



Review

Recycling technologies for thermosetting plastics and their composites: Advances through vitrimerization

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Abstract: The proliferation of thermosetting plastics—materials engineered to maintain stable thermal properties across defined temperature ranges—has enabled critical applications in electronics, automotive components, and household appliances. However, their inherent cross-linked or highly stable polymer structures, designed for thermal durability, simultaneously render them difficult to recycle through conventional melt-processing methods. This has led to a growing accumulation of thermosetting plastics and their composite wastes, which often incorporate reinforcing fibers (glass, carbon) or metallic inserts, further complicating waste management. This review examines the current landscape of thermosetting plastic waste streams, focusing on the challenges posed by their high-performance matrices. We critically assess emerging recycling strategies, including thermal, mechanical, and chemical recycling methods, with particular attention to methods that enable the recovery of both the polymer fraction and embedded reinforcements. Furthermore, the potential for emerging vitrimerization technology is explored and extensively reviewed. Key findings indicate that while landfilling and incineration remain prevalent, advanced separation and low-degradation chemical recycling routes presently offer better pathways toward circularity, particularly so because vitrimerization is at its infant stage. The review concludes by presenting a SWOT analysis of the various valorization methods, identifying vitrimerization technology as the future process capable of addressing the gaps in thermosetting plastics and their composite waste management.

Keywords: vitrimers; vitrimerization; thermosetting plastics, thermosetting plastic composites; mechanical recycling; thermal recycling; chemical recycling

1. Introduction

Polymers are a class of modern materials occurring in nature or synthesized from petroleum resources. While natural-occurring polymeric materials degrade through biological activities after end-of-life [1–3], petroleum-derived polymeric materials do not, persisting in the environment and causing significant environmental pollution [2,4]. Petroleum-derived polymers can be categorized into plastics (thermoplastics, thermosets) or elastomers; an exception is when a blend is made from any two or even three different classes of polymers [5,6]. Each petroleum-derived polymer class (thermoplastics, thermosets, or elastomers) possesses unique structural properties; hence, they behave differently in their response to heat, load, and other environmental factors. While items made from thermoplastics can be recycled repeatedly by application of heat, thermosetting plastic articles, once made, cannot be remolded by heat application [7]. After the end of life, thermoplastic materials can be reused for the same or a different product through reprocessing, as their structural bond type can be broken easily through melt processing (reversible heat cycle). Thermosets, on the other hand, have network structures that make the breaking of bonds via melt-processing methods impossible (irreversible heat cycle).

It is therefore important to note that end-of-life thermosetting plastics present bigger recycling challenges [8], while thermoplastics can be recycled by mechanical methods and melt-processing methods (rotational, compression, injection, blow molding), creating new articles. Today, thermosets can only endure mechanical recycling techniques through crushing to derive fillers or reinforcing materials. This irreversible heat cycle makes it impossible to reuse end-of-life thermoset articles for the same purpose; only repurposing becomes possible. Consequently, a high percentage of thermosets are still landfilled or incinerated, causing serious environmental concern. Furthermore, the most versatile method of recycling plastics is the mechanical technique due to its ease, low cost, and rapid processing of high volumes for new products. Unfortunately, thermosets can only be recycled through mechanical (shredding/grinding), chemical (solvolysis), and complete thermal degradation (pyrolysis), which have high energy demand, high process cost, and less throughput [9].

This review therefore seeks to critically assess advances in these recycling strategies, including thermal, mechanical, and chemical recycling methods, with particular attention to methods that enable the recovery of both the polymer fraction and embedded reinforcements. Having laid a firm foundation of the existing techniques of end-of-life thermoset valorization, an emerging non-degrading processing method termed vitrimerization of end-of-life thermosetting plastics and their composites will be explored in greater detail.

Thermosets are a class of plastics that undergo an irreversible chemical curing process to form a rigid material. The curing process triggered by the catalyst, hardener, heat, or irradiation introduces a bond, which makes for a highly cross-linked network structure [10–12].

Generally, the 3D structures of thermosets endow them with superior strength, creep resistance, dimensional stability, and excellent heat and chemical resistance compared to thermoplastics. On the other hand, they are less flexible, have low impact strength, and are very brittle. The latter properties are drawbacks where high impact energy and flexibility are of the essence. However, a balance in desired properties can be achieved by introducing fibers or fillers or through structured blending [13–15].

Furthermore, the inherent lightweight associated with plastics has made thermosets the choice materials for most engineering applications where high thermal, chemical, and mechanical stability is desirable. This includes structural and protective applications, such as aerospace components and wind

turbine blades [16,17]. Additionally, unlike thermoplastics, thermosets bond excellently with fibers such as carbon, glass, or aramid for engineering composite materials. They do not require additional compatibilizers for proper interfacial bonding. Consequently, fiber-reinforced thermosetting plastic composites are gaining new ground in boat hulls, car parts, and other complex applications [18].

Global thermoset production is estimated at 42–48 million tons annually, representing roughly 10%–13% of total global plastics production. This is propelled by demands in the automotive, construction, electronics, and aerospace industries [19]. This calls for a more sustainable approach to thermosetting plastics valorization. Predominant among thermosets are epoxies, polyurethanes, silicones, polyesters, and phenolics.

1.1. Epoxies

Epoxies are high-performance thermoset plastics known for superior adhesion, excellent chemical resistance, and durability. They are formed by the reaction of epoxy resin with a hardener. Epoxies have been widely employed as matrices for structural composite materials, adhesives, coatings, electronics, and aerospace components [20,21].

The global epoxy composite market is a high-growth sector with over 3 million tons of global epoxy resins used in composite material for automotive, aerospace, wind energy, and construction sectors, projected to reach over 4 million tons by 2030 [22]. This suggests that a significant volume of used epoxy materials is ending up in dump sites.

1.2. Polyurethane (PU)

Polyurethane can be either a thermoplastic or a thermoset, depending on its chemical formulation. Most traditional polyurethanes are thermosets that are cured into permanent cross-linked materials. They are characterized by high durability, superior abrasion resistance, and excellent load-bearing capacity. Polyurethane is utilized as foam for insulating materials, in furniture, and in structural applications. The annual PU market size was worth USD 78.1 billion in 2023 and is expected to reach a 3.9% compound annual growth rate up to 2030 [23].

1.3. Silicones

Silicones, also known as polysiloxanes, are versatile, synthetic inorganic polymers made from silicon, oxygen, and organic groups. They exist as oils, gels, or rubbers and are characterized by excellent flexibility and softness, high chemical resistance, high temperature resistance, and very low glass transition temperature (T_g) [24,25]. This has positioned them as choice materials for heat insulation, gaskets, and applications where soft surfaces are desired. Additionally, their excellent flexibility makes them suitable for the preparation of α , ω -reactive terminated silicone oligomers with controlled molecular weights or a wide variety of reactive organofunctional end-groups [26,27]. Global silicone consumption is on a steady upward trend, with total annual consumption reaching an estimated 4.1 million tons in 2024, with the expectation to reach 5 million tons in 2035 [28].

