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Review

Pultrusion and Vitrimer Composites: Emerging Pathways for Sustainable Structural Materials

Vishal Kumar [1,*], Khaled W. Shahwan [1], Wenbin Kuang [1], Kevin L. Simmons [1], Philip Taynton [2] and Emily R. Cieslinski [3]

[1] Energy and Environment Directorate, Pacific Northwest National Laboratory, Richland, WA 99354, USA; khaledshahwan@yahoo.com (K.W.S.); wenbin.kuang@pnnl.gov (W.K.); kl.simmons@pnnl.gov (K.L.S.)
[2] CEO Mallinda Inc., Denver, CO 80229, USA; philip@mallinda.com
[3] PulFlex Technologies, LLC, Ford City, PA 16226, USA; ecieslinski@pulflex.com
* Correspondence: vishal.kumar@pnnl.gov

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Abstract

Pultrusion is a manufacturing process used to produce fiber-reinforced polymer composites with excellent mechanical, thermal, and chemical properties. The resulting materials are lightweight, durable, and corrosion-resistant, making them valuable in aerospace, automotive, construction, and energy sectors. However, conventional thermoset composites remain difficult to recycle due to their infusible and insoluble cross-linked structure. This review explores integrating vitrimer technology a novel class of recyclable thermosets with dynamic covalent adaptive networks into the pultrusion process. As only limited studies have directly reported vitrimer pultrusion to date, this review provides a forward-looking perspective, highlighting fundamental principles, challenges, and opportunities that can guide future development of recyclable high-performance composites. Vitrimers combine the mechanical strength (tensile strength and modulus) of thermosets with the reprocessability and reshaping of thermoplastics through dynamic bond exchange mechanisms. These polymers offer high-temperature reprocessability, self-healing, and closed-loop recyclability, where recycling efficiency can be evaluated by the recovery yield retention of mechanical properties and reuse cycles meeting the demand for sustainable manufacturing. Key aspects discussed include resin formulation, fiber impregnation, curing cycles, and die design for vitrimer systems. The temperature-dependent bond exchange reactions present challenges in achieving optimal curing and strong fiber–matrix adhesion. Recent studies indicate that vitrimer-based composites can maintain structural integrity while enabling recycling and repair, with mechanical performance such as flexural and tensile strength comparable to conventional composites. Incorporating vitrimer materials into pultrusion could enable high-performance, lightweight products for a circular economy. The remaining challenges include optimizing curing kinetics, improving interfacial adhesion, and scaling production for widespread industrial adoption.

Keywords: pultrusion; vitrimer; recyclability; self-healing; sustainability

1. Introduction

1.1. Overview of Composite Materials

Composites have had a significant impact on human civilization, ranging from prehistoric mud and wooden shelters to innovative spacecraft components. The term “composite” refers to a combination of two or more materials designed so that the properties of the sum have more desirable attributes than those of the individual constituents. This augmentation occurs by incorporating a discontinuous or reinforcement phase into a continuous matrix [1]. Polymers have progressively replaced metals and ceramics in a variety of applications over the last few decades due to some of their attractive attributes such as resistance to corrosion, lower density, manufacturability into more complex shapes, and sometimes lower cost [2–4]. However, virgin polymers often do not have adequate strength, stiffness, and other key characteristics, limiting their usefulness in some demanding applications. To enhance the properties of the polymer, fibrous reinforcement is added. Incorporating fibers into polymers can significantly enhance their mechanical properties including tensile strength, flexural modulus, and fatigue resistance often surpassing those of metals in strength-to-weight ratio, stiffness, and durability. Fiber-reinforced polymers (FRPs) have significantly replaced metals in industries ranging from construction to aerospace, military, automobiles marine, sports, storage containers, and transportation [1,5–10]. Typically, these composites include matrices like epoxy, phenol, polyester, or vinyl ester combining with reinforcing fibers like carbon, glass, aramids, or natural fibers. Owing to their superior properties, durability, and lightweight nature, fiber-reinforced polymer (FRP) composites are widely used in various industrial applications. However, despite their advantages, FRP composites present considerable difficulty in terms of reuse and recycling offering substantial environmental and economic issues [11–15]. While fiber-reinforced thermoplastics can be recycled through mechanical, thermal, or chemical methods, the majority of high-performance FRPs are made from thermoset resins, which pose greater recycling challenges due to their crosslinked structure. This inherent rigidity prevents remelting, making conventional recycling techniques ineffective and necessitating the development of alternative recycling strategies. Historically, discarding including burial into landfills and incineration remained the most common methods of waste disposal for thermoset-based FRP materials. Currently, significant efforts are being made to address the issue using improved mechanical, thermochemical, and chemical recycling, as well as reclamation techniques [16–21]. Nonetheless, the difficulty of recovering thermoset resins and efficiently reusing recovered fibers has prompted substantial study into the development of reprocessable and adaptable thermosets. This is accomplished by utilizing reversible and dynamic cross-linked covalent bonds, which include transesterification, disulphide exchange, imine exchange, and other reversible bond exchange mechanism [22–25]. These developments in recyclability are accompanied by increasing focus on environmental effect and sustainability. In parallel, extensive research is being performed to substitute synthetic fibers and polymers with bio-based polymers and natural fibers, with the goal of achieving biodegradability at the end of their lifespans [26–31].

1.2. Introduction to Vitrimers

One of the most intriguing and crucial approaches for recycling thermosets is to change the chemical structure of the polymers before using them in composite manufacturing. Over the last 10 years, major efforts have been made to manufacture thermosets that are reprocessable and recyclable. The crosslinked network structure of thermosets, while necessary for their exceptional mechanical qualities, often precludes reprocessability. To overcome this issue, researchers have created polymer networks with reversible or exchangeable covalent networks, known as covalent adaptive networks (CANs), that combine the benefits of thermosets and thermoplastics. These networks enable thermoset mechanical stability while providing thermoplastic malleability and high temperature reprocessability through reversible bond exchange processes. This novel class of polymers, with a permanently crosslinked structure and dynamic topology that allows for bond rearranging, is known as “vitrimer,” a term coined by Leibler and colleagues [32–35]. Using vitrimer in FRP composites is a highly sustainable and promising recycling strategy that allows for the manufacture of, reprocessable thermoset components in an economically and environmentally beneficial manner. Extensive research is already ongoing to develop reprocessable, repairable FRP composites with vitrimer matrices. Mallinda Inc. created VITRIMAX™, a polyimine-based vitrimer resin that can be bent, molded, and welded for closed-loop reuse with both solution-based and solid states [36]. Notably, this vitrimer is employed as a matrix in carbon-fiber composites, resulting in a fully closed-loop system for recovery and reuse of both the fiber and the resin. The recyclability of vitrimers is not only structural but also quantifiable, with efficiency evaluated by retained mechanical performance and material recovery ratios across multiple recycling cycles.

1.3. Manufacturing of Polymer Composites

Polymer composites are produced through a variety of processes that are specifically designed to accommodate the matrix properties. Thermoset composites, which are thermally stable and have high strength, are typically manufactured using hand lay-up technique, resin transfer molding (RTM), filament winding, and pultrusion. In contrast, thermoplastic composites, which are regarded for their toughness and recyclability, are frequently made by injection molding, compression molding, thermoforming, and pultrusion. Among these methods, pultrusion is a continuous, automated process that enables excellent production efficiency and cost-effectiveness, particularly for components with uniform cross-sections [37,38]. Moreover, pultrusion supports both thermoset and thermoplastic resins, offering material versatility and flexibility in design for complex shapes.

1.4. Pultrusion Process: Overview

Pultrusion is a continuous manufacturing process that produces fiber-reinforced composite products with uniform sections, such as prismatic beams and rods. Reinforcing fibers, such as synthetic and natural fibers, are pulled through a resin bath to impregnate them with a thermosetting or thermoplastic resin, after which the composite material is shaped and cured in a heated die while being continuously pulled, hence the name "pultrusion". The process of pultrusion is displayed in Figure 1 shoes how the fiber enters the resin bath through guide plate and forms a required shape in the die and curing takes place. It also demonstrates how the cured composite is pulled out of the die and cut in to required size. Pultrusion is an attractive manufacturing process because it is highly automated that can produce large quantities of profiles at relatively much lower capital investment and manufacturing costs for industries such as building, automotive, and aerospace [39–42]. Vitrimers are currently being investigated as a potential type of recyclable thermosetting polymer, characterized by dynamic covalent networks that enable reprocessing and repair while preserving structural integrity [33]. They have shown potential for conventional molding and composite production, but their application in continuous manufacturing processes like pultrusion is still limited. Current research indicates that the versatility of resin formulations poses a considerable challenge, as vitrimer matrices must simultaneously satisfy the requirements of low viscosity, rapid curing, and controlled topology exchange reactions during pultrusion techniques [35]. Furthermore, the combination of process parameters such as pulling speed, die temperature, and resin infiltration with dynamic bond exchange kinetics presents optimization challenges that are markedly discrete from those encountered with traditional thermosets. Even though more people are interested in sustainable composites, there are not many thorough studies on these subjects. This study seeks to fill existing gaps by investigating the amalgamation of vitrimer and pultrusion, pinpointing current formulation and manufacturing constraints, and proposing strategies for the development of scalable, reusable alternatives to conventional pultruded thermoset composites.

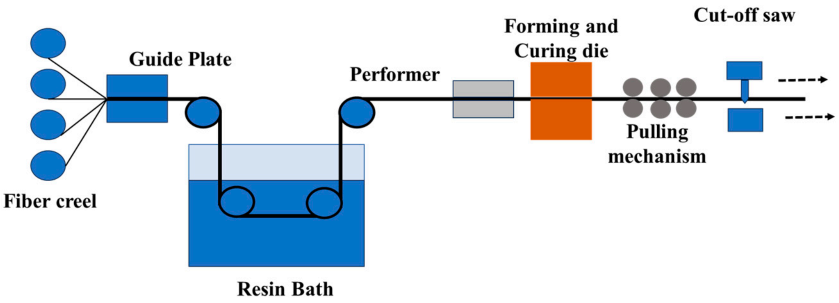

Figure 1. Schematic of pultrusion process with key stages (fiber impregnation, resin curing, die shaping, pulling, and cutting) indicated.

This paper provides a comprehensive analysis of the pultrusion process, detailing each critical stage, including fiber impregnation, resin curing, die shaping, continuous pulling, and precise cutting. It explores the influence of key processing parameters such as resin type, viscosity, fiber type, volume fraction, fiber orientation (predominantly longitudinal in pultrusion, with occasional transverse or ±45° reinforcements), die geometry, temperature, and cure cycle on the final properties of the composite product. Additionally, the review highlights emerging trends in pultrusion technology, focusing on the integration of advanced materials and improved process control strategies. Notably, the development of innovative polymer resins, particularly dynamic covalent network polymers such as vitrimer, is expected to revolutionize pultruded composites by enhancing their recyclability, durability, and overall performance. These advancements are poised to expand the scope of pultrusion applications, driving the production of more sustainable, high-performance composite materials across a wide range of industries.

2. Pultrusion Process

2.1. Preparation of Resin

The processing of resin is a critical step in the pultrusion process, essential for producing composite materials with consistent and uniform properties. The process begins by choosing a resin system that aligns specific manufacturing and target performance requirements, with common options including polyester, vinyl ester, polyurethane, and epoxy resins [43–45]. Precise quantification of the resin components, including the basic resin, curing agents, accelerators, and additives, is essential for achieving the desired material properties [46–48]. The components are carefully blended to produce a uniform mixture with acceptable viscosity that minimizes air bubbles and ensures a uniform distribution of additives [49–51]. A degassing approach can be utilized to remove entrapped air, hence minimizing bubbles and voids, and improving resin performance and manufacturing quality [52,53]. Fillers and additives, like calcium carbonate and alumina trihydrate, are commonly employed as cost-effective alternatives to enhance properties such as creep resistance and fire resistance. However, careful selection and precise dosage of these fillers are crucial to maintain optimal workability, surface quality, and overall performance of the composites [54]. These additives can also influence factors such as velocity, die temperature, and tool wear [55–57]. The resin is stored under controlled conditions to prevent premature curing, maintaining its stability until it is used in the pultrusion [56].

