (h) imines.

performance applications (*e.g.* transportation, infrastructure, energy, sports equipment, construction, and chemical industries) they are instead typically designed to be highly resistant to corrosion and chemical degradation. This resistance to chemical degradation is why the harsh temperatures, pressures, and chemical conditions described above are typically required for the solvolysis of thermoset matrices. Furthermore, many thermoset matrices (*e.g.* polydicyclopentadiene and styrenic resins) have only inert carbon–carbon covalent bonds in their backbone, making them entirely resistant to chemical deconstruction by solvolysis.

An emerging approach to overcoming these limitations is to redesign thermosets to be susceptible to selective deconstruction in response to milder conditions.[178] This can be achieved by installing 'weak-link' bonds throughout the thermoset backbone that can be selectively broken under specific external conditions. These cleavable bonds, which can be added to the thermoset matrix in either the crosslinkers or as cleavable comonomers (Fig. 7) can be broken in response to specific stimuli including heat, irradiation, acidic conditions, or mechanical stress, allow the thermoset matrix to be broken down and separated from the fillers. Ideally, these bonds are designed to render the thermoset deconstructable while maintaining the material's mechanical properties during use.[59,202] Common types of cleavable bonds that have been successfully incorporated into thermoset composites include esters,[203–205] disulfides,[206] thioesters[207] acetals,[208,209] boronic esters,[210] amides,[211] imines,[212,213] and silyl ethers.[214] In some cases, the bond cleavage is reversible, allowing the post-degradation

matrix fragments to be reconnected and reused. With these chemically recyclable 'deconstructable-by-design' matrices, recovery and circularity can be achieved for both the filler and matrix phases.

This approach has been extensively studied for recovery of fiber from conventional composites like CFRP. For instance, CFRP composites have been prepared with matrices that contain reversible amidation chemistry between linear polyethyleneimine and bifunctional maleic anhydride cross-linkers. These composites exhibited exceptional mechanical properties (tensile strength >450 MPa, Young's modulus >12 GPa), thermal stability, and solvent resistance. Under acidic conditions at ambient temperature, the matrices could be fully depolymerized, enabling complete separation and recovery of both carbon fibers and monomers with high separation yields (>93%). The recovered carbon fibers retained nearly identical mechanical properties to pristine ones and could be reused for multiple cycles, with new composites recovering over 95% of their original properties.[211] Two other examples of deconstructable-by-design thermoset matrices for CFRP include poly(hexahydrotriazine) matrices,[215] silyl ether-containing polydicyclopentadiene matrices,[214] and acetal-containing epoxy resins,[205] all of which exhibited performance comparable to commercial counterparts while allowing for recovery of the carbon fiber under mild acidic conditions.

An alternative approach is to construct the entire thermoset network from cleavable bonds, rather than incorporating them as weak links within a conventional thermoset backbone. Polyimine networks exemplify this strategy, where the crosslinked

structure is formed entirely through dynamic imine bonds that can undergo selective cleavage under acidic conditions through hydrolysis reactions. For instance, Zou *et al.* demonstrated a polyimine-based electronic skin containing silver nanoparticles that could be completely deconstructed by soaking in an acidic recycling solution, causing the imine bonds to hydrolyze and the crosslinked network to depolymerize into soluble oligomers and monomers while allowing clean recovery of the metallic nanofillers. The authors successfully recycled their conductive ink up to four printing cycles and demonstrated the quality of the remade material through multiple characterization methods. Furthermore, the recycled films actually exhibited increased Young's modulus and tensile strength compared to the original films, likely due to more complete crosslinking reactions during repolymerization. The authors also demonstrated functional recovery by fabricating sensors from the recycled materials and testing their sensing capabilities, confirming that the recycled e-skin components retained comparable performance to the original devices. This comprehensive approach validates both the chemical recyclability of the imine network and the practical reusability of the recovered materials for functional device applications.[216]

A particularly advanced example of deconstructable polyimine networks involves vanillin-based liquid crystalline thermosets with graphene nanoplatelet fillers designed for thermal management applications. Under mild acidic conditions (HCl/acetone mixture), the polyimine networks underwent complete depolymerization through hydrolysis of the imine bonds, enabling selective recovery of both the vanillin-derived monomer and the graphene nanoplatelets with high efficiency (97.5% graphene nanoplatelet recovery). The recovered components retained their structural integrity and functionality, and the regenerated nanocomposites prepared from the recovered materials demonstrated comparable thermal properties to the original materials, validating the closed-loop recyclability of these high-performance thermal management materials.[217]