1.4. Polyesters

Polyesters exist as both thermoplastics and thermosets. The former are a saturated form, while the latter are unsaturated polyester resins (UPR), in which the introduction of a third network through free radical polymerization is catalyzed and accelerated under controlled temperature. Cross-linked unsaturated polyester resins are commonly used as matrices for reinforced fiber-glass composites used in marine, construction, and automotive industries [29]. Their ability to withstand high temperatures, corrosion resistance, and high strength make them ideal for these purposes.

The global production volume of thermoset unsaturated polyester resin is estimated at about 1.4 million tons annually. This represents approximately 75% of thermoset composite annual production, with approximately 30% used in the construction industry as glass-reinforced plastic composites (GRP). The global thermoset polyester is projected to grow 12% annually until 2033 [30].

1.5. Phenolic polymers

Phenolic polymers (phenolic resins) are synthetic thermosetting plastics formed by the polycondensation of phenols and aldehydes. They have excellent dimensional stability, low cost, good thermal resistance, and excellent electrical insulation properties, hence being widely used in circuit boards, automotive parts, and construction and aerospace industries [31]. Phenolic resins occur as Novolacs or Resols. The former is formed with a less stoichiometric amount of formaldehyde with an acid catalyst and a curing agent, while the latter is formed with an excess stoichiometric amount of formaldehyde with an alkaline catalyst; curing can occur by heat without any curing agent. Phenolic resin is not environmentally friendly, as it can emit toxic substances at elevated temperatures; its high levels of free phenol and formaldehyde are carcinogenic and pose significant harm to humanity and the environment [32–34]. Currently, researchers are focusing on green processes for producing phenolic polymers by replacing petroleum-based alternatives, reducing energy consumption, and implementing solvent-free synthesis methods. This strategy is aimed at reducing toxic phenol and formaldehyde footprint by utilizing lignins, tannins, and cardanol from plants such as cashew nut shell liquid, as well as applying oxidative polymerization to eliminate formaldehyde usage [32,35].

Global phenolic resin production was estimated at approximately 6.1 million tons in 2024, with a steady growth of 2.2% compound growth rate since 2014 [36]. Currently, the market size is valued to increase by USD 4.56 billion at a compound annual growth rate of 5.4% from 2024 to 2029, largely driven by demands in the automotive, construction, and insulation industries [37].

The increasing demand for the above key thermosetting plastics, coupled with the challenging recycling of thermosets due to their high network structure, will require intensive research for a more viable, sustainable approach to reduce the adverse consequences of indiscriminate disposal of end-of-life thermosetting articles. The projected global annual production by volume for the major thermosetting plastics up to the year 2030 according to global information research [38] is presented in Figure 1 below.

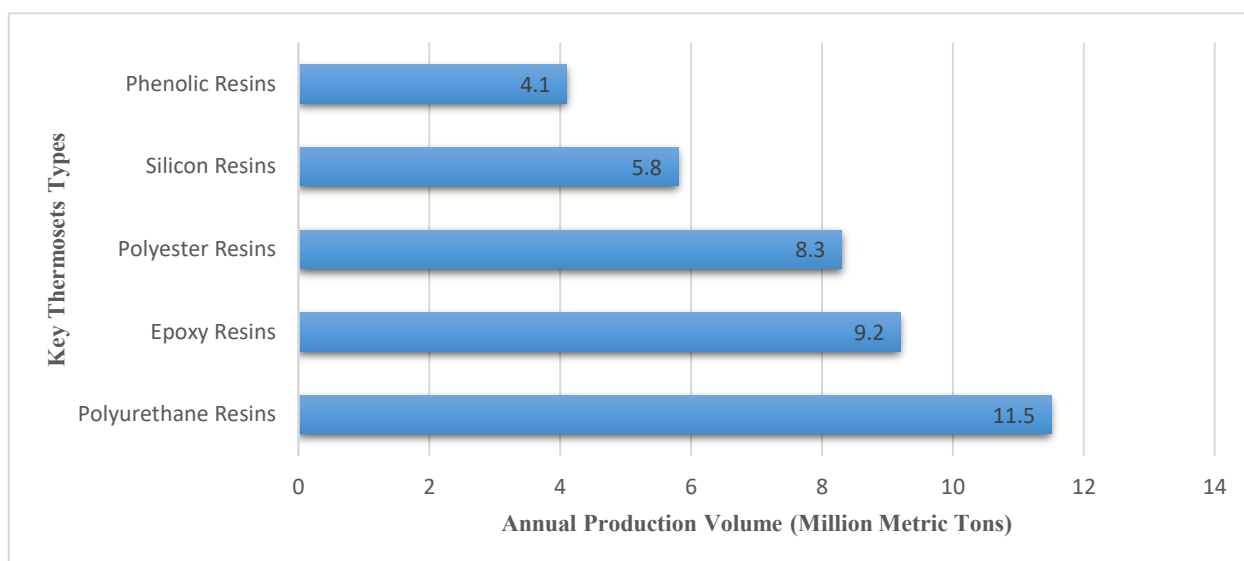


Figure 1. Projected global annual production volume (by 2030) for the major thermosetting polymers, based on current growth analysis [38].

2. Thermosetting plastics and composite recycling methods

Thermosets can be recycled using primary approaches employed for their thermoplastics counterparts; however, due to their durable cross-linked structure, the recycling product is not molten plastics, which could be remolded into the same starting article. Consequently, recycled thermosets are mostly repurposed, either downcycled or upcycled [39–42]. Currently, three primary methods are widely practiced: mechanical recycling, chemical recycling, and thermal recycling approaches [43]. Recent advances in thermoset recycling involve in situ chemical modification of the matrices of the thermoset with a linking agent, thereby introducing an easy-to-break bond without destroying the inherent properties of the thermoset material. This approach has been receiving attention as capable of redefining the use and recycling of end-of-life thermosets.

This section will provide extensive insights and up-to-date details for each of the approaches. Figure 2 presents a graphical summary of these methods.