2.2. Impregnation of Fiber

The fiber impregnation phase is a crucial step in the pultrusion technique. The process begins with the preparation of reinforcing materials, typically in the form of roving composed of glass, carbon, or aramid fibers [58]. The fibers are drawn into a resin bath for wetting which is essential to achieve consistent strength and interfacial bonding characteristics. After impregnation excess resin is subsequently removed, and the impregnated fibers pass through a guiding system for proper alignment [59]. Ensuring material compatibility of the resin and fiber is necessary for good adhesion and mechanical properties [60]. Moreover, factors such as resin viscosity, pulling speed, and the taper angle at the die entrance impact the pressure within the pultrusion die, thereby directly impacting fiber impregnation quality and void formation [61,62]. Manufacturers often strive to maintain void content below 5% to ensure optimal mechanical properties. Beyond traditional methods, advanced techniques have been developed to improve impregnation efficiency. Volk et al. [62] and Dubé et al. [63] examined a closed injection impregnation procedure, where resin is injected directly into the die. This method enables precise control over the resin-to-fiber ratio, promotes rapid impregnation, and minimizes exposure to volatile solvents, enhancing both process efficiency and environmental safety compared to conventional pultrusion techniques. The closed injection impregnation technique uses resin better and makes less pollution than open-bath systems, however it is harder to use with vitrimer resins. Vitrimer viscosity is substantially affected by dynamic bond exchange processes, which makes it behave in a non-linear way with temperature. This is different from regular thermosets. For instance, vitrimer matrices that use transesterification chemistry usually have a sharp decrease in viscosity beyond 120–140 °C. On the other hand, disulfide- and imine-based systems soften at lower temperatures, between 80 and 110 °C. These changes make it harder to inject stable resin, moisten it, and soak up fibers. To deal with these problems, process changes can consist of (i) preheating the resin container to stabilize viscosity before injection, (ii) adjusting the catalyst concentration to find the right balance between reactivity and flow stability, (iii) adding reactive diluents or rheological additives to lower baseline viscosity, and (iv) using in-line viscosity monitoring with feedback-controlled pumps. Literature indicates that sustaining viscosity between the 0.5 and 2.0 Pa·s during impregnation is generally adequate to provide uniform fiber wetting in closed injection settings [33–36].

2.3. Curing Process and Die Shaping

The next step entails regulated curing and polymerization within the die by an exothermic reaction [64,65]. The molding temperature strongly depends on the resin system employed with thermosetting resins typically processed at lower curing temperatures and engineering thermoplastics requiring higher melting or softening ranges. Knowing the thermochemical behavior of the resin during pultrusion is crucial for optimizing performance, as described in detail by [36], which provides background on curing kinetics applicable to composite processing. At this point the polymer's molecular structure can be modified, which impacts the final qualities and performance of the composites. Various parameters affect the curing process, such as resin composition, temperature, pressure, dwell time, and the choice of catalysts and initiators [48]. Achieving an ideal balance among these factors is crucial to ensuring consistent curing and achieving superior mechanical performance of the composite material. The residence time of the reinforcing fibers and resin mixture spent inside the heated die during the pultrusion process determines the velocity, which requires careful control to synchronize the curing rate with overall process efficiency [66]. An optimal residence time ensures that the resin cures sufficiently to achieve the desired mechanical strength and dimensional stability while maintaining production efficiency. On the other hand, an improper balance can result in defects, such as incomplete curing that weakens the structure or excessive curing that leads to brittleness and diminished durability [67]. The selection and amount of catalysts and initiators, including organic peroxides such as methyl isobutyl ketone peroxide, tert-butyl perbenzoate, and di(4-tert-butylcyclohexyl) peroxide, can substantially influence the initiation and progression of the curing reaction [48].

Following the curing stage, the pultrusion method entails molding the reinforced fibers and resin into the desired cross-section within a custom designed die. Pultrusion dies often possess a short length, varying from 2 to 4 feet (0.6–1.2 m), making them suitable for cost-effective mass production [68]. The design and temperature management of the die are essential for shaping the pultruded product. In practice, the die is often designed such that the temperature is gradually increased at the entrance region, which promotes controlled curing and minimizes thermal stresses, thereby enhancing structural integrity [69]. Accurate temperature control is crucial for managing the curing and polymerization processes of the resin matrix. Fluctuating or incorrectly selected temperatures can result not only in premature curing, incomplete polymerization, uneven curing, and void formation, but also in practical problems such as product stickiness, pulling difficulties, and resin dripping [70]. Effective die heating systems are essential for ensuring consistent heat delivery throughout the process. Furthermore, the thermal expansion and contraction of die materials must be considered to reduce dimensional inconsistencies and prevent stress concentrations, enabling the manufacturing of high-quality, durable composite materials.

2.4. Pulling and Cutting of the Prepared Sample

The final stage of the pultrusion process involves extracting the cured composites from the die and cutting it to the desired length. Optimizing both the pulling rate and force and the cutting phases is essential for enhancing the efficiency and quality of fiber-reinforced polymer composites [71]. Increasing the pulling velocity reduces the time the materials remain in the preheating chamber. Pulling speed has been shown to affect the composite's mechanical characteristics, such as strength and stiffness [72]. However, maintaining proper fiber alignment and preventing distortions during extraction requires careful tension control. While higher pulling speeds might enhance manufacturing productivity; however, too high rates could affect resin impregnation and curing, potentially compromising the product quality including surface appearance. Conversely, slower pulling speeds allow for prolonged resin impregnation and curing time, reducing the production efficiency. Determining the right balance often requires trial-and-error iterations to achieve optimal pulling speed that supports both efficient processing and sufficient resin curing. The optimal pulling speed depends on the resin, fibers, and desired characteristics of the final product.

Once the pultruded material has been cured and aligned, precise cutting is necessary to ensure that the finished product meets the specified specifications. The cutting mechanism and speed are critical in ensuring accuracy, minimizing edge damage, and lowering dust generation throughout the cutting operation [73]. Synchronizing the pulling and cutting stages is critical for reducing material waste, maintaining product quality, and increasing overall process efficiency.

3. Factors Affecting the Quality of the Pultruded Product

3.1. Type of Resin Used and Their Viscosity

The viscosity of the resin used in pultrusion profoundly influences the quality and performance of the finished product [74]. Different resin systems exhibit unique properties that influence their suitability for various applications. Epoxy resins are renowned for their exceptional mechanical characteristics and robust fiber adhesion, resulting in durable composites. However, their limited pot life, due to fast reactivity, requires higher processing temperatures compared to polyesters [75]. Despite these advantages, epoxy resins often display a tendency to adhere to die walls potentially causing surface defects like exposed fibers and dimensional irregularities. In contrast, polyester is valued for its cost-effectiveness, easy processing, rapid curing, UV resistance, and versatility. However, they may undergo volumetric shrinkage up to 7% due to extensive crosslinking which affects the dimensional stability [76]. In the case of polyester matrices, styrene is commonly present as a reactive diluent, which facilitates processing by lowering viscosity; however, its volatility can lead to emissions that require careful control due to health and environmental concerns. Vinyl esters offer a compromise between epoxies and polyesters, providing commendable chemical resistance, mechanical strength, and fiber adhesion [75]. Meanwhile, Polyurethane (PU) resins are widely used in structural applications owing to their low viscosity, high reactivity, design, and potential cost savings of up to 20%. They produce strong, stiff, and lightweight components, suitable for both simple and complex profiles.

Effective viscosity regulation is crucial for optimizing fiber impregnation, as resin flow within the die directly impacts the uniformity of fiber wetting and composite integrity. As crosslinking progress due to viscosity increases, the risk of cavity development rises, which reduces the mechanical characteristics of the composite. A thorough understanding of the relationship between types of resin, viscosity, and rheology is essential to ensuring the production of high-quality pultruded composites. Recent research has delved into the rheological behavior of various resin systems under pultrusion conditions. Research performed by Yuksel et al. [77] and Baran et al. [78] studied the curing kinetics of polyurethane and polyester resins using differential scanning calorimetry (DSC), providing valuable insights into their performance during the pultrusion process. Rheological investigations using a rheometer have helped establish viscosity models dependent on temperature and curing progression, shedding light on changes in storage and loss moduli throughout the process. Baran et al. [78] performed a comprehensive three-dimensional characterization of the pultrusion techniques, combining thermochemical and mechanical components to assess temperature and curing profiles. Their findings highlighted the effects of thermal conditions on resin behavior, process-induced stresses, and deformations. Numerical simulations based on these studies have enhanced process control, contributing to improved product quality. In addition to resin properties, additives and fillers play a crucial role in modifying resin viscosity and enhancing composite performance. For effective stress transfer and composite performance, there must be a strong bond between the fiber surface and the polymer matrix [79–81]. Surfactants or coupling agents, such as silanes for glass fibers, titanates/zirconates for mineral fillers [82], or maleic anhydride–grafted polymers for polyolefin matrices, are frequently used to improve this. By improving wettability and interfacial bonding, these additives increase mechanical strength and durability.

3.2. Type of Fiber, Volume of Fiber, and Fiber Orientation

Glass fibers are widely used in pultrusion due to their versatility, cost-effectiveness, and high tensile strength, making them a popular choice for various industrial applications [83]. For applications requiring superior mechanical performance, carbon fibers offer enhanced strength, stiffness, and rigidity, making them ideal for high-performance aerospace, automotive, and structural applications. Aramid fibers, in contrast, are valued for their high specific strength and impact resistance relative to glass fibers, although they generally exhibit lower compressive properties and significantly lower thermal stability than carbon or glass fibers. Bio-based natural fibers such as kenaf, hemp, and coir are being explored as sustainable reinforcement options for pultrusion technique [84,85]. These fibers provide environmental and cost benefits while also helping to build more eco-friendly composite materials. The mechanical property of the pultruded composite depends upon the fiber volume fraction which ranges from 50% to 70% [86–89].

In addition to fiber type and volume, the architecture and orientation of reinforcing fibers significantly influence the directional characteristics and overall performance of composites. Fiber alignment can be adjusted to provide specific mechanical properties based on application requirements. Unidirectional fibers are often used to maximize strength and stiffness along a single axis, making them ideal for load-bearing applications. Pultrusion predominantly employs continuous unidirectional fibers or non-crimp fabrics that maintain alignment under tension; woven fabrics are more common in roll-wrapped applications, while chopped strand mats are generally not suitable for pultrusion due to the lack of continuous fiber tension.

Textile fiber architectures, such as braided structures, promote conformability and are particularly useful for complex geometries, while knitted fabrics provide flexibility and resistance to delamination, making them well-suited for dynamic applications. Additionally, random fiber mats (chopped strand mats) exhibit isotropic properties, delivering uniform strength distribution in all directions, though with lower mechanical performance compared to aligned architectures. Three-dimensional woven and stitched fibers enhance through-thickness reinforcement, reducing delamination and increasing impact resistance. Optimizing fiber architecture and orientation is essential for modifying the composite's mechanical, thermal, and structural properties. By selecting the appropriate fiber orientation (e.g., unidirectional, cross-ply, or multidirectional), engineers can tailor the anisotropic behavior of pultruded composites, thereby achieving specific performance outcomes such as higher tensile strength in the fiber direction, improved fatigue resistance under cyclic loading, or enhanced dimensional stability.

3.3. Die Geometry and Its Temperature

The die plays a crucial role in shaping and forming pultruded components, as it establishes the profile of the final product and dimensions of the composites [90]. An effective die design must ensure proper fiber alignment during the pultrusion process, which is critical for achieving the desired mechanical properties and structural integrity of the composite [91]. Various factors, including die material, size, cavity dimensions, and geometry, influence resin flow, fiber orientation, and, ultimately, the overall quality of the pultruded product [92]. A well-designed die ensures proper resin distribution and consistent fiber alignment, which are essential for avoiding defects such as voids, fiber misalignment, or delamination [93,94]. Controlling tension within the die is vital to prevent defects, as excessive stress concentrations can lead to fiber distortion or material inconsistencies [90]. The die must facilitate a controlled deformation of the composite, ensuring uniform thickness and proper structural formation. Temperature control within the die is another critical factor influencing resin curing and the final composite properties [95]. While most thermosetting pultrusion dies lack dedicated cooling zones, some advanced configurations include both heating and cooling elements near the die entrance to regulate the curing process and maintain consistent pultrusion speeds [96]. Maintaining the correct temperature profile is crucial, as excessively high temperatures can cause premature curing, leading to processing challenges, while insufficient temperatures may result in incomplete polymerization. Ideally, the cooling zone should sustain a temperature above the resin's gelation point but below the full cure temperature to ensure optimal curing. For specialized applications, curved dies allow for the continuous production of components with intricate contours and consistent cross-sections, allowing specific functional or aesthetic requirements [97–99]. The precision in designing curved dies, particularly the accurate control of heating and cooling, substantially influences the dimensional accuracy and overall quality of pultruded components.

4. Pultrusion of Curved Components

Pultrusion is traditionally employed to produce straight, elongated composite. But, when fibers are bent, twisted, or crimped within a cured matrix, they can lose some of their advantageous mechanical properties, compromising the structural integrity and desired functionality of the composite [100,101]. Although standard pultrusion techniques are ideal for creating straight profiles, recent advancements have enabled the production of curved parts for various applications [102], as illustrated in Figure 2.