Similarly, Peng *et al.* demonstrated recycling of supramolecular polyimide nanocomposites with functionalized boron nitride nanosheet fillers. Deconstructability was based on imines and hydrogen bonding interactions, enabling fully closed-loop recycling in both liquid and powder states. The recovered materials maintained their original thermal and electorical properties, demonstrating the viability of designing multifunctional electronic materials with built-in recyclability through strategic incorporation of dynamic bonds.[218]

Beyond these polyimine-based approaches, deconstructable matrices that enable nanofiller recovery have also been designed from polyhemiaminal and polytriazine (PHT) thermoset matrices.[219,220]

This approach to designing recyclable thermoset nanocomposites represents a significant shift from traditional recycling methods, addressing recycling challenges at the material design stage rather than at the materials end-of-life. By enabling the non-destructive recovery of valuable fillers and the matrix, these innovations offer promising pathways for developing sustainable, high-performance composite materials.

The future of filler phase recovery: transitioning to nanomaterials. The recovery of nanomaterials from thermoset nanocomposites represents a critical challenge in achieving a circular economy for advanced materials. While significant progress has been made in developing various recovery methods – physical, thermal, chemical, and deconstructable thermoset approaches – most research to date has focused on conventional composites, particularly on carbon fiber recovery from CFRP. Limited data is available specifically for nanomaterial recovery. This knowledge gap presents both challenges and opportunities.

While the fundamental principles developed for carbon fiber recovery may provide valuable insights for nanomaterial recovery, the unique characteristics of nanomaterials – particularly their size, surface area, and tendency to agglomerate – mean that direct application of these methods may not be suitable. Physical recovery methods that work well for carbon fibers may be too aggressive for nanoscale materials, while thermal and chemical processes may affect nanomaterials differently due to their high surface-to-volume ratios. Furthermore, minor disruptions to nanomaterials surface chemistry can drastically change their properties, stability and interactions with the matrix phase. This gives rise to challenges with agglomeration and poor dispersion of the nanofiller phase, which can compromise the performance of thermoset nanocomposites. Notably, nanomaterials are an incredibly broad class of materials that have varying length scales, shapes, chemistries, and properties. By extension, they also have widely varying sensitivities to recovery processes. Therefore, recovery techniques will likely need to be tailored to meet the specific challenges associated with specific nanocomposite materials. Overall, significant work remains to understand how these systems behave when incorporating nanoscale fillers rather than carbon fibers.

Looking ahead, research priorities must expand beyond carbon fiber recovery to specifically address nanomaterial recovery challenges. This includes developing cost-effective recovery methods tailored to the characteristics of specific nanomaterials, understanding the behaviour of nanomaterials during recovery processes, and establishing appropriate testing and characterization protocols to evaluate the quality of recovered nanomaterials. The field also needs to investigate potential aggregation and re-dispersion issues specific to recovered nanomaterials, aspects that are less critical for carbon fiber recovery.

Inherently recyclable nanomaterials. An alternative approach that could circumvent many of these challenges involves designing nanomaterials with inherent recyclability from the outset. For example, Zeng *et al.* demonstrated this concept with biophenolic nanospheres based on reversible supramolecular assembly. These nanospheres could be completely disassembled in DMF at room temperature and reassembled upon cooling, achieving a remarkable 96.7% recovery rate from flexible polyurethane foam through simple soaking for 5 minutes. The recovered nanospheres maintained identical structure and functionality to virgin materials, enabling direct reuse without

additional treatment. This design-for-recycling approach suggests that incorporating stimuli-responsive assembly behaviour into nanomaterial architecture could achieve nearly perfect closed-loop recycling.[221]

Through focused research on nanomaterial recovery, the field can progress toward truly sustainable life cycle management of thermoset nanocomposites, ultimately contributing to a more circular materials economy while maintaining the unique properties that make these materials valuable in modern applications.

Reuse of the recovered nanofiller. The successful reuse of recovered nanomaterials is critical for achieving true circularity in thermoset nanocomposites and represents the ultimate validation of recovery efforts. For circularity to be achieved, recovered nanomaterials should effectively replace virgin materials in new applications. While the recovery techniques discussed above show promise for extracting nanomaterials from thermoset matrices, the ultimate value of these approaches depends on the quality and performance of the recovered nanomaterials in subsequent applications.