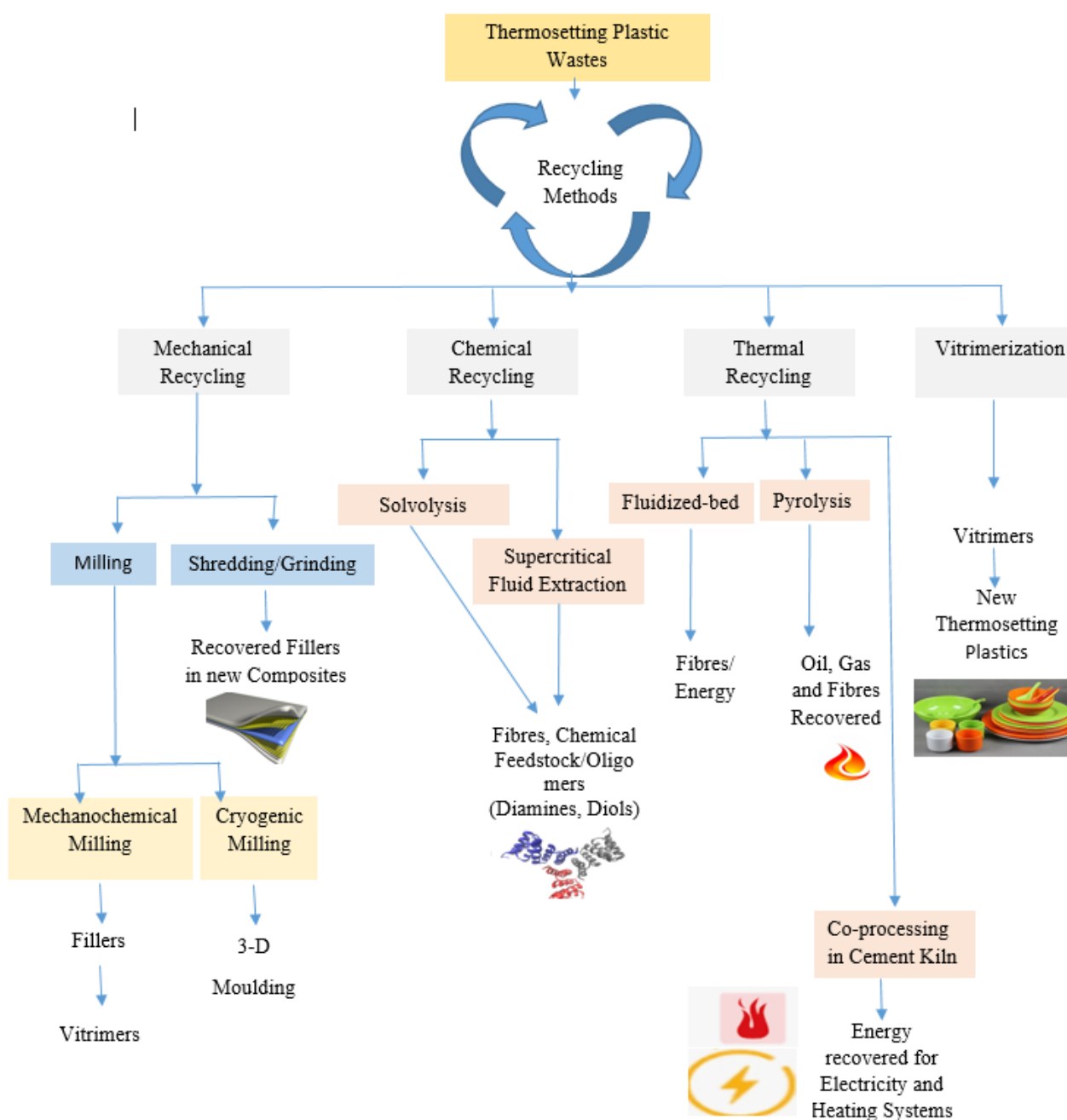


Figure 2. Graphical representation of various methods of thermoplastic and composite waste recycling.

2.1. Mechanical recycling

Mechanical recycling is currently the most commercially available method of recycling plastic waste, including thermosets and their composites [44]. The method is less technical, easier, and more cost-effective when compared to other methods. It consists of collecting, sorting, washing, and size reduction. However, it comes with some challenges for thermosets due to their cross-linked nature. In this method, thermosetting wastes, including their composites, are reduced into smaller particle sizes using any of the following techniques.

2.1.1. Shredding

A plastic shredding machine is used to reduce thermoset waste to a size range of 50–100 mm. The resulting new materials are usually reused as cheapening fillers or reinforcing fillers in asphalt and cement for construction [45,46]

2.1.2. Milling

Specialized hammer mills or ball mills are employed to break down thermoset waste to particles of a size range of 10 mm to 50 μm . This technique results in a finer particle size, which can lead to a matrix-rich product from thermoset waste or a fiber-rich product in the case of thermosetting composite waste. Matrix-rich products are utilized as fillers in new composites, while fiber-rich products are used as reinforcing fillers in new materials where strength is desired. Recent advances in thermosetting plastics milling involve the use of catalysts and/or controlled temperature to prevent significant damage to the chemistry of the material during milling through cryogenic milling and mechanochemical milling.

2.1.2.1. Cryogenic milling

This technique leverages the brittleness of thermosets at low temperature, usually below the glass transition temperature (T_g), to produce ultra-fine powder after impact milling. It involves the use of liquid nitrogen or carbon dioxide to make the thermoset waste extremely brittle while preserving the chemistry of the thermoset without significant degradation of its chemical characteristics [47]. This process produces high-quality impact-milled thermoset powder suitable for 3D printing.

Erin et al. [48] obtained very neat thermoset powder from used 3D materials. These fine thermoset powders were recycled back into neat liquid monomer resin. Their results show that monomers diffused into recycled powder particles create interpenetrating networks upon exposure to ultraviolet light. 3D printing of commercial brittle material with 10 wt% recycled content produced high-quality parts with a maximum stress of about 46.7 MPa and a strain break of 11.6%. Results show that the 3D-printed parts with recycled content presented errors of the same order as neat analogs (52 MPa, 13%).

Cryogenic milling offers several advantages in thermoset processing over conventional milling techniques by ensuring micronizing, nano-scaling end-of-life (EoL) thermosets, which improves solubility and reactivity. Additionally, the cold operating temperature minimizes oxidation and contamination, while the fine particles improve the strength of the new materials, making them suitable for functional applications.

George et al. [49] explored the cryogenic milling of silicone wastes for reuse as soft/elastic filler in silicone formulations for pressure sensors. They reported that the ground silicones increased the dielectric permittivity and mechanical strength of the virgin silicone matrix. They concluded that the silicone-based filler exhibited significant capacitance changes under mechanical compression and maintained a stable response to repeated stress, making it a promising material for soft robotics and medical devices.

2.1.2.2. Mechanochemical milling

This technique involves incorporating a chemical catalyst such as zinc acetate into the thermoset during the ball milling process to enable bond breaking, which renders the thermoset reprocessable as

a vitrimer for remolding [50]. Vitrimers have emerged as a new class of polymers derived from thermosetting plastics that can be reshaped, repaired, and reprocessed without losing their mechanical properties [51, 52]. This novel class of polymer relies on dynamic covalent bonds (DCBs) [50], introduced by esters, disulfides, and silyl ethers to exhibit properties close to those of thermoplastics. A better understanding and subsequent commercialization of the vitrimer technology (vitrimerization) will solve environmental concerns arising from end-of-life thermosetting materials. This process will be discussed in detail below.

2.2. Thermal recycling

Thermal recycling is one of the few commercially viable options for processing thermosets and thermosetting composite wastes due to their cross-linked structure, which prevents melting. The thermal method focuses on the recovery of fibers from thermoset composite waste, generating energy directly or in the form of liquid fuel from the resins.

The two main industrial methods are pyrolysis and fluidized-bed processing, complemented by co-processing in cement kilns.

2.2.1. Pyrolysis of thermoset waste

This method decomposes materials in an oxygen-free environment to recover energy and fibers, typically operating at 450–100 °C [53 and 54]. It avoids combustion by using an inert atmosphere, breaking down the resins into oil and gas while leaving the fibers in the case of thermosetting composite wastes. Studies have shown that catalyzed pyrolysis favors a higher yield of syngas and gaseous fuel compared to uncatalyzed (thermal) pyrolysis for most plastics [53, 54]. Several materials have been employed as catalysts, being widely reported in the literature, with each offering a unique advantage. Key advantages are the lowering of the process temperature, thereby preserving the mechanical properties of the recovered fibers. The quality of the recovered fibers depends on their nature and ability to withstand heat during the process [55]. Consequently, most fibers often suffer a reduction in strength.