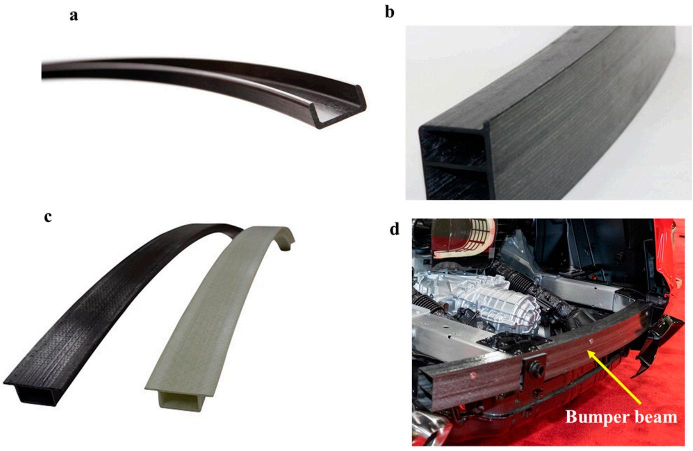

Figure 2. Examples of curved pultruded parts: (**a**) elegant arcs, (**b**) curved hollow part, (**c**) curved CFRP bumper, and (**d**) rear bumper beam (highlighted with arrow) of a Chevrolet Corvette sports car's curve pultruded part [102].

Curved pultrusion overcomes the challenges associated with high deflection and bending, which often limit the performance of pultruded glass fiber-reinforced polymer (GFRP) beams [103]. Research shows that failure in pultruded GFRP components is often governed by bending-induced stresses and deflection, rather than by exceeding the intrinsic tensile or compressive strength of the material [103–106]. While standard straight pultrusion effectively produces linear profiles, it does not meet the demand for cambered or arched beams, which are often necessary to minimize deflection in structural applications [103]. Moreover, research on curved pultrusion using thermoplastic materials remains limited compared to thermosetting resins. To address these limitations, various advanced manufacturing processes have been developed for producing curved pultruded components, each offering distinct advantages and challenges. One such method, introduced by Talabi et al. [96], is known as "bent pultrusion," "3D pultrusion," or "post-die manipulation" (Figure 3). This process involves pulling resin-saturated rovings through a die using a robotic arm at room temperature. The die shapes the material into the desired cross-section and removes surplus resin, while the uncured resin serves as a lubricant to minimize friction through fiber movement.

After exiting the die, UV light cures the resin allowing for precise manipulation of the fiber placement during curing [107]. This technique limits the die's function to shaping the profile, significantly reducing pulling pressures and enabling the formation of radii and corners. However, this approach is primarily suitable for simple curves and lacks the versatility required for more complex geometries. An alternative technique, molded pultrusion, integrates a curved mold or die into the production process, allowing for the continuous formation of intricate shapes [107] (Figure 4a). Figure 4b illustrates how the die heaters are positioned along different angular sections (6°, 17°) to gradually guide the composite into the desired curved profile without inducing excessive stresses or defects.

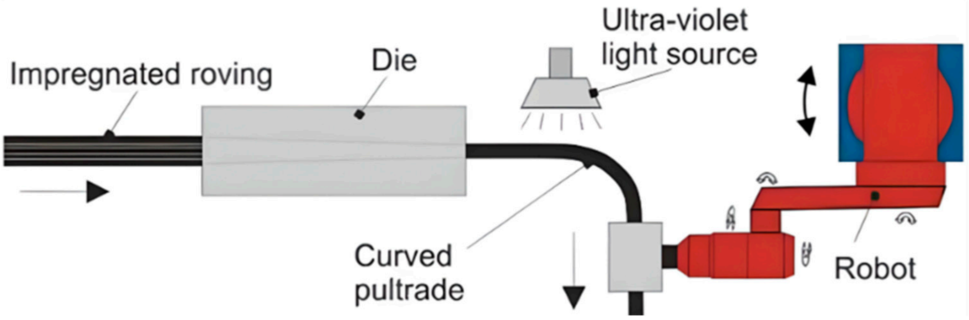

Figure 3. Schematic representation of curved pultrusion [96].

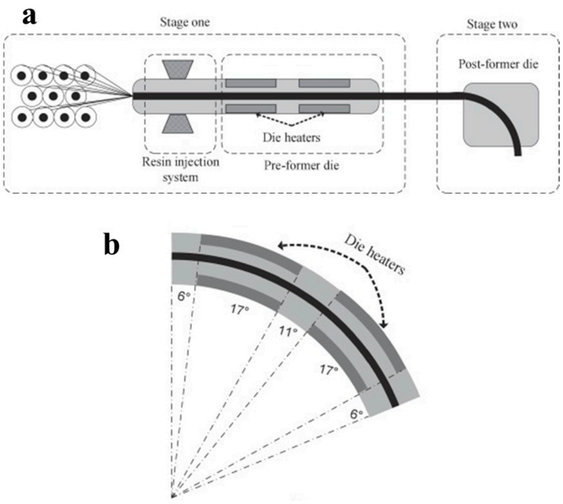

Figure 4. Schematic representation of emphasizing curved die in curved pultrusion: (**a**) schematic of pultrusion and (**b**) cross-section of the T-stiffener [107].

While effective for producing complex profiles, molded pultrusion involves higher tooling and mold costs, making it a more expensive option compared to conventional pultrusion methods [108]. The complexity of mold design also imposes limitations on the range of geometries that can be economically manufactured. A notable innovation in this field is TTI's patented moving mold system, which enables the creation of both straight and continuous curved profiles. The dynamic die moves back and forth along the profile, allowing the formation of constant 2D or 3D radii, alternating straight and curved sections, and even coil spring-like features [85]. The process uses heated chrome-plated dies to reduce the risk of fiber withdrawal from the hot matrix. However, this technology is currently restricted to producing profiles with consistent cross-section.

Deflection rollers offer another tool for making intricate curves and forms in pultruded components by applying controlled pressure along the pultrusion line, guiding the material as it cures [109]. The curved-pultrusion approach has been successfully employed to manufacture arched and cambered profiles for structure applications [110]. Further advancements include a patented pultrusion process by Klimovski et al. [111] for manufacturing curved gap filler preforms compatible with curved stringers in aerospace structures. This method utilizes a pultrusion die with a curved path, a cooling chamber, and heat-transfer channels to enhance the precision and speed of production. The process involves heating the fiber-reinforced material to its adhesion temperature, feeding it into the curved die, and continuously forming it into the desired preform shape while maintaining dimensional accuracy. Another significant innovation focuses on the production of curved pultruded beams for automotive applications [112,113]. This patent outlines a method for manufacturing vehicle bumper systems with polymeric impact beams, incorporating at least 30% continuous fiber reinforcement for improved structural integrity and impact resistance. The technique allows for the creation of curved beams with reinforced corners and attachment features, using a combination of tows, rovings, and fiber fabrics. Recent studies improved the pultrusion of curved composites via innovative die and mold designs. For instance, new segmented and adaptive mold geometries have been created to lower residual stresses and fiber misalignment during curved pultrusion [96]. Real-time process monitoring and online sensing technologies, such as fiber Bragg grating sensors and infrared thermography, have been used in parallel to keep an eye on the evolution of the cure, the infiltration of the resin, and the formation of defects during pultrusion runs [110]. These monitoring tools allow for closed-loop control of pull speed and die temperature, which makes it easier to repeat and scale up pultrusion. Using these high-tech mold designs and digital process control methods is an important step toward encouraging industries to use vitrimer composites for structural parts with complex shapes that last a long time.

Overall, these innovations in curved pultrusion enable the fabrication of complex geometries with enhanced mechanical properties, expanding the potential applications of pultruded composites in industries such as automotive, aerospace, and civil engineering. As technology progresses, the focus remains on overcoming current limitations, such as cross-sectional variability and tooling costs, to improve manufacturing efficiency and broaden the application range of curved pultruded components.

5. Properties of Pultruded Composites

Analyzing the properties of pultruded composites is essential for optimizing structural design, ensuring safety, and maximizing material efficiency. Due to their anisotropic nature, their stiffness and strength depend on load direction, making it crucial to consider these factors in applications involving complex loading conditions, such as earthquakes or wind forces. Understanding these characteristics improves design standards, enhances failure mode predictions, and ensures structural reliability. Additionally, it drives material innovation, enabling the development of stronger, more durable, and cost-effective composites tailored to specific applications.

Pirchio et al. [113] investigated the tensile strength and modulus of pultruded fiber-reinforced polymer (FRP) composites under varying load angles relative to the pultrusion direction. These properties are crucial for structural design, as they vary with load orientation due to the anisotropic nature of FRP composites. A generalized Hankinson's equation was developed to predict these variations, showing good accuracy with deviations ranging from 4.4% to 8.3%. The study revealed a sharp decline in tensile strength from 0° to 45°, which then plateaued, indicating reduced sensitivity at higher angles. This anisotropic trend, consistent with classical laminate theory, is illustrated in Figure 5. It should be noted that the original study employed dog bone specimens, which are not recommended for composite tensile testing, and therefore the schematic is provided here only to represent the general behavior rather than specific test data.

Landesmann et al. [114] conducted an experimental investigation on pultruded glass fiber-reinforced polymer (GFRP) elements, reporting an average tensile strength of 253.3 MPa and a compressive strength of 262.3 MPa. The composites also exceeded E23 class standards for flexural and pin-bearing strengths. The modulus of elasticity averaged 24.5 GPa, and the interlaminar shear strength was 20.6 MPa, both meeting E17 class standards. As shown in Figure 6, different failure modes were observed during testing. Higher fiber content improved mechanical performance, with flange samples showing greater strength than web samples due to better fiber alignment. These findings confirm the suitability of E17-class GFRP elements for structural applications requiring high strength and stiffness.

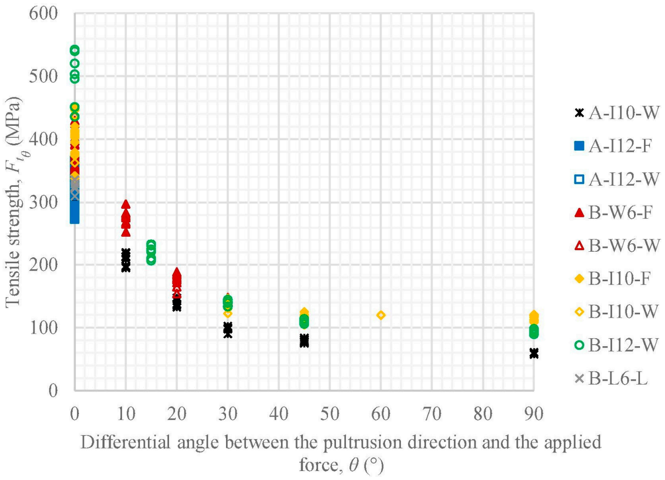

Figure 5. Tensile strength variation in pultruded FRP composites with load angle relative to fiber orientation [113].

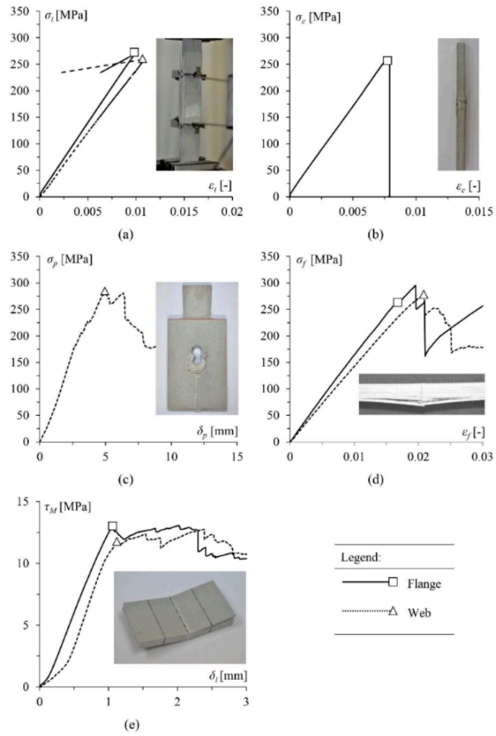

Figure 6. Pultruded composite samples and their failure mode: (**a**) tensile, (**b**) compression, (**c**) pin-bearing pushed-out, (**d**) flexural, and (**e**) interlaminar shear strength [114].

Hashemi et al. [115] investigated pultruded hybrid kenaf/glass fiber composites and found that increasing the proportion of kenaf fibers led to a reduction in interlaminar shear strength (ILSS). The highest ILSS was recorded in non-hybrid glass fiber composites (21.5 MPa), while kenaf-rich composites exhibited the lowest values (10–12 MPa). This was due to the higher kenaf content resulting in increased impregnation issues and interface voids, weakening the fiber–matrix bonding. Irfan et al. [116] explored the characteristics of glass fiber vinyl ester polymer composites produced using a modified pultrusion process.