The reuse of recovered nanomaterials faces several key challenges. First, the recovery processes themselves may introduce defects or alter the surface chemistry of nanomaterials, potentially compromising their performance in new applications. Second, ensuring adequate dispersion of recovered nanomaterials in new matrices can be challenging, particularly if the nanomaterials have undergone agglomeration during the recovery process. Third, the interfacial properties between recovered nanomaterials and new polymer matrices may differ from those of virgin materials, affecting the overall composite performance.

Despite these challenges, recent work has demonstrated the feasibility of successfully reusing recovered nanomaterials from both thermoset composite recycling and alternative waste streams. A notable example was reported by Mikhalchan et al., who demonstrated the successful reuse of CNT sheets recovered from thermoset composites via thermal pyrolysis.[222] Using a two-step thermal process involving treatment in nitrogen up to 500 °C followed by oxidation at 500 °C, the recovered CNT sheets retained remarkable properties: commercial sheets maintained 95% of their original tensile strength and 100% of their electrical conductivity, while laboratory-grade sheets retained 54–59% of their strength. Critically, the recovered CNT sheets could be directly reused in new thermoset composite manufacturing without additional treatment, demonstrating the potential for truly circular material flows. The preservation of both mechanical and electrical properties enabled reuse in the same high-performance applications, representing the highest level of material circularity.

Complementing this thermal recovery approach, Poutrel et al. demonstrated successful nanomaterial recovery from vitrimer matrices using mild chemical dissolution. Their study focused on graphene-based nanoparticles incorporated into epoxy vitrimer matrices at loadings up to 1 wt%. Using ethylene glycol dissolution at 180 °C, they achieved recovery yields of 92–93% for graphene nanoparticles and reduced graphene oxide, and 48–73% for more highly functionalized particles (graphene

oxide and silane bearing epoxy functionalised graphene nano-platelets (GPTS-GO)). Comprehensive characterization using XPS, XRD, Raman spectroscopy, and SEM revealed that recovered nanoparticles retained their core structural integrity, though some surface chemistry modifications occurred. Notably, low-functionality particles, graphene nanoparticles and reduced graphene oxide, showed minimal structural changes and received additional hydroxyl functionalization, while highly functionalized particles underwent partial defunctionalization during extraction. Despite these chemical changes, all recovered nanoparticles showed no polymer residues and maintained their graphitic structure, indicating successful separation from the vitrimer matrix and demonstrating sufficient quality for reuse applications.[223]

Beyond direct composite recycling, the successful implementation of recovered nanomaterials from alternative waste streams further validates the circular economy potential of these approaches. Lorero et al. demonstrated that ZnO micro-particles recovered from waste alkaline batteries can serve as effective fillers in epoxy thermoset nanocomposites. The recycled ZnO particles, which exhibited unique "desert rose" hierarchical morphologies with nanoscale substructures, showed comparable or superior performance to commercial ZnO and TiO_2 nanoparticles in several key properties. Notably, the recycled ZnO-reinforced composites demonstrated enhanced hydrophobic properties, achieving water contact angles up to 85° after surface etching treatment—significantly higher than neat epoxy (61°) and commercial particle-filled systems. The study revealed that the hierarchical structure of the recycled particles provided enhanced surface roughness while maintaining thermal and mechanical properties comparable to virgin nanofiller systems. This work illustrates how waste-derived nanomaterials can not only match the performance of commercial alternatives but also provide unique morphological advantages that enhance specific composite properties.[224]

These examples collectively demonstrate that recovered nanomaterials, whether from direct composite recycling or alternative waste streams, can successfully replace virgin materials in high-performance applications. The key to successful reuse lies in understanding and leveraging the unique characteristics that recovery processes may impart to nanomaterials—whether through preserved functionality, modified surface chemistry, or enhanced morphological features. This approach not only validates the technical feasibility of nanomaterial circularity but also highlights the economic potential of recovering valuable nanomaterials from diverse waste streams, contributing to a more sustainable materials economy.

Conclusions and outlook

There is clearly a need for further innovation toward a circular economy for thermoset nanocomposites to become a reality. As highlighted above, continued efforts towards this goal are motivated by the environmental impact of nanomaterials production, the environmental hazards associated with disposing of thermoset nanocomposite waste, and the

economic value that can be captured by recycling and reusing valuable nanomaterials. Notably, more comprehensive life-cycle analysis data for different thermoset nanocomposites could further illuminate the value of recycling these materials.