Recent developments have demonstrated the use of diverse catalysts and two-step pyrolysis to improve the overall process and quality of products. De Fazio et al. [56] provided a comprehensive overview of recycling technologies for fiber-reinforced plastics (FRP), emphasizing the efficiency of pyrolysis techniques, particularly the two-step approach, in fiber recovery. They reported that pyrolysis of FRP with carbon fibers (CF) demonstrates high efficiency by preserving 93%–96% of the fiber's original mechanical properties, compared with solvolysis, which achieved only 50% of the original mechanical properties. Ginder and Ozman [57] observed that the two-step pyrolysis for glass fiber (GF) recovery, initially at 350 °C and followed by a short period at 450 °C, effectively separates the glass fibers from the matrix. They reported that the glass fiber recovered demonstrated good strength and strain properties.

Wu et al. [58] explored the catalytic pyrolysis of carbon fiber–reinforced epoxy in molten salt as a strategy for recovering carbon fibers. Their results show that the mechanical and surface properties of the recovered fibers were preserved.

In addition, the quality of pyrolysis oils has also been improved by incorporating biomass into the pyrolysis process of thermosetting plastic waste to remove excess moisture and oxygen and also

lower the corrosiveness and viscosity of the oil [59, 60 and 61].

Based on numerous studies reported in the literature, the quality of pyrolysis oil and retention of fiber qualities are critical for thermosetting composite recycling. Pyrolysis and its variants have demonstrated high efficiency in recycling the materials sustainably in a linear loop and in ensuring the realization of a closed loop.

2.2.2. Fluidized-bed processing of thermoset wastes

The goal of this method for thermosetting composite waste valorization is to recover fibers that can be reused, like glass and carbon fibers, while resins serve as fuel to generate energy that is utilized for heating systems. In this thermal oxidative process, shredded mixed thermoset composite wastes are fed into a hot sand bed at a temperature range of 450–600 °C (Figure 3). The resin decomposes, leaving the recovered fibers, which are separated by a rotating screen. The temperature of the fluidized bed is set so as to be able to decompose the thermoset matrices while preserving the fiber. This technique has been widely employed for recovering glass (GF) and carbon fibers (CF) from their respective composites. For polyester resins, a temperature of 450 °C is adequate, while epoxy resin may need up to 550 °C to release fibers.

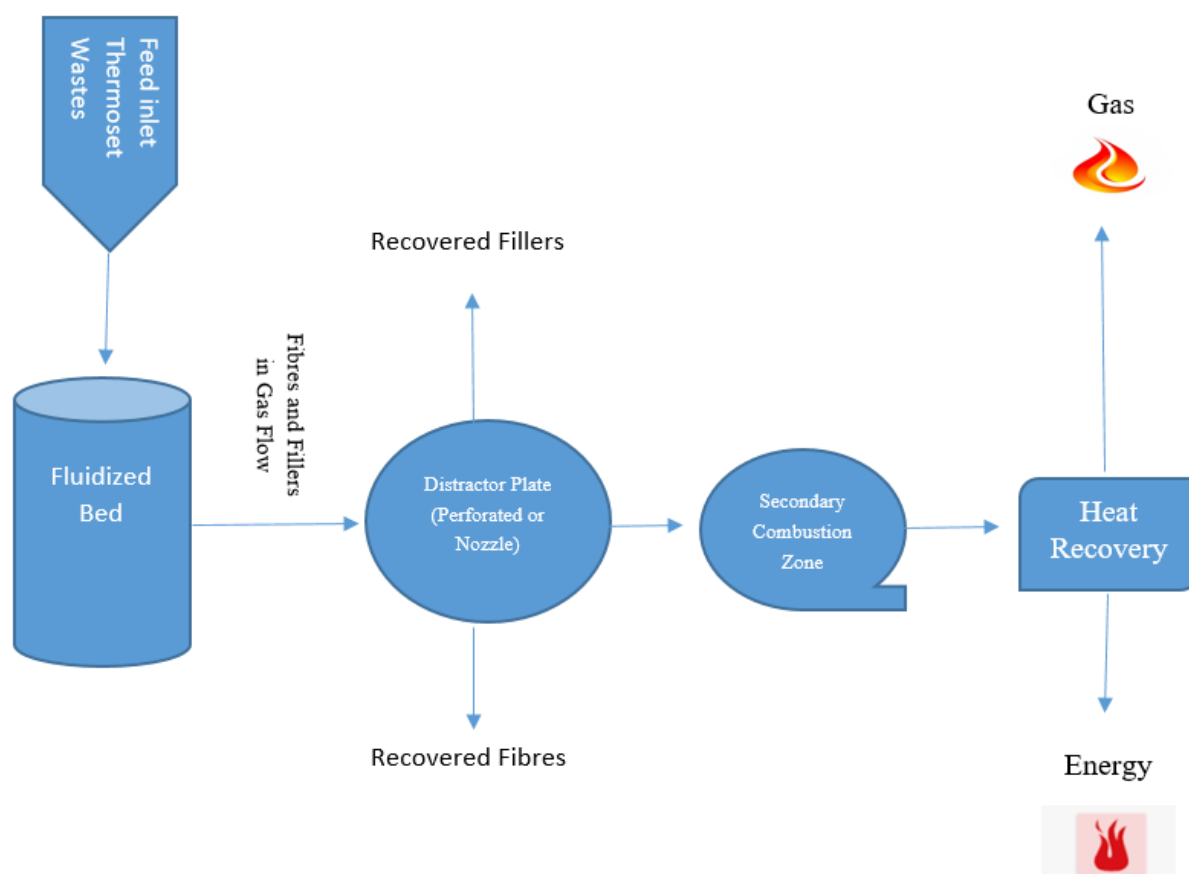


Figure 3. Schematic representation of fluidized-bed processing of thermoset wastes.

The purity of the fiber is usually around 80%, with fiber strength dropping significantly up to 50% due to the high heat generated by the process [62]. Recent research has focused on the modification of

process parameters and post-chemical treatment as a way of improving the recovered fiber properties, particularly for glass fiber [63].

Yaang et al. [64] studied the effects of chemical etching and post-silanization treatment of GF with 80% decreased tensile strength at 500 °C from fluidized bed recovery. However, the post-chemical treatment regained 30%–70% of the lost mechanical properties. Similarly, Thomason et al. [65] employed short-hot sodium hydroxide solution treatment on GF and reported 75% recovery of the lost properties of the fiber.

Pender and Yang [66], on the other hand, studied the effects of CuO, CeO₂, and Co₃O₄ as catalysts in boosting resin decomposition in the fluidized bed process. They reported a 20% increase in the strength of recovered GF, attributed to CuO and CeO₂, whose activities effectively lowered the process temperature to 375 °C and process time to 20 minutes, drastically reducing energy requirement.