The experiments compared composites made with filled and unfilled vinyl ester resins combined with glass fibers. The results, summarized in Figure 7, highlighted key differences in mechanical behavior. The study found that unfilled resin composites exhibited lower flexural strength, likely due to de-bonding at the fiber–matrix interface. While interlaminar shear strength (ILSS) values were similar between the two pultrusion techniques, composites with filled resin demonstrated higher ILSS. Additionally, the storage modulus remained consistent across both pultrusion methods; however, composites made with unfilled resin showed a higher overall modulus.

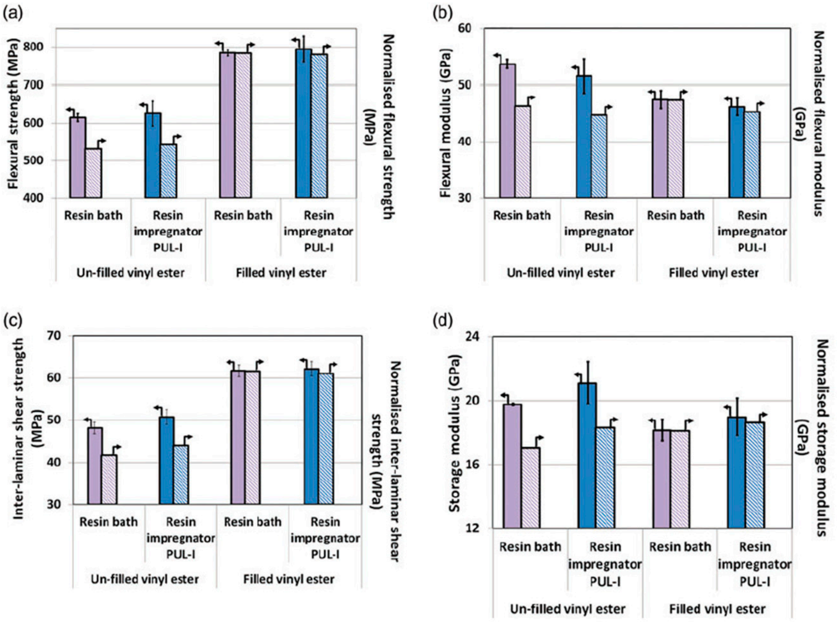

Figure 7. Mechanical and dynamic mechanical analysis of the pultruded composite: (**a**) flexural strength, (**b**) flexural modulus, (**c**) ILSS, and (**d**) storage modulus [116].

Balakrishnan et al. [117] studied the mechanical performance of kenaf/glass fiber hybrid pultruded composites. Compared to pure kenaf or glass fiber composites, the hybrids exhibited superior fatigue resistance, making them suitable for cyclic loading applications. Factors such as the kenaf-to-glass fiber ratio, loading conditions, and fiber orientation influence fatigue life. Natural fibers (kenaf) enhance sustainability and reduce costs, while synthetic fibers (glass) improve strength and durability. Luo et al. [118] investigated the transverse tensile strength of pultruded glass fiber composites, comparing non-standard and standard coupons. As shown in Figure 8, the non-standard coupons exhibited failure modes like standard specimens. The results indicated that transverse tensile strength is lower than longitudinal strength due to the resin matrix's dominance in the transverse direction. Common failure mechanisms included fiber–matrix debonding and matrix cracking, with non-standard coupons proving to be a reliable alternative for testing.

Liu et al. [119] established a process for producing curved-pultruded GFRP arch beams. The beams underwent three-point bending tests using strain gauges and linear variable differential transducers (LVDT) for monitoring. The tensile strength and modulus of the flange were found to be 768 MPa and 49.2 GPa, respectively. The web plate exhibited a transverse strength of 61.6 MPa and a modulus of 12.6 GPa, with in-plane shear strength and modulus recorded at 71.3 MPa and 3.4 GPa, respectively. The study demonstrated that curved pultrusion offers enhanced fiber dispersion and flexural properties compared to traditional methods like filament winding. Struzzieroa et al. [120] focused on modeling curved carbon/epoxy composite components for aerospace applications. The study analyzed aerospace epoxy resin, developed models, and created a 3D Finite Element (FE) model for manufacturing simulation. The results indicated that a 62% degree of cure in the pre-former stage is optimal for achieving an 87% cure post-forming. The method proved effective, with maximum stresses of 54 MPa in compression and 200 MPa in tension, confirming the feasibility of defect-free manufacturing.

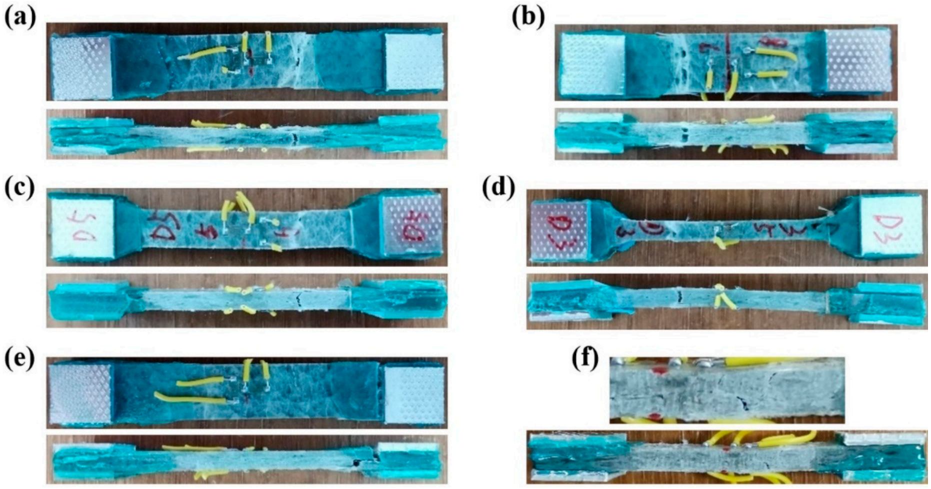

Figure 8. Failure modes of transverse short coupons: (**a**) fracture of rectangular long coupons; (**b**) fracture of rectangular short coupons; (**c**) fracture of dog-bone-shaped wide coupons; (**d**) fracture of dog-bone-shaped narrow coupons; (**e**) end failure; and (**f**) premature failure [118].

6. Vitrimers: A New Class of Polymer

Thermosetting polymers are characterized by covalently crosslinked three-dimensional (3D) networks that form during the curing process [121,122]. Unlike thermoplastic polymers, these covalent bonds in thermosets are typically irreversible, even at elevated temperatures, leading to decomposition rather than melting. As a result, thermosets are traditionally considered non-recyclable, whereas thermoplastics can be readily remolded and recycled into new products. In some cases, thermoset resins have also been used as additives or fuel components [123]. Recent research has increasingly focused on developing polymers that combine the durability of thermosets with the processability of thermoplastics [124]. This has led to the creation of covalent adaptable networks (CANs), modified thermosetting materials that can respond to external stimuli, such as light, heat, electricity, pH, and moisture, allowing for reversible bond reconfiguration. Polymers incorporating these networks have been classified as vitrimers.

Vitrimers exhibit a unique viscosity-temperature relationship that follows Arrhenius' law, like vitreous silica [125]. They behave like thermosets under normal conditions, offering high mechanical strength and stability. However, when exposed to external stimuli, vitrimers demonstrate thermoplastic-like behavior due to their dynamic crosslinked structure, allowing bond dissociation and reformation without changing the overall crosslink density. One of the key features of vitrimers is their ability to undergo a solid–liquid transition facilitated by a prolonged rubbery phase, enabling reshaping and reprocessing while maintaining structural integrity. Additionally, their dynamic covalent bonds provide excellent solvent resistance, making them suitable for high-performance applications [126]. Their rheological characteristics are controlled by bond-exchange reactions that occur by either associative mechanisms (where a new bond forms before the old one breaks, as in transesterification and transamination) or dissociative mechanisms (where the old bond breaks first, as in imine or disulfide exchange) [124]. When associative CANs undergo exchange, the crosslink density remains constant. In contrast, during the exchange of dissociative CANs, the crosslinking is temporarily reduced, which affects relaxation and reprocessability. Their processing and reprocessing mechanisms rely on various addition-elimination reactions, making their structural and functional characteristics difficult to fully comprehend [127]. Figure 9 illustrates the potential of vitrimer materials, highlighting their dynamic covalent bond networks within their structure.

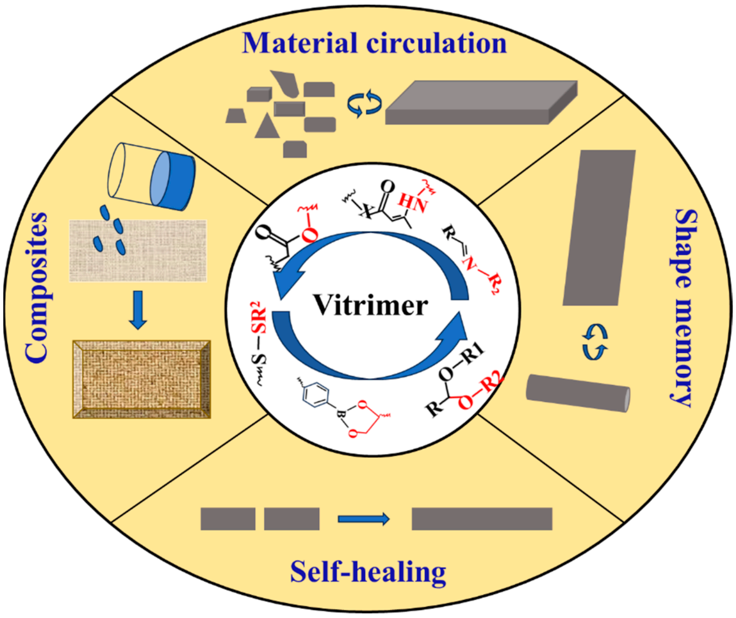

Figure 9. Schematic representation of dynamic covalent bonds in vitrimer materials.

Vitrimer materials offer significant advantages across various material characteristics, properties, and applications. These adaptable polymers have been extensively explored for their potential in developing composite and nanocomposite, enabling the creation of self-healing and self-morphing structures, and improving material processing techniques. These dynamic networks allow reversible bond exchanges in response to external stimuli, such as heat, light, or pH changes, facilitating reshaping, repair, and enhanced durability [128]. Figure 10 illustrates the relationship between the covalent adaptable networks in vitrimer materials and their response to reversible stimuli, highlighting how these dynamic bonds contribute to the material's unique adaptability and functionality.

When vitrimer are exposed to external stimuli such as heat, light [129], or chemical triggers, these networks undergo bond exchange reactions. Due to these reversible bond-exchange mechanisms, vitrimers can be considered recyclable thermosetting materials, offering a sustainable alternative to conventional thermosets. Figure 11 illustrates the fundamental mechanics of vitrimer recycling, including processes such as hot pressing and solvent-assisted reprocessing, which facilitate reshaping and reuse without compromising structural integrity.

In solvent solutions, vitrimers can be depolymerized into monomers or oligomers. Once the solvent evaporates, these materials can be reprocessed or repolymerized, effectively restoring the vitrimer's original structure and properties. This ability to maintain their persistent crosslinked network after multiple processing cycles enhances their longevity and usability. Owing to their unique reprocessability and structural resilience, vitrimers and their composite and nanocomposite derivatives are being applied across various fields. These applications include shape memory materials, self-healing polymers, adhesives, advanced printing processes, and other emerging sectors yet to be fully explored.

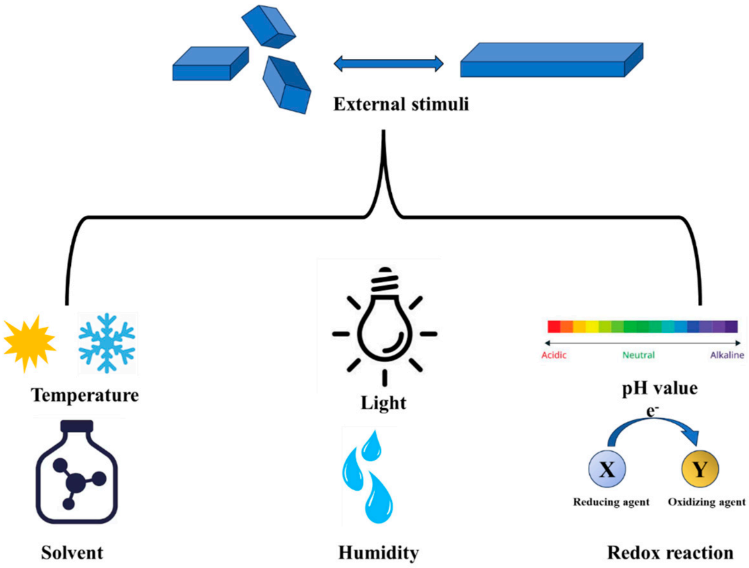

Figure 10. Schematic representation of external agent to activate the reversible bond.