Both approaches for recycling thermoset nanocomposite, reprocessing (Fig. 4 and 5) and extracting and recovering the filler phase (Fig. 6), have progressed rapidly in recent years. Yet, they still face obstacles to achieving real-world, large-scale utilization that require targeted research efforts to overcome.

For reprocessable thermoset nanocomposites using CANs and vitrimers, the primary challenge lies in balancing recyclability with nanocomposite performance requirements. This necessitates developing temperature-responsive dynamic chemistries that can operate at moderate processing temperatures while maintaining high-temperature service capabilities, thereby expanding their applicability to key sectors that utilize nanocomposites such as aerospace and automotive. Additionally, controlling nanofiller dispersion during multiple reprocessing cycles remains critical, requiring systematic investigation of surface functionalization strategies that prevent agglomeration while preserving interfacial bonding. Building on this foundation, multi-dynamic systems that combine complementary bond exchange mechanisms show particular promise for optimizing the trade-off between mechanical performance and processability, though they require careful optimization of bond hierarchy and processing conditions.

As a second approach, nanomaterial recovery strategies have been developed to help address the distinct technical challenges associated with nanoscale materials in thermoset systems. The key research priorities include developing mild chemical recovery methods that preserve nanomaterial surface chemistry and prevent agglomeration during extraction, establishing standardized characterization protocols to evaluate recovered nanomaterial quality, and investigating selective recovery techniques for complex mixed nanofiller systems. These advances would enable the design of nanomaterials with inherent recyclability features, creating a more sustainable approach to nanocomposite design from the outset.

The convergence of these technical advances opens immediate opportunities across multiple industrial sectors, where implementation strategies can be tailored to specific performance requirements and risk tolerances. In the automotive industry, recyclable CNT/epoxy nanocomposites could initially target interior panels and non-critical structural components, where the slightly reduced service temperature of vitrimers is acceptable. Similarly, the aerospace sector could implement recyclable graphene/epoxy nanocomposites in secondary structures and fairing components, particularly as environmental regulations increasingly favor sustainable materials. The electronics industry presents a compelling case for nanomaterial recovery, where valuable metal nanoparticles from printed circuit boards could be extracted using deconstructable matrices, simultaneously addressing material scarcity and e-waste challenges.[216]

These sector-specific applications can be further enhanced by creating synergistic material flows between industries. For example, integrating recovered nanomaterials from alternative waste streams, such as ZnO from batteries[224] or metal nanoparticles from electronic waste,[216] into new nanocomposite applications could establish symbiotic industrial ecosystems where one industry's waste becomes another's raw material. Such cross-industry collaboration would maximize the economic value of recovered nanomaterials while minimizing waste generation across the entire materials lifecycle.

Realizing this vision requires a coordinated, staged implementation approach that acknowledges both technical limitations and market realities. Near-term efforts should focus on low-risk applications where performance compromises are acceptable, allowing for technology maturation and market acceptance. Medium-term developments must address scalability through standardized recycling protocols, industrial-scale processing equipment, and supportive regulatory frameworks. Long-term success ultimately depends on achieving cost parity with virgin materials while demonstrating equivalent performance in the most demanding applications.

Critical enabling factors span technical, economic, and regulatory domains. These include establishing robust collection and sorting infrastructure for end-of-life nanocomposites, developing economic incentives that make circular materials cost-competitive, and fostering industry consortiums that can share the costs and risks of developing new recycling technologies. The integration of these elements will determine whether the vision of truly circular thermoset nanocomposites can be realized at the scale necessary to meaningfully address current sustainability challenges in advanced materials manufacturing.

In sum, while significant technical and economic hurdles remain, the convergence of environmental pressures, advancing recycling technologies, and growing economic opportunities provides compelling motivation for continued innovation toward a circular economy for thermoset nanocomposites. The strategic research directions and staged implementation approach outlined above offer concrete pathways for achieving this critical transformation, ultimately contributing to a more sustainable future for high-performance materials across diverse industrial applications.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this perspective.

Author contributions

EP and ZR developed the concept of the perspective. EP prepared the figures. ZR wrote the first draft of the manuscript, which was further developed, added to, and edited by EP.

Conflicts of interest

There are no conflicts to declare.

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