Currently, research has shown that fluidized-bed processing of thermoset wastes is viable for recovering GFs and CFs from glass fiber–reinforced composites (GFRPs) and carbon fiber–reinforced polymer composites (CFRPs). Overall, this process closes the linear loop of end-of-life (EoL) GFRPs and CFRPs and promotes circularity, lowering pressure on landfills and the environment.

2.2.3. Co-processing in cement kilns

Cement kiln co-processing is currently considered one of the most effective and sustainable methods for recycling glass fiber–reinforced thermoset composites [67]. The aim is to integrate the mineral component into cement production as an alternative raw material, while the high-calorific-value thermoset matrices serve as fuel in place of coal or gas. The silica, calcium, and alumina from the glass are similar to the minerals used in cement manufacturing [67]; therefore, they can be utilized as clinker. This is an excellent valorization technique for glass fiber composites, although not suitable for carbon fiber composites due to their lower heat tolerance compared to glass fibers. The process is highly efficient, offering up to 67% material recovery and 33% energy recovery, with carbon dioxide emissions as low as 16%, without any residual wastes left [68].

2.3. Chemical recycling

Chemical recycling of thermosetting and plastic composite wastes is a recovery method that converts end-of-life materials into valuable resources for new applications. In this process, highly cross-linked matrices are recovered as oligomers, while glass and carbon fibers are simultaneously recovered for reuse. The advantage of this method, though expensive, is that high-quality fibers and usable starting materials (oligomers) are obtained. Key techniques of the process are categorized into two major methods: solvolysis and supercritical fluid extraction.

2.3.1. Solvolysis

Solvolysis is a chemical recycling method that uses solvents under suitable temperature and pressure to break down highly cross-linked end-of-life thermoset or thermosetting composites, making up for the recovery of useful chemicals and reinforcing materials for reuse. In this method, waste thermosets are dissolved using suitable solvents, heat, and catalysts to recover oligomer and fibers such as glass and carbon fibers. The glass and carbon fibers that have been recovered through this

method retain up to 95% of their original strength [69–71]. The major concern of this method is the environmental and health concerns associated with most hazardous solvents, its high energy demand, and the solvent recovery efficiency.

Solvolysis is classified according to the type of solvent employed based on suitability for a particular matrix: hydrolysis (water as solvent), methanolysis (methanol as solvent), and glycolysis (glycol as solvent). The key solvolysis reactions are as follows:

Epoxy + Glycol $\rightarrow$ Bisphenol A-based oligomers

Polyester + Water $\rightarrow$ Diol + Dicarboxylic acids

Epoxy + Amine $\rightarrow$ Functionalized oligomers

The choice of solvent, catalyst, and other processing parameters depends on the solid–solvent interaction parameter. Pantazidou et al. [72] reported an 83% recovery of glass fibers from end-of-life wind turbine blades (WTB) made from glass fiber–reinforced polyester composites. In their study, polyethylene glycol was used as the solvolysis agent at 200 °C and ambient pressure. They employed 12.5 g of sodium hydroxide as the catalyst on 10 g of WTB for 6 h. Furthermore, Omar et al. [73] investigated the chemical recyclability of glass fiber–reinforced epoxy composites from four-layer waste mill blades. They identified sulfuric acid as the most effective solvent, with maximum yield obtained at 87.5 °C, 100% (v/v) sulfuric acid in distilled water, and a reaction time of 48 h.

2.3.2. Supercritical fluid extraction

Supercritical fluid (SCF) recycling is a form of chemical recycling that uses high-pressure, high-temperature fluids to break down the cross-linked structure of thermosets and thermosetting composites, allowing for the recovery of clean fibers and chemicals for reuse. The process involves using supercritical water, supercritical methanol, or supercritical carbon dioxide with co-solvent-linked polyurethane matrices. At the supercritical state, the fluid has gas-like penetration (low viscosity, high diffusion) for dissolving materials. Supercritical water is a greener and safer solvolysis agent, with no harmful emissions, but is associated with high supercritical parameters: a critical point temperature of 374 °C and pressure of 22.1 MPa [74–76]. Alcohol, on the other hand, with a critical point temperature of 250 °C and pressure of 4.8 MPa [77,78], is the better alternative. Recent research works have focused on the potential use of various organic solvents and catalysts to lower the process temperature and pressure to save energy and time [79].

Raul et al. [80] studied the recycling of carbon fiber–reinforced epoxy composites using subcritical and supercritical alcohols as extraction media. The alcohols methanol, ethanol, 1-propanol, and acetone were used as solvent reagents in experiments in batch and semi-continuous reactors at a temperature range of 200–450 °C while investigating the impact of flow rate and the addition of sodium hydroxide and potassium hydroxide as catalysts. Results showed 98 wt% resin recovery at 350 °C with 1.1 kg alcohol/kg fiber/min; at the same solvent flow rate but at 275 °C with 0.02 mol/L of KOH, a 96.5 wt% resin recovery was obtained.

Raul et al. [81] carried out chemical recycling of carbon fiber–reinforced composites in near-critical and supercritical water. They investigated which parameters influenced the recovery of useful resources, including time, temperature, oxidant, and catalyst concentration. The experiment was conducted in a batch reactor at 250–400 °C and 4.0–27 MPa, with a reaction time of 90 min. The

efficiency of resin recovery was reported to be 78.3 wt% under the supercritical water condition. When potassium hydroxide was used as the catalyst, resin recovery significantly improved to 95.3 wt%, as did fiber strength.

Currently, many compounds are being investigated to find more effective catalysts for SCF recycling of thermosets and thermoset composites [82–84].

3. Vitrimers

Thermosetting plastic wastes cannot be melted to be reshaped into new items due to their highly cross-linked structural chemistry; as such, they are said to have an irreversible heat cycle. This property becomes a limitation to their recycling through commonly used secondary processing methods like injection, compression, blow, and rotational molding techniques [85,86]. Consequently, the techniques discussed above (pyrolysis, solvolysis, SCF, and other pathways) are a common practice for thermoset materials. However, these methods also present challenges: high equipment cost, huge energy demand, high technological know-how, and environmental and health risks. Recent breakthroughs have emerged regarding the possibility of incorporating compounds into thermosets to make them melt upon application of heat rather than char. The full benefits of highly cross-linked thermoset materials can be realized simultaneously, as their end-of-life waste can also be heated and reused like thermoplastics. This has been achieved by introducing dynamic covalent bonds (DCBs) into thermosets; the resulting new materials are called vitrimers and were first reported by Leibler and his group in 2011 [87].

Vitrimers are a class of plastics that combine the characteristics of thermosets with the recyclability of thermoplastics. Vitrimers contain DCBs, which impact associative covalent adaptable networks (CANs) that ensure a steady cross-link density of the material regardless of the type of stimulus. These compounds include boronic esters, disulfides, and silyl ethers, which allow bonds to swap partners at high temperatures within the solid networks. This results in a material flow while retaining its cross-linked structure. Interestingly, vitrimers are not only recyclable but also repairable. They do not dissolve in solvents but can be reshaped under heat and pressure. Therefore, self-healing can be triggered thermally. Key properties of these new materials are compared with those of thermoplastics and thermosets in Table 1.

Table 1. Properties of thermoplastics, thermosets and vitrimers.