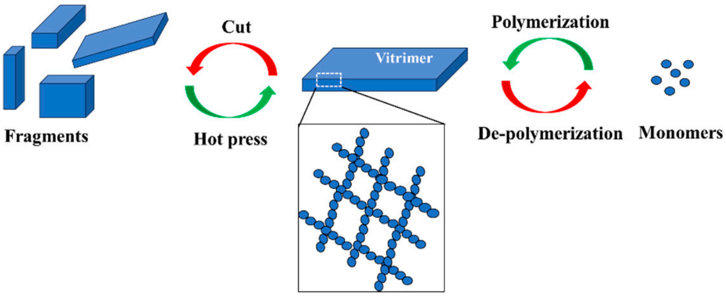

Figure 11. Schematic representation of vitrimer recycling.

6.1. Vitrimer Chemistry and Its Key Properties

Vitrimer-based polymers are synthesized through various chemical reactions and covalent bond exchange mechanisms, which enable their dynamic and reversible nature. Research has shown that the presence of a catalyst can significantly influences the exchange reaction pathway, often enhancing reaction rates and facilitating faster network rearrangement. In contrast, several studies have explored catalyst-free systems, revealing distinct material properties and processing behaviors compared to catalyst-assisted systems. Figure 12 illustrates the chemical pathways involved in vitrimer formation, highlighting both catalyst-assisted and catalyst-free processes used in developing associative dynamic covalent adaptive networks (ADCANs) within thermoset polymers. These networks enable unique features such as self-healing, reprocessability, and stress relaxation under specific conditions. Table 1 provides a comparison of epoxy vitrimer materials synthesized with and without a catalyst, detailing their mechanical strength, glass transition temperature (Tg), healing conditions, and healing efficiency. This comparison highlights how catalyst incorporation can influence vitrimer performance, providing valuable insights for designing materials tailored for specific applications.

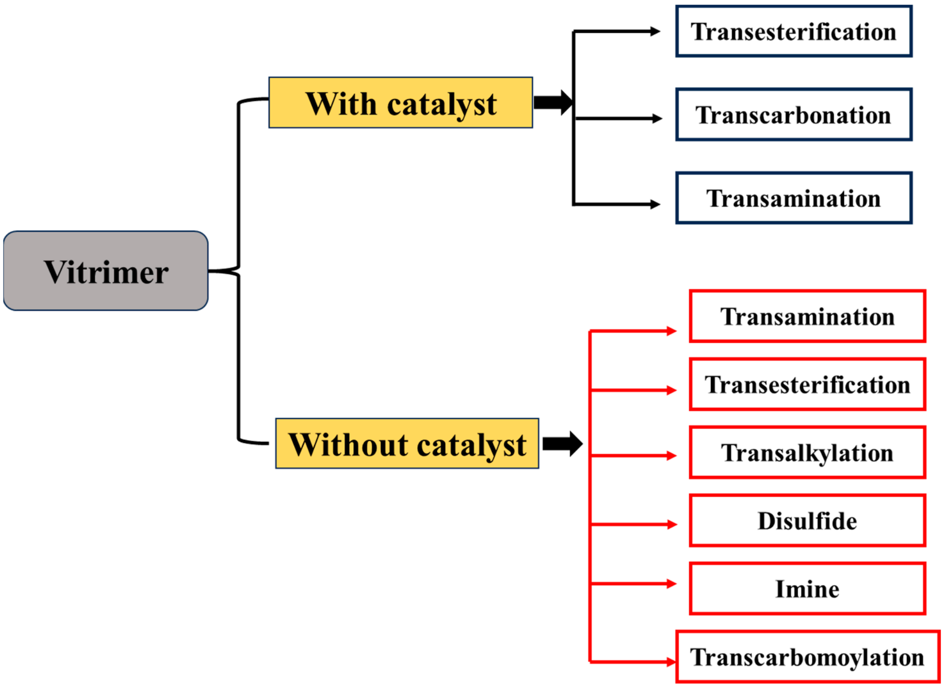

Figure 12. Chain exchange chemical reaction of vitrimeric material with and without catalyst.

Table 1. Properties of vitrimer material in presence and absence of catalyst [130].

Types of Mechanism	Types of Materials	Types of Catalyst	Transition Temperature (Tg)	Healing Conditions	Healing Efficiency	Mechanical Strength (MPa)	References
Transesterification	Biobased triepoxy	$[Zn (Ac)_2]$	187 °C	220 °C, slight pressure	90%	62.8–69.2	[128]
Transesterification	Epoxy	Tributylamine (TBA)	130 °C	160 °C, 3 h	57%	20	[129]
Transesterification	Liquid crystalline vitrimer with amino-capped trimer	Triazobicyclodecene (TBD)	58 °C	IR laser	100%	41.4	[131]
Transesterification	Polydopamine-modified gold nanosheet epoxy vitrimer	Triazobicyclodecene (TBD)	42 °C	Light, 120 min	90–100%	90	[132]
Transamination	Poly(vinylogous urethane)	Catalyst free	87 °C	150 °C, 30 min	76%	25	[133]
Transalkylation	Poly(1,2,3-triazolium ionic liquid)	Catalyst free	−8, −23 °C	-	-	15	[134]
Transalkylation	Poly(thioether)	Catalyst free	−20 °C	160 °C, 45 min	100%	80	[135]
Transcarbamoylation	Polyhydroxy urethane	Catalyst free	54 °C	160 °C, 4 MPa for 8 h	76%	2.2 ± 0.4	[136]
Disulfide	Epoxy with silica nanoparticles	Catalyst free	151–155 °C	180 °C, 30–120 min	36.8–78.1%	58.5 ± 1.3	[137]
Disulfide	Polyhydroxy urethane	Catalyst free	66 °C	150 °C, 510 MPa for 30 min	65%	35	[138]
Disulfide	Epoxy with FRP	Catalyst free	127 °C	200 °C,100 bar for 5 min	-	43 ± 7	[139]
Imine	Polyimine	Catalyst free	102 °C	110 °C for 30 min	-	Dry and wet samples, 49 and 32	[140]

Note: The "Mechanical Strength" values represent the specific property reported in each cited study (e.g., tensile, flexural, or compressive strength), as indicated in the respective references.

6.2. Key Properties of Vitrimer

Vitrimers are primarily defined by their viscoelastic characteristics, which vary with temperature and are closely linked to the rate of covalent bond exchange, corresponding to the material's transition temperature. When heated beyond the glass transition temperature (Tg), thermoplastic materials and dissociative covalent adaptable networks (CANs) typically experience changes in their molecular topology, following the Williams–Landel–

Ferry (WLF) behavior on a macroscopic scale. In contrast, vitrimers incorporate dynamic covalent bonds that exhibit temperature-dependent behavior, following an Arrhenius-type relationship [130]. This dynamic bond exchange allows for controlled viscosity, enabling processing over a broad temperature range while maintaining the material's crosslinked network integrity. Unlike thermoplastics and dissociative CANs, which show a significant reduction in viscosity and crosslink density with increasing temperature, vitrimers retain structural stability during thermal processing [130]. A key feature distinguishing vitrimers from traditional polymers is the introduction of the topological freezing transition temperature (Tv), which represents the point at which the material transitions from a viscoelastic solid to a liquid state. This transition occurs when the viscosity approaches 10^{12} Pa·s [141]. Tv sits between the solid and liquid phase transitions and is critical for understanding vitrimer flow behavior. The rate of covalent exchange in vitrimers varies with temperature: slow exchange reactions dominate below Tv, while faster exchanges occur above Tv. Research suggests that topological rearrangement begins around Tg and accelerates as the material approaches Tv. The position of Tv relative to Tg significantly influences vitrimer behavior and can be categorized into two pathways:

1. Tv Above Tg: The material behaves in a rubbery or viscoelastic state between Tg and Tv. As the temperature reaches Tv, it transitions into a viscoelastic liquid, following Arrhenius behavior.
2. Tv Below Tg: The polymer chains remain rigid until reaching Tg, after which bond exchange initiates. Initially, the material exhibits WLF behavior for a specific period before shifting to Arrhenius behavior as the temperature increases.

This temperature-dependent behavior is illustrated in Figure 13 [141], highlighting how vitrimer properties evolve through different phases depending on the relationship between Tv and Tg.

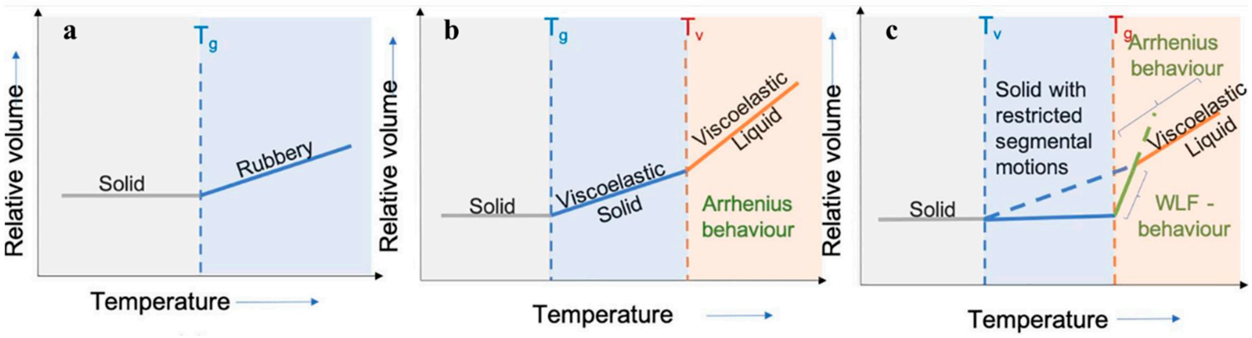

Figure 13. (**a**) Phase transition on thermoplastic and dissociative covalent material thermosetting vitrimer following (**b**) Tg below Tv and (**c**) Tv above Tg [141].

6.3. Mechanism

Figure 14 illustrates a novel approach developed for dynamic covalent adaptive networks (DCANs) in thermoset materials, designed to enhance their self-repair capabilities through both dissociative and associative bond exchange mechanisms [142–144]. In dissociative covalent adaptable networks (CANs), dynamic bonds can break and reform even within highly crosslinked systems like thermosets. This results in a temporary reduction in crosslink density as bonds dissociate and then re-form, allowing network rearrangement and facilitating material repair. As shown in Figure 14a, bond breakage leads to a proportional drop in crosslink density, weakening system connectivity during the process.

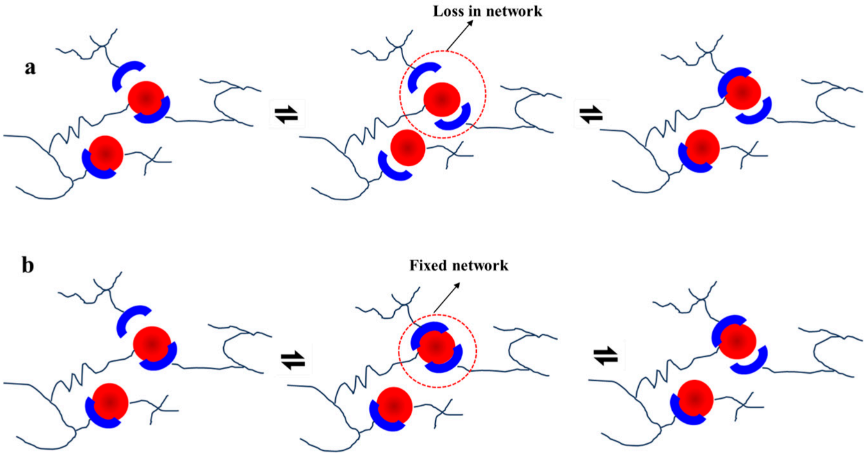

Figure 14. Schematic representation of CAN: (**a**) dissociative and (**b**) associative.

A significant example of this mechanism was demonstrated by Wudl and coworkers [145], who developed a recyclable, self-healing thermosetting polymer based on the thermoreversible Diels–Alder reaction. This reversible reaction allows the material to self-heal and regains its original mechanical properties. Other studies have explored similar chemistries for healing thermosetting polymers, showing that when crosslink density decreases nonlinearly with rising temperature, material flow changes significantly but can be reversed upon cooling, restoring the material's strength [146]. In contrast, the associative dynamic covalent adaptive network (ADCAN) maintains a stable crosslink density during the bond exchange process. As illustrated in Figure 14b, bond breaking and reformation occur simultaneously, allowing the network structure to rearrange without reducing connectivity [144,145]. This mechanism results in minimal changes to the macromolecular structure during the exchange, preserving the material's integrity.

The first ADCAN was observed in 2005, based on a reversible addition-fragmentation chain transfer (RAFT) reaction in thiol-ene monomer units. A significant advancement came in 2011 when Leibler and colleagues [146] introduced an ADCAN-based vitrimer using a transesterification reaction within an epoxy system. This innovation led to the development of materials that show characteristics like vitreous silica, with properties controllable across two temperature ranges defined by the glass transition temperature (Tg) and the Tv.

Vitrimers have been the focus of extensive research since Leibler's work, leading to the advancement of various chemical platforms tailored for dynamic covalent networks. While contemporary reviews and perspective articles provide detailed insights into vitrimer chemistry, only a limited range of chemistries has been adopted in the composite industry due to the restricted availability of dynamic chemical precursors at an industrial scale [147,148]. For vitrimer-based composites to be viable in manufacturing, the selected dynamic chemistry must be compatible with the overall crosslinking process and should not release volatile byproducts, such as water, which could introduce defects during curing. Additionally, the viscosity and other processing parameters must align with industrial manufacturing techniques such as resin transfer molding (RTM), vacuum infusion, and impregnation. These processes require materials capable of withstanding high temperatures, both during structural composite manufacturing and in real-world applications. To ensure proper vitrimer processing, the composite's production temperature must exceed the material's glass transition temperature (Tg). Furthermore, the reprocessing temperature (Tr) and degradation temperature (Td) of the polymer matrix must be carefully considered to establish suitable conditions for reshaping or repairing the vitrimer. Typically, Tr is set approximately 50 °C above Tg to allow sufficient segmental motion of polymer chains, facilitating dynamic bond exchange. However, for vitrimer matrices with a high Tg, the range of possible reprocessing temperatures becomes more limited. This necessitates a balance between avoiding degradation reactions and maintaining enough polymer flexibility for effective reshaping. These thermal and chemical constraints have restricted the formulation of vitrimer composites to a narrow range of chemical substrates that meet both processing and performance requirements [149].

6.4. Bond Exchange Mechanism

Over the past decade, a diverse range of vitrimers has been developed, each featuring distinct bond exchange mechanisms. Notable examples include transesterification, disulfide exchange, and imine exchange mechanisms, each contributing unique properties and functionalities to the material [150–152]. These mechanisms, illustrated in Figure 15, enable dynamic covalent bond rearrangement, enhancing the material's reprocessability, self-healing capabilities, and overall mechanical performance. The effectiveness of vitrimer pultrusion relies on the selected bond exchange mechanism, which influences activation energy, the suitable range of temperature, and process compatibility.

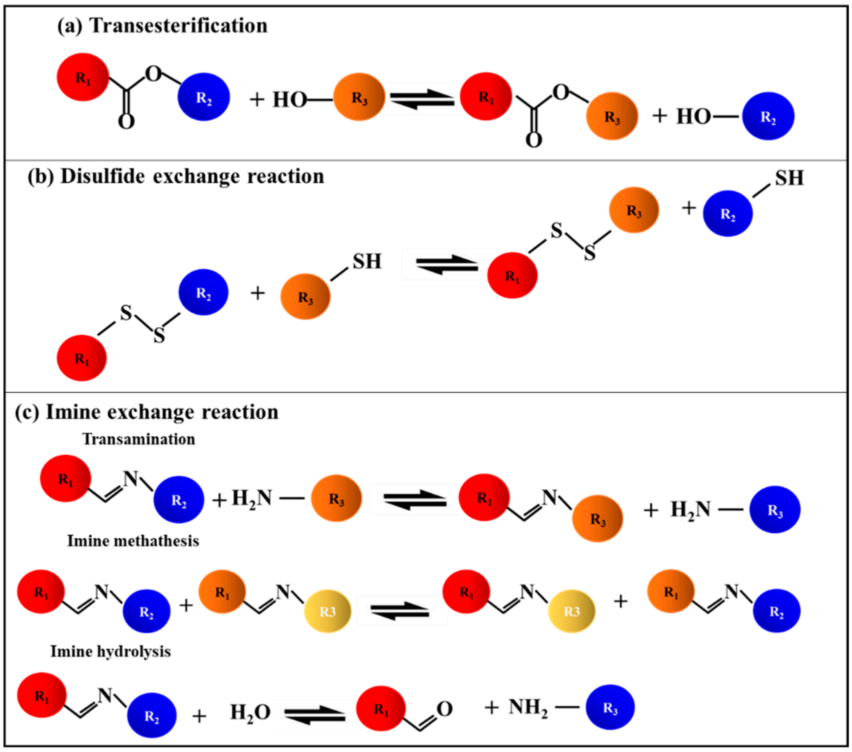

Figure 15. Bond exchange mechanism: (**a**) transesterification, (**b**) disulfide exchange reaction, and (**c**) imine exchange reaction.

6.4.1. Transesterification

Transesterification linkages in epoxy-acid and epoxy-anhydride systems typically require a Lewis acid or base catalyst, often using metallic or organic compounds. However, these catalysts can pose risks, including toxicity, resin incompatibility, and degradation during thermal recycling. Recent research focuses on biobased epoxy vitrimers that eliminate the need for external catalysts, using self-catalytic tertiary amines within monomers to accelerate bond exchange. Increasing hydroxyl and ester group content, or adding hyperbranched polyols, can further enhance reaction rates, promoting faster self-healing and stress relaxation. However, these changes may reduce the glass transition temperature (Tg) and affect mechanical performance. Transesterification processes usually have activation energies of about 80–120 kJ·mol^{-1}, therefore they need to be carried out at temperatures over 150 °C. This makes it more durable at high temperatures, but it also uses more energy.

6.4.2. Disulfide Bond

Disulfide bonds are weak, reversible covalent bonds capable of dynamic exchange, even at room temperature, without the need for a catalyst. Aromatic disulfide bonds generally exhibit higher exchange reactivity compared to other disulfide types. Incorporating these bonds with petroleum-based hardeners has enabled the development of various epoxy vitrimers featuring disulfide linkages, enhancing their self-healing and recyclability properties. Disulfide exchange processes have lower activation energies of about 50–70 kJ·mol^{-1}, which allows for reconfiguration at 90–120 °C. This is more energy-efficient, but it might make oxidative stability worse.

6.4.3. Imine Bond

Aldehydes and primary amines form imine bonds (Schiff bases) through condensation reactions without a catalyst. These bonds can undergo metathesis or react with primary amines to form new imines. With a lower activation energy than transesterification bonds, imines enable faster stress relaxation and milder reprocessing conditions, even without catalysts. Aromatic imines are more reactive than aliphatic ones due to the conjugation effect. While stable in neutral and alkaline conditions, imine bonds readily hydrolyze in acidic environments, especially with organic solvents. Imine exchange is in the middle range (around 60–90 kJ·mol^{-1}) and works best between 110 and 140 °C, which is a good balance between reactivity and stability.

From a pultrusion point of view, these ranges suggest that transesterification-based systems are better for aerospace or high-performance uses that need a high Tg, while disulfide and imine systems may be better for automotive and construction uses that need lower processing temperatures and energy savings. Case studies demonstrate that modifying the kind of catalyst (e.g., Zn^{2+} salts for transesterification, tertiary amines for imine exchange) and its concentration facilitates the synchronization of bond-exchange kinetics with the residence times in the pultrusion die (30–90 s). This comparative framework elucidates the selection and optimization of vitrimer chemistry for various pultrusion scenarios [31,33].

6.5. Vitrimer Composites: Key Characteristics and Properties

Pure polymers, including vitrimers, often fall short of meeting the increasing performance demands of modern industries. To overcome these limitations, incorporating reinforcements and fillers into vitrimer matrices has become essential. These composites enhance properties such as mechanical strength, electrical and thermal conductivity, and responsiveness to external stimuli. The performance of vitrimer composites depends on several factors, including the type of reinforcement, aspect ratio, fiber loading, degree of dispersion, and the quality of polymer-reinforcement bonding. However, in most epoxy vitrimer composites, there is often a trade-off between enhanced properties and slower topological rearrangement due to restricted polymer chain mobility caused by the reinforcement. While low filler loading may be insufficient to improve desired properties, excessive loading can hinder the dynamic network's ability to rearrange effectively. Recent research has focused on developing fiber-reinforced polymer (FRP) composites that are reprocessable, repairable, and weldable using vitrimer matrices. A notable advancement is the development of VITRIMAX™ by Mallinda Inc., a polyimine-based vitrimer resin capable of forming robust, dynamic, crosslinked thermoset networks that can be reshaped, molded, and welded. These resins support closed-loop recycling in both solution-based and solid-state environments [152]. Most notably, this polyimine resin has been employed as a matrix for carbon-fiber FRP composites, enabling a fully closed-loop system that allows the efficient recovery and reuse of both the fiber and resin [36]. Malleable polyimine demonstrates a tensile strength of approximately 40 MPa and exhibits significant mechanical property recovery after reprocessing. A single-ply FRP composite reinforced with woven carbon fiber displays a tensile strength of approximately 400 MPa and a flexural strength of around 150 MPa. These composites can be molded into various 3D curved configurations through a straightforward heat-pressing process and demonstrate excellent recovery of mechanical properties after reprocessing [36]. Imine-cured DGEBA vitrimer and trifunctional epoxy matrices that utilize dynamic imine exchange have been used to fabricate carbon fiber-reinforced composites. These composites exhibit a modulus of 56 GPa and a flexural strength of 1028 MPa. They retain 70–80% of their mechanical properties after three chemical recycling cycles (Figure 16) [153].

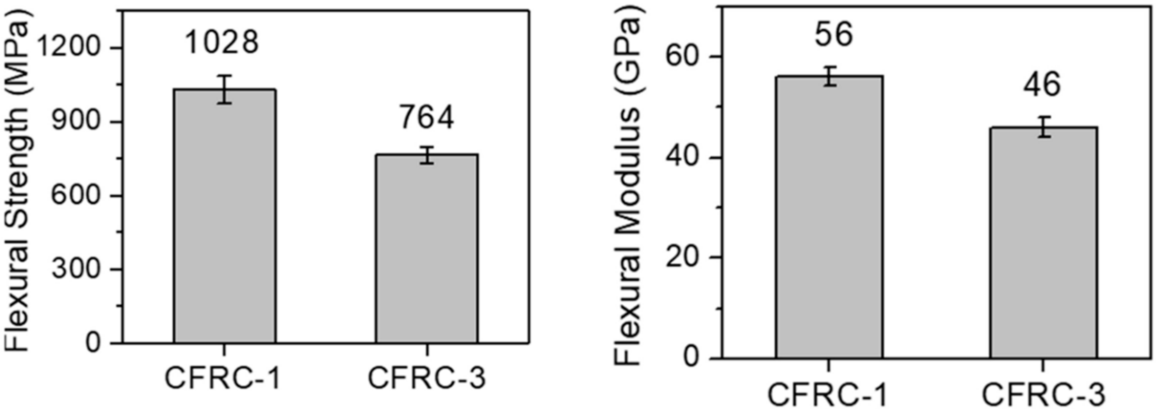

Figure 16. Flexural strength and modulus of the vitrimer composites measured by three-point bending test (ASTM D7264) [153].

Denissen et al. [154] studied glass fiber-reinforced composites containing vinylogous urea matrices that undergo dynamic amine exchange reactions, achieving a tensile strength of 791 MPa and a modulus of 40 GPa. These composites can be reshaped at 150 °C and reprocessed in N-methylpyrrolidone at 100 °C after 15 min, or at room temperature after 6 h. Similarly, composites containing aromatic disulfide bonds enable dynamic metathesis processes, achieving tensile strengths of approximately 800 MPa and flexural strengths of 903 MPa. These composites recover nearly all their flexural properties after hot-pressing at 160 °C for 5 min [155]. Bio-based epoxy vitrimers synthesized from vanillin and 4-aminophenol show a tensile strength of 145.4 MPa and a modulus of 1.18 GPa. After reprocessing at 180 °C and 20 MPa for 10 min, they form short-fiber composites with a modulus of 760 MPa and a tensile strength of 11 MPa [155]. Liu et al. [156] investigated carbon fiber-reinforced composites based on vanillin and diamines (m-xylylenediamine and 1,6-hexanediamine) cured with imine-linked DGEBA epoxy. These composites demonstrated a tensile strength of 622 MPa and a modulus of 19.3 GPa. After chemical dissolution in hydrochloric acid, the recovered carbon fibers retained similar surface morphology and chemical structure to virgin fibers. In another study, a carbon fiber-reinforced vitrimer composite cured with 4-aminophenyl disulfide and DGEBA epoxy achieved a remarkable tensile strength of 1460 MPa and a modulus of 144 GPa. Table 2 mechanical property data across conventional thermosetting composites and vitrimer-based composites. It also includes retention rates of mechanical performance a recycling, highlighting that epoxy-based vitrimer composites retain 70–80% of flexural strength after three recycling cycles, while conventional epoxies show irreversible property loss.