Properties	Thermoplastics	Thermosets	Vitrimers
Reshapeability	Yes	No	Yes (Heat/Pressure)
Recyclability	Yes	Yes	Yes (Heat/Pressure)
Self-healing	No	No	Yes (Heat/Pressure)
Cross-linked	No	Yes	Yes
Creep resistance	No	Yes	Yes
Solvent resistance	Variable	Yes	Yes
Bond exchange	Not applicable	No	Yes
Reworkable	Yes	No	Yes

Since the first report on vitrimers in 2011, several vitrimeric materials have been studied and reported in literature, exploring different polymeric backbones, chemical bonds, and catalyst systems [88–90]. All vitrimer materials share the properties presented in Figure 4, making them suitable for

applications such as shape memory materials, reusable adhesives, and 3D printing.

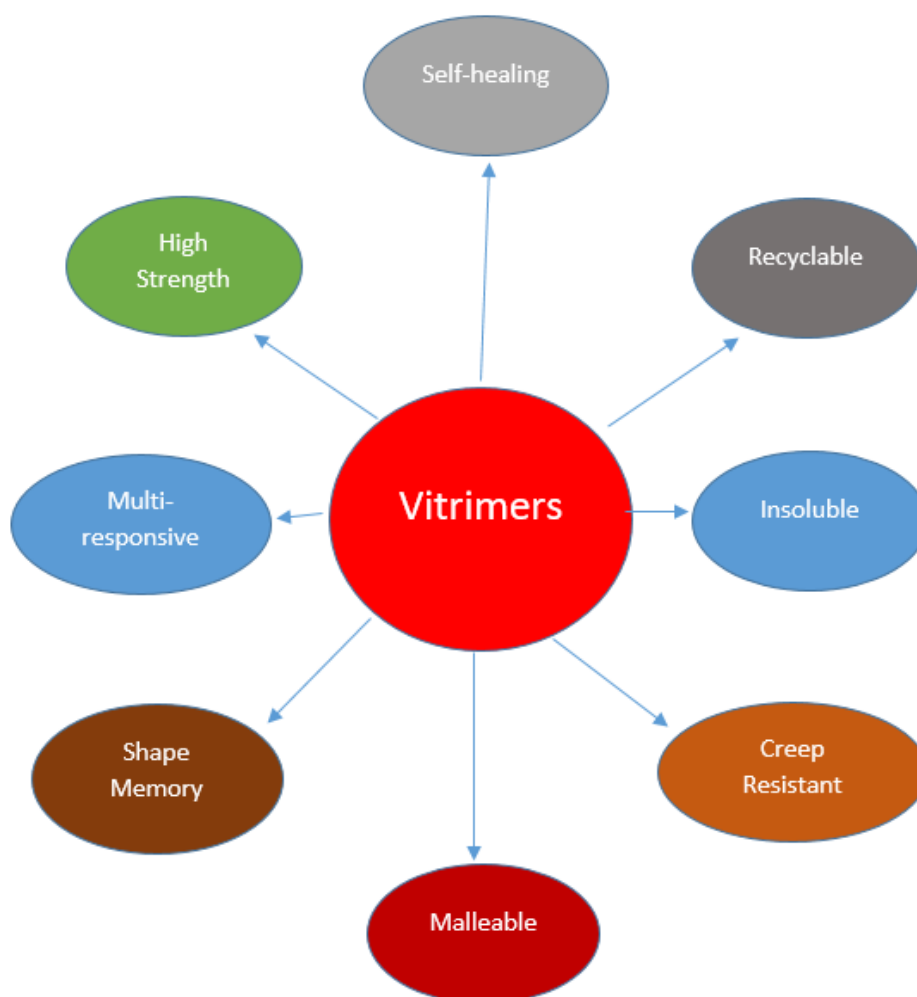


Figure 4. Unique properties of vitrimers.

The choice of appropriate DCBs for a particular material will enable the bonds to swap effectively and flow like thermoplastics when triggered by external stimulus, such as heat. The chemistry of vitrimers is such that the bonds are associative, meaning that old bonds break and new bonds recombine simultaneously, ensuring that the cross-linked density is preserved, thereby retaining the properties of thermosets (Figure 5). Hence, DCBs can be termed associative dynamic network (ADN) structures. Some of the groups that can create DCBs include hydroxyl groups, ester groups, disulfide exchange, imine exchange, and vinylogous urethane groups.

The choice of DCB and specific chemistry depends on the monomers or the EoL thermoset and the desired properties of the vitrimer material, such as its mechanical strength, thermal stability, and recyclability.

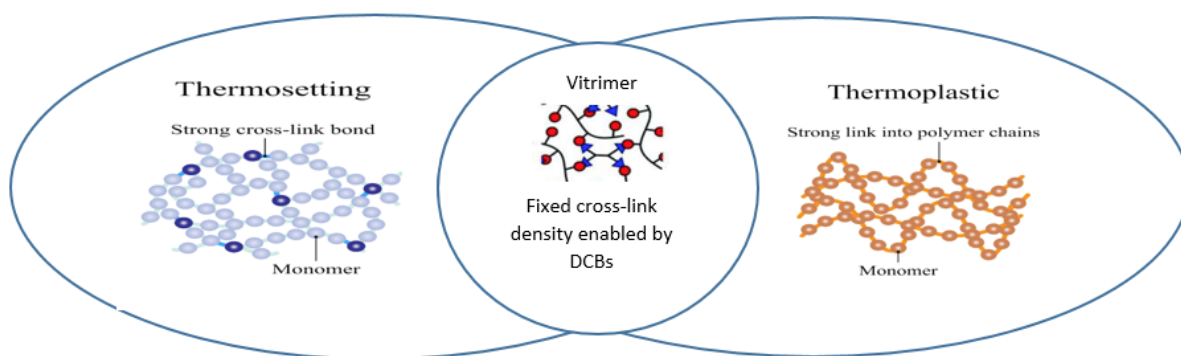


Figure 5. Characteristics of bonds in thermosets, thermoplastics, and vitrimers.

3.1. Primary and secondary bonds in thermoplastics, thermosets, and vitrimers

The thermal behavior of polymers is the basis for their classification into three major classes. Their behavior after heat exposure is dictated by two distinct types of bonds: intramolecular bonds (primary bonds) and intermolecular bonds (secondary bonds).

The primary bonds are those that link individual atoms together to form long, continuous polymer chains; these are referred to as the backbones. The secondary bonds are those between the separate polymer chains, which enable the bulk state of the polymer.

Three distinct classes of plastics are based on the nature, strength, and type of primary and secondary bonds holding atoms and chains within the plastic, as shown in Table 2. How these bonds respond to heat and pressure also determines the recyclability, reshapeability, and self-healing ability of the plastics (Figure 6).

Table 2. Plastic types and nature of primary and secondary bonding.