Table 2. Comparison of tensile, flexural, and modulus values of vitrimer-based and conventional pultruded composites, including performance retention after recycling.

Matrix Type	Reinforcement	Tensile Strength (MPa)	Flexural Strength (MPa)	Modulus (GPa)	ILSS (MPa)	Retention After Recycling	Test Standard	Ref.
Polyimine vitrimer	Neat polymer	~40	–	1.0		Significant recovery after reprocessing	ASTM D3039/ASTM D790	[36]
Polyimine vitrimer	Carbon fiber (single ply woven)	~400	~150	14.2		Excellent recovery after reprocessing	ASTM D3039/ASTM D790	[36]
Epoxy vitrimer (imine-cured DGEBA)	Carbon fiber	–	1028	56		70–80% after 3 chemical recycling cycles	ASTM D790	[153]
Epoxy vitrimer (disulfide cured DGEBA)	Carbon fiber	1460	–	144		Retained ~33 MPa tensile after grinding & hot-pressing	ASTM D3039	[75]
Bio-based epoxy vitrimer (vanillin-based)	Neat polymer	90	–	1.18		~85 MPa tensile after reprocessing	ASTM D3039	[132]
Conventional GFRP (pultruded)	Glass fiber	253	–	24.5		Not recyclable	ASTM D3039/ASTM D790	[114]
Conventional hybrid (kenaf/glass)	Hybrid fibers	–	–	–	21.5	Not recyclable	ASTM D2344 (ILSS)	[115]

After mechanical grinding and hot-pressing at 190 °C under 100 bar for 5 min, the composite maintained a tensile strength of 33 MPa and a modulus of 16 GPa [75]. Zaho et al. [157] highlight the formulation of sustainable epoxy vitrimer materials utilizing epoxidized soybean oil (ESO), tyramine-based bisphenol derivatives (TTP), and 4,4′-diaminodiphenyl disulfide (4-AFD). The resultant epoxy vitrimer (ETA) has superior thermal stability, mechanical strength (29 MPa), self-healing ability (within 30 min), solvent resistance, and shape memory functionalities attributed to the incorporation of dual dynamic covalent bonds (imine and disulfide).

6.6. Recyclability and Repairability of Fiber-Reinforced Vitrimer Composites

This section discusses the recyclability and self-healing properties of vitrimer composites, focusing on physical and chemical recycling methods, followed by an evaluation of their benefits and limitations. Additionally, the self-healing capabilities of vitrimers offer promising potential for extending the lifespan of polymer composites.

6.6.1. Physical Recycling Technique

In vitrimer recycling, physical recycling involves mechanically breaking down materials, often by crushing them into powders or granules for reuse [53,157]. Common physical recycling methods include extrusion molding, injection molding, and hot pressing. The effectiveness of these methods depends on factors such as temperature, pressure, and particle size, which directly influence the properties of the recycled material [158,159]. Yu et al. [159] explored the effect of reprocessing parameters on transesterification-based vitrimers. Epoxy resin cured with a 5 mol% zinc catalyst and carboxylic acid (77% triacid, 23% diacid) was ground into powder (25–120 μm) and reprocessed. After 30 min of pressing at 180 °C and 45 kPa, the recycled sample retained 76.7% of its original tensile strength (2.9 MPa). Zhang et al. [160] investigated the influence of various processing conditions—such as particle size distribution, temperature, and pressure—on recycled vitrimers. Finer powders improved contact surfaces and mechanical properties, while higher temperatures enhanced mechanical strength during epoxy-anhydride vitrimer recycling. The impact of temperature on the recycling of epoxy-anhydride vitrimers demonstrated that temperature is essential for increasing the mechanical properties of the recycled vitrimer [161].

6.6.2. Chemical Recycling of Vitrimer

Imine Exchange-Based Recycling

Chemical recycling of vitrimer involves breaking down vitrimer networks into oligomers or monomers using solvents [162]. Zhao et al. [24] studied an imine-embedded bisphenol network (EN-VAN-AP), which was chemically broken down in DMF with hydrochloric acid and later reformed into a solid upon reheating as displayed in Figure 17.

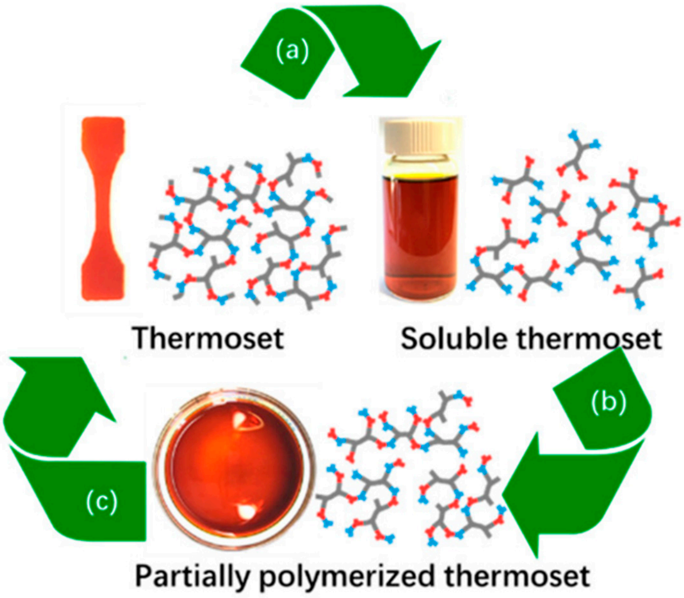

Figure 17. Chemical recycling of vitrimer [24].

Lessard et al. [162] developed a one-step method to transform linear prepolymers—synthesized via controlled radical copolymerization of methyl methacrylate and (2-acetoacetoxy)ethyl methacrylate—into vitrimers using a trifunctional amine treatment. The chemical recovery process illustrated in Figure 18 involves introducing an excess of a low-boiling-point monofunctional amine (e.g., n-butylamine) into the solvent, facilitating the breakdown of the vitrimer network. A precipitation technique is then used to recover the modified prepolymer from the solution. Finally, tris(2-aminoethyl)amine (TREN) is reintegrated into the system during a continuous curing process, allowing for network regeneration and restoring the vitrimer's original properties.

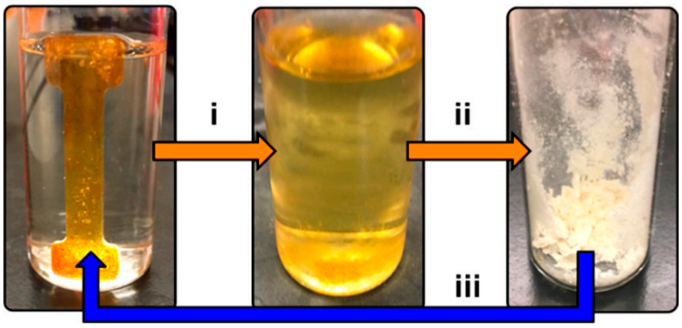

Figure 18. Chemical recycling of catalyst free vitrimer [162].

In the context of the circular economy, there has been a growing emphasis on utilizing reusable and bio-sourced materials for developing covalent adaptable networks (CANs) and their corresponding carbon fiber composites (CFRPs). Among these materials, vanillin, a derivative of lignin has gained significant attention due to its potential for creating sustainable and recyclable polymer systems. Researchers have developed various vanillin-based crosslinkers to fabricate dynamic polyimine thermosets and their composites. Notable examples include vanillin-terminated phosphazene monomer (HVP), methacrylated vanillin (MVL), and vanillin-terminated bisaminomethyl cyclohexane (VAN-BAC). For instance, Zamani et al. [163] used vanillin as a precursor to synthesize imine bond-based polymers and composites with high crosslink densities as depicted in Figure 19. These composites exhibited material properties comparable to conventional, non-recyclable epoxy-based carbon fiber composites. Compared to traditional composites, the imine-based composites reinforced with carbon fiber demonstrated impressive mechanical properties, including tensile strengths of 455–606 MPa, a Young's modulus of 42.3–47.3 GPa, and bending strengths ranging from 461 and 622 MPa [163]. The key advantage of these imine-bonded composites lies in their closed-loop recyclability, particularly in acidic environments, making them more sustainable than conventional epoxy systems. In another study, Zhao et al. [164] synthesized a vitrimer system by combining a synthetic monoepoxide containing a formyl group with vanillin and a diamine, leading to the formation of a Schiff base structure within an epoxy network. This vitrimer exhibited properties comparable to a non-recyclable bisphenol A epoxy resin, including a glass transition temperature (Tg) of 172 °C, a tensile strength of 81 MPa, and a modulus of 2112 MPa. The vitrimer demonstrated excellent thermal stability, with only 5% weight loss at 323 °C. When applied in carbon fiber composites, this material showed enhanced mechanical performance, achieving a tensile strength of 763 MPa and a tensile modulus of 35.3 GPa, while retaining the ability to be recycled under mild conditions. Beyond vanillin, researchers have explored other renewable bio-sourced materials such as soybean oil, glycerol triglycidyl ether, and cellulose to develop CAN-based systems. These materials offer significant potential for creating recyclable carbon fiber-reinforced polymers (CFRPs) with superior mechanical properties, both before and after recycling, contributing to a more sustainable composite manufacturing process.

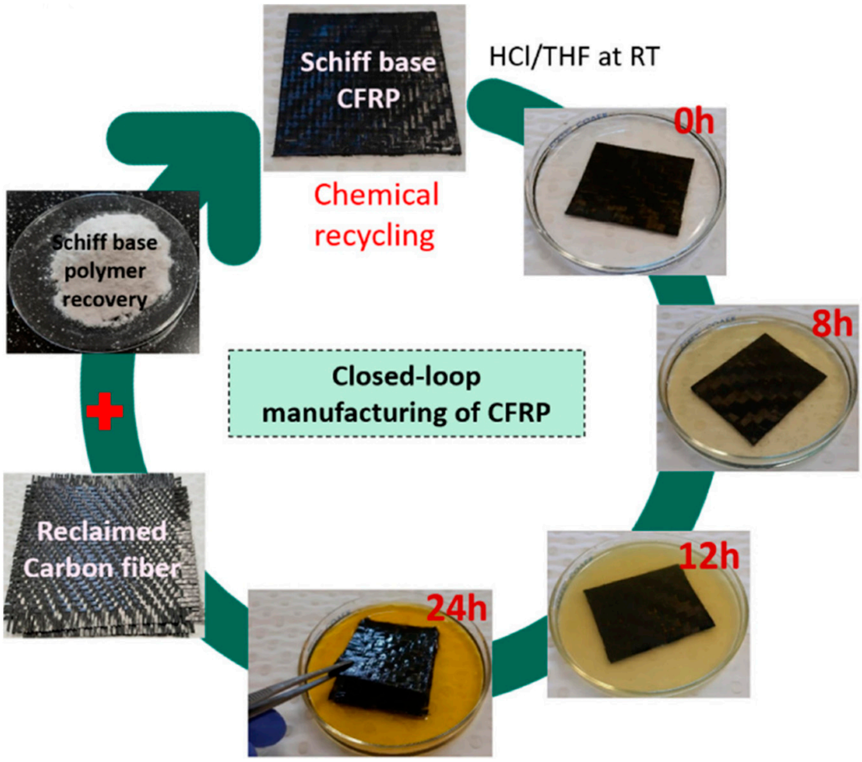

Figure 19. Closed-loop chemical recycling process of carbon fiber composite [163].

Transesterification-Based Recycling

Epoxy-based polymer systems containing ester linkages are commonly used in the manufacturing of recyclable carbon fiber composites (CFRPs) due to their favorable reprocessing characteristics. Hao et al. [165] developed a recyclable composite using an epoxy/anhydride thermosetting system with a high glass transition temperature (Tg > 200 °C). In this system, tetraglycidyl methylenedianiline (TGDDM) served as the epoxy matrix, while triethanolamine (TEOA) acted as the co-curing agent. The composite could be effectively recycled under mild conditions without the need for external catalysts. This recyclability is attributed to the catalytic activity of tertiary amines present in TGDDM and TEOA, which facilitate hydrolysis of the polymer matrix in aqueous solutions. Additionally, the hydroxyl groups in TEOA promote transesterification reactions with ester bonds, enhancing the material's degradability and recyclability [166]. The recyclability of these CFRPs under moderate conditions allows for nearly complete preservation of the carbon fiber structure, enabling their reuse in producing new composites. This process is illustrated in Figure 20. The tensile strength of the recycled composite reached 437 MPa, retaining approximately 87% of the original composite's tensile strength (502 MPa) [167].