Plastic types			
Bond types	Thermoplastics	Thermosets	Vitrimers
Primary bonds (intramolecular forces)	Strong covalent carbon–carbon and carbon–hydrogen	Strong covalent	Strong covalent
Secondary (intermolecular forces)	Weak electrostatic attraction (Van der Waals)	Weak electrostatic attraction (Van der Waals); strong covalent bonds form cross-links as a third bond type within the secondary or intermolecular structure	Dynamic covalent bonds (DCBs) form cross-links

Thermoplastics have strong covalent primary bonds that provide the plastic with its fundamental structural integrity and tensile strength. These primary bonds remain intact during heating, so that the plastic does not decompose or burn upon melting for semi-crystalline plastics. Thermoplastics also have secondary bonds—mainly weak physical attractive forces between separate chains, including Van

der Waals forces and hydrogen bonds. Because these secondary bonds are weak, they are sensitive to temperature, enabling the polymer chains to slide past one another upon heating and reforming upon cooling. As such, thermoplastics flow like viscoelastic fluids when heated above the glass transition temperature (T_g) or melting temperature (T_m) for semi-crystalline polymers [91].

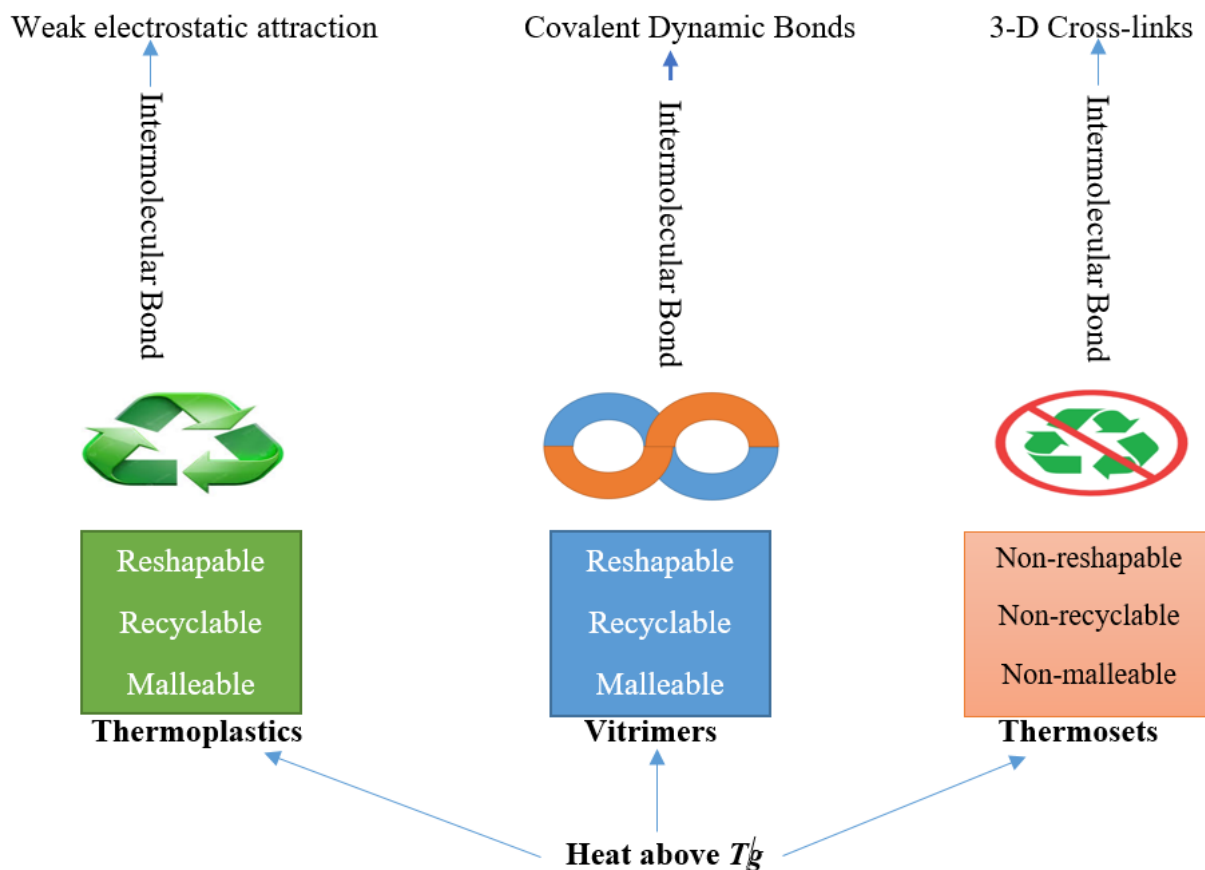


Figure 6. Reclassification of plastics based on the dominant intermolecular bond type and response to heat.

Thermosetting plastics, on the other hand, exist as independent linear chains, possessing strong covalent primary bonds and weak secondary bonds before cross-linking [92]. Upon the application of heat and catalysts, additional third network bonds (dimensional network) form from reactive or functional monomers through strong covalent bonds. Once this 3D network is formed, they dominate the weak secondary bonds in the thermosets, impose rigidity, and hinder the mobility of secondary bonds after heat application. However, above T_g , the cross-linked polymer chains gain significant mobility to soften and behave like tough rubber, but remain in shape because they never melt [93].

Vitrimers or covalent adaptable networks (CANs) are a class of plastics having covalent primary bonds, such as thermoplastics and thermosets. They also possess cross-linked covalent bonds (similar to thermosets) but, in addition, present dynamic covalent bonds (DCBs) as secondary or intermolecular bonds. These bonds can undergo thermally activated exchange reactions; they break and reform in a controlled manner [94], allowing the polymer to flow and reshape or recycle without destroying its geometry. These materials retain their structure or geometry upon heating due to their ability to

maintain a steady cross-link density. The thermal behavior of vitrimers is governed by both T_g and topology freezing temperatures (T_v). Like thermosets, vitrimers become softer and more flexible above T_g ; above T_v , the dynamic exchange is activated, allowing the material to flow like thermoplastics.

Vitrimers, therefore, are a class of plastics that merges the malleability of thermoplastics with the performance advantages of thermosets.

3.2. *Production processes of vitrimers*

There are two primary routes to create vitrimers, as described below.

3.2.1. Direct synthesis from small molecules (monomers)

The first step is the synthesis of the pre-polymer followed by the mixing of multifunctional monomers containing DCBs such as epoxy, acids, or amines with catalysts under appropriate temperature. A typical example is the heating of epoxidized soybean oil with a diamine and diacrylate to build a cross-linked material with exchangeable bonds.

3.2.2. Upcycling from existing polymers

This method gives a new life to common EoL plastics, using techniques like reaction extrusion (REX). This process is particularly beneficial for EoL thermosets, being termed vitrimerization.

4. **Vitrimerization**

Vitrimerization is the process of breaking down the permanent network structure of plastics and rebuilding them into CANs by introducing DCBs; the newly formed material is called a vitrimer.

The emergence of vitrimers is dealing with the major limitation of end-of-life thermosetting items; nowadays, the durability, strength, and heat/chemical resistance of thermosets can be fully utilized, together with their recyclability, similar to thermoplastics.

Vitrimerization of thermoset waste is the process of altering the bond chemistry of a thermoset by introducing dynamic covalent bonds, which replace the highly cross-linked bonds with covalent adaptable networks (CANs), enabling associative exchange reactions such as transesterification, disulfide exchange, siloxane exchange, silyl ether exchange, olefin metathesis, and imine/vinylogous urethane exchange [95–97]. These allow network rearrangement without losing integrity. Because new bonds must form before old bonds are broken down, the cross-link density never lowers, unlike in thermosets [98]. Vitrimers present DCBs that are associative rather than dissociative; consequently, they possess infinite viscosity (they flow, but never dissolve) and no creep, i.e., below the topology freezing temperature.

The standard procedure for this process is as follows:

- i. **Size reduction:** Waste is ground into fine particles to minimize the surface area for a better chemical reaction through mechanochemical or cryogenic milling (both processes were discussed in the previous section).
- ii. **Catalyst integration:** A catalyst is mixed with fine particles, occasionally with a solvent or hydroxyl-providing agent, to facilitate catalyst diffusion.

- iii. **Network activation:** The mixture undergoes a mechanochemical process to drive the catalyst into the polymer network, forming active sites for future exchange reactions.
- iv. **Remolding:** The activated powder is heated and compressed using standard plastic molding equipment, such as compression molding, which activates the DCBs.
- v. **Confirmatory test:** Comparative studies of Fourier-transformed infrared spectroscopy (FTIR) spectra of vitrimerized and non-vitrimerized samples provide information on the formation of metal–ligand complexes. Dynamic mechanical analysis (DMA) evaluates the stress–temperature relation.

These procedures were reported by Liang et al. [99], who demonstrated the conversion of epoxy thermosets to epoxy-vitrimer through transesterification using zinc acetate as the catalyst. The fabricated epoxy-vitrimer displays comparable properties with the virgin epoxy; when reprocessed, it retains its properties. The mechanochemical vitrimerization process allows for the formation of metal-carboxylate ligands that serve as junctions for the transesterification exchange reaction. Consequently, polyester-based thermosets, such as epoxy, vinyl ester, and polyurethane, can be converted into vitrimer networks relying on metal-carboxylate ligands. Any thermoset material that has ester groups and freely available hydroxyl groups can be infused with a catalyst that supports transesterification, thereby being converted into a vitrimer through the procedure described above.

Liang et al. [100] demonstrated the vitrimerization of epoxy and polyurethane thermosets using tin (II) 2-ethylhexanoate ($\text{Sn}(\text{Oct})_2$) as the catalyst for the transesterification reaction. They compared the properties of vitrimerized and non-vitrimerized epoxy and polyurethane samples. Stress relaxation showed that unrecyclable thermosets were successfully converted into vitrimers using the vitrimerization process stated above. Both the vitrimer and vitrimerized networks relaxed significantly due to the transesterification reaction. Furthermore, after recycling the vitrimers, 44% of modulus, 30% of ultimate strength, and 78% of extensibility for PU could be recovered. In the case of the epoxy system, 73.5% of modulus, 63% of ultimate strength, and 82% of extensibility were recovered. The decrease in properties was attributed to the infusion of the catalyst. The presence of the catalyst has a deleterious effect on the mechanical and thermal properties of the vitrimer. These findings align with the work reported by Leibler et al. [101].

Bandegi et al. [102] employed a mechanochemical approach to study the vitrimerization of cross-linked poly(ethylene-vinyl acetate) (EVA) using zinc acetate as the catalyst and varying contents of poly(vinyl-alcohol) as a feedstock of hydroxyl groups. They reprocessed the vitrimerized EVA three times without additional catalyst, in order to study its recyclability. Results showed that the EVA sustained its properties after being recycled multiple times and relaxed rapidly at temperatures as low as 100 °C.

Other metal ligands have also been reported in the literature, such as zinc acetylacetonate, zinc octoate, zinc stearate, and zinc trifluoroacetate hydrate [103–105].

Vitrimerization allows for the reusing, reworking, and upcycling of unrecyclable thermosets, with no significant loss in their original properties, thereby enabling zero waste from the thermosetting plastic fraction.

SWOT analysis of the methods for recycling thermosetting plastics and their composites.

Strength Mechanical recycling *Low energy consumption *Technologically simple *High throughput Thermal recycling *Handles mixed wastes effectively *Recovers energy *Recovers fibers Chemical recycling *Clean long fibers are recovered *Pure monomers and chemicals are recovered Vitrimer *Reparable *Weldable *Recyclable like thermoplastics *Enables innovation	Weakness Mechanical recycling *Produces low-value fibers *Other recovered materials are downcycled Thermal recycling *High energy demand *Fiber surfaces often have char residue *Degrades fiber strength *Risk of harmful emissions Chemical recycling *High chemical costs *High capital costs *Process is resin-specific, i.e., careful sorting is required Vitrimer *Technology is recent and expensive *Currently associated with trade-offs such as improved strength, which makes reprocessing difficult
Opportunities Mechanical recycling *Serves as a low-cost feedstock for construction materials Thermal recycling *Recovered energy can reduce plant operating costs *Viable for low-value waste Chemical recycling *Enables true closed-loop recycling for high-value sectors such as aerospace and wind energy Vitrimers *Technology is highly promising as the future of thermosets	Threat Mechanical recycling *Market saturation of low-value fillers and completion from cheaper virgin materials Thermal recycling *Stringent emission regulations required *Volatility in recovered oil prices Chemical recycling *Economic viability depends heavily on the resale value of recovered fibers, which is volatile Vitrimers *Stiff competition against established low-cost thermoset reprocessing alternatives *Scalability is still at an infant stage

The SWOT analysis of the various techniques for recycling thermosets and their composites shows that each method is aimed at avoiding end-of-life thermosetting materials making their way to

landfills. However, the choice of a particular method will largely be influenced by factors such as the desired output quality based on the application of the recovered materials, process cost, energy recovery quality, and technological readiness level (TRL) of the method. Mechanical recycling provides the cheapest option, thermal processing provides a more energy-recovery option, and the chemical process provides high-quality fibers through solvolysis. The technique of the future is clearly vitrimerization, but its TRL is still at an infant stage. Scalability issues of transitioning from laboratory-scale synthesis to a large-scale, cost-effective industrial production remain a current hurdle.

5. Conclusions

The inherent cross-linked structure of thermosetting plastics and their composites precludes direct melt-processing; yet, this review confirms that viable recycling pathways exist, each with distinct technical and economic trade-offs. Mechanical recycling offers a low-energy, surface-level solution but produces irregular fillers with diminished properties. Thermal recycling (pyrolysis and fluidized bed) efficiently recovers energy and fibers but tends to degrade the reinforcing materials and release hazardous byproducts. Chemical solvolysis emerges as the most promising approach for high-value material recovery, capable of selectively breaking down the resin matrix to reclaim both clean fibers and oligomeric fractions. However, widespread industrial adoption remains hindered by high solvent costs (supercritical fluid extraction), long reaction times, and the complexity of managing mixed waste streams. Future progress will depend on integrating low-temperature catalytic systems, which are already being explored, and developing selective separation technologies.

Ultimately, the emergence of vitrimers, though still at a low technological readiness level, will change the entire plastic technology ecosystem—enabling designing recyclable plastics that will combine the durable properties of thermosets while still ensuring a sustainable circular economy for these durable, high-performance materials.

Use of AI tools declaration

The authors declare they have not used Artificial Intelligence (AI) tools in the creation of this article.

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Conflict of interest

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

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