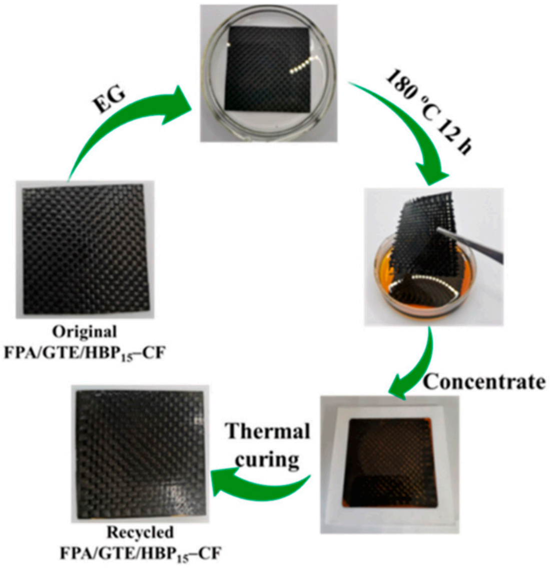

Figure 20. Recycling carbon fiber composite [167].

In another study, Zhang et al. [166] developed covalent adaptable networks (CANs) by curing phthalic anhydride (PA) with glycerol and diglycidyl 4,5-epoxycyclohexane-1,2-dicarboxylate (DGEDC). The polymer network, rich in free hydroxyl groups, allowed for rapid transesterification at 180 °C within 205 s, facilitating degradation, reprocessability, and recycling. The resulting carbon fiber composites exhibited superior mechanical properties, including a modulus of 8.5 GPa and a tensile strength of 559 MPa significantly higher than those of pure CANs (tensile modulus: 3.2–3.9 GPa, tensile strength: 65–78 MPa). Additionally, the composites could be broken down at 190 °C in ethylene glycol, and the recycled fibers retained their mechanical properties, with an elongation at break of 1.24% and a tensile strength of 3.55 GPa, comparable to the original fibers (1.35% and 3.78 GPa).

The further advanced recyclable vitrimer composites by using fumaropimaric acid (FPA), glycerol triglycidyl ether (GTE), and hydroxy-terminated hyperbranched polyesters (HBPs) [168]. The incorporation of HBPs significantly improved the composite's mechanical properties, achieving an interlayer shear strength of 43 MPa and a tensile strength of approximately 585 MPa. The reprocessed composites retained strong mechanical properties, with an interlayer shear strength of 35 MPa and a tensile strength of 467 MPa [169]

(Figure 21b). Similarly, Qi et al. [170] demonstrated that carbon fibers could be effectively recycled by immersing composites in heated ethylene glycol. The process dissolved the epoxy matrix and facilitated transesterification reactions, enabling the recovery of carbon fibers with unchanged mechanical characteristics. The recycled fibers exhibited a modulus of 223 GPa and a tensile strength of 4.4 GPa, nearly identical to the original fibers. Furthermore, recycled carbon fiber composites retained almost 100% of their original mechanical properties, highlighting the effectiveness of this recycling method.

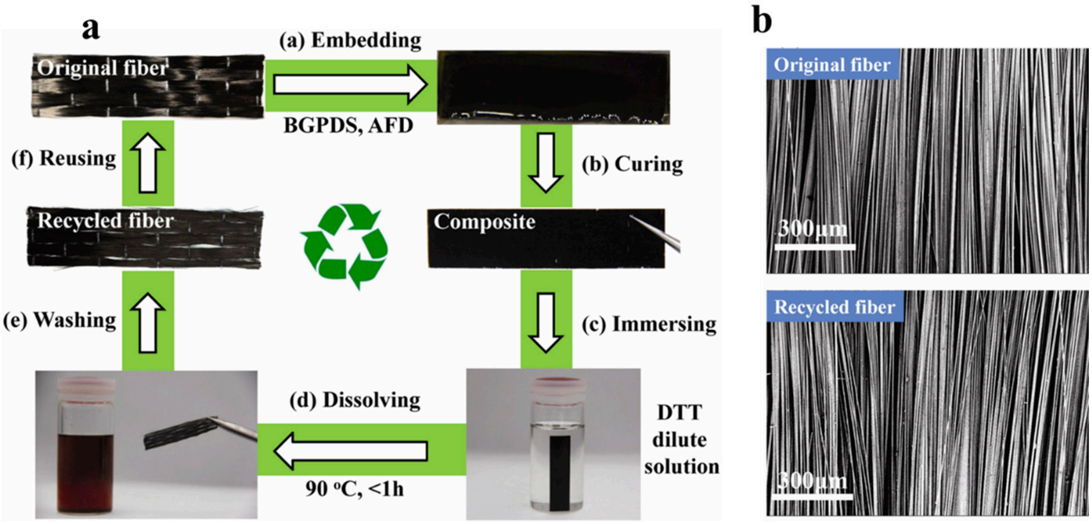

Figure 21. (**a**) Recycling of carbon fiber vitrimer composites, and (**b**) SEM image of original and recycled fiber [169].

Disulfide Exchange-Based Recycling

Epoxy resins have gained significant attention for formulating cross-linked networks (CANs) using disulfide linkages to develop recyclable carbon fiber-reinforced polymers (CFRPs). These disulfide bonds facilitate dynamic covalent exchanges, enhancing recyclability and mechanical performance [168]. Si et al. [169] synthesized an epoxy vitrimer enriched with a high concentration of exchangeable disulfide bonds. This design enhances metathesis reactions within the polymer network, improving the recycling performance of vitrimer-based CFRPs. When treated with dithiothreitol (DTT), the vitrimer undergoes complete dissolution, with dual disulfide bonds degrading more rapidly than those with a single disulfide linkage. The CFRPs developed using these vitrimers exhibited excellent mechanical properties, including a tensile strength of 334.5 MPa and a storage modulus of 10.5 GPa. Following DTT treatment, both the carbon fibers and polymer matrices could be effectively recycled to regenerate CFRPs, as illustrated in Figure 21a. Importantly, the recycling process preserved the structural integrity of the carbon fibers, as shown in Figure 21b. The recycled CFRPs retained impressive mechanical capabilities, including a tensile strength of 320.9 MPa and a storage modulus of 8.2 GPa, indicating that their performance remained comparable to the original material supporting the concept of closed-loop recyclability. Zhou et al. [155] further advanced this approach by developing CFRPs using dynamic polyurea/epoxy (DPE) vitrimers featuring disulfide linkages. In their synthesis, diglycidyl ether of bisphenol A (DGEBA) and hexamethylene diisocyanate (HDI) were combined with 4-aminophenyl disulfide (AFD), which served as a dynamic crosslinker. The fast disulfide metathesis reactions allowed the DPE resins to be reused at low temperatures, facilitating efficient recycling. The CFRPs fabricated from DPE resins exhibited outstanding mechanical performance, including flexural strength of 903 MPa, Interlaminar shear strength of around 50 MPa, tensile strength of around 400 MPa, and tensile strain of 3.5%. These CFRPs demonstrated excellent chemical recyclability, allowing for the recovery of carbon fibers (CFs) without compromising solvent resistance. The ability of these CFRPs to undergo multiple recycling cycles without significant degradation highlights their potential for sustainable composite manufacturing. This advancement paves the way for next-generation CFRPs that incorporate highly sustainable production techniques while maintaining strong mechanical properties and structural integrity.

Qin et al. [170] developed closed-loop recyclable carbon fiber-reinforced polymers (CFRPs) using reversible amidation chemistry. This approach is based on the cleavage of tertiary amide bonds formed from the reaction between maleic anhydride and secondary amine. These bonds can be fully broken under highly acidic conditions. The polymer networks were constructed using a multifunctional macromonomer (LPEI) and a bifunctional crosslinker (BA), which served as the polymer matrix for fabricating the CFRPs. The resulting composites exhibited excellent mechanical performance, including tensile strength of 466 MPa, elongation of break around 4%, and Young's modulus of 13.1 GPa. Under acidic conditions, the polymer matrices underwent depolymerization, allowing for the efficient extraction of intact carbon fibers (CFs) and recovery of the pure monomers (LPEI and BA). Both the reclaimed carbon fibers and monomers retained their original structural and chemical properties. The recovered monomers demonstrated high purity, enabling multiple reuse cycles in the synthesis of new CFRPs. This closed-loop recycling process allowed for the preservation of over 95% of the original mechanical properties in the regenerated composites, making this method highly effective for sustainable CFRP production.

Vitrimer composites enhance the repair capabilities of thermoset materials through dynamic covalent bonding. When the original covalent bonds break due to damage, the material can largely recover its mechanical performance through the reformation of dynamic covalent bonds. A key feature of vitrimers is their ability to undergo reversible crosslinking at elevated temperatures, allowing for self-healing under suitable conditions. For effective repair, applying pressure during the vitrimer healing process is essential to ensure sufficient contact at the damaged interface. This promotes efficient bond exchange and structural restoration. Wu et al. [171] synthesized epoxy vitrimers through transesterification, creating a crosslinked network enriched with primary amines and a high density of hydroxyl groups. This chemical modification enhanced the bond exchange rate, significantly improving the material's self-healing properties. As illustrated in Figure 22, the vitrimer composite effectively repaired visible surface damage. The study concluded that applying elevated temperatures and pressure can efficiently heal matrix cracks and delamination in the composite material. This ability to restore structural integrity through dynamic bond exchange offers significant advantages for extending the lifespan and durability of vitrimer-based composites.

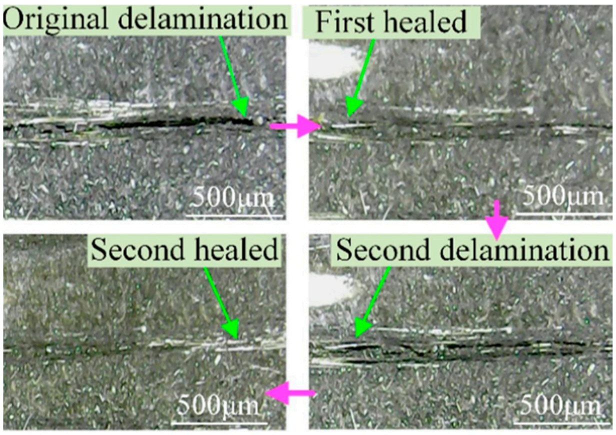

Figure 22. Reparability of the vitrimer carbon fiber composite [171].

In another research Patel et al. [172] investigated the self-healing ability of vitrimeric poly(hindered urea) (PHU) networks for maintaining triboelectric device performance after surface damage. Micron-scale abrasions were introduced on a 0.2 wt% CNT/PHU composite sheet using sandpaper. The material's reversible hindered-urea chemistry enabled surface restoration through a self-healing process at 90 °C and 80% humidity for 20 min. As shown in Figure 23, the composite effectively recovered its original state, demonstrating strong potential for durable energy-harvesting applications.

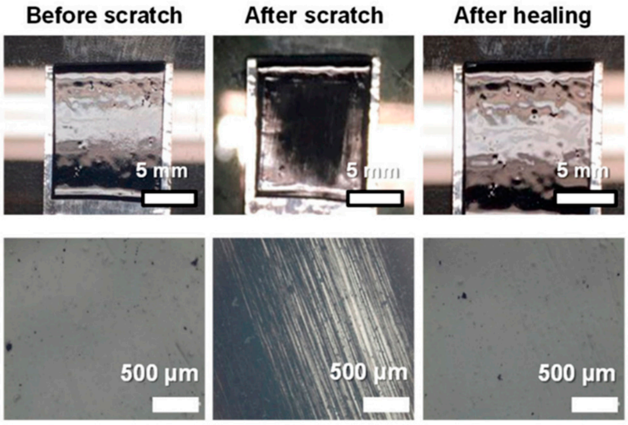

Figure 23. Self-healable vitrimer composite (0.2 wt % CNT) before and after healing [172].

Yu et al. [173] introduced a novel self-healing method for carbon fiber-reinforced vitrimer composites using dynamic imine bonds that reversibly break and reform upon heating. A mixture of diethylenetriamine (DETA) and ethanol dissolves the polyimine vitrimer at room temperature. To initiate the repair, the vitrimer powder is heated and sprayed onto the damaged surface, triggering bond reformation, as shown in Figure 24. This process effectively restores the mechanical integrity of the composite, enabling full recovery of its structural performance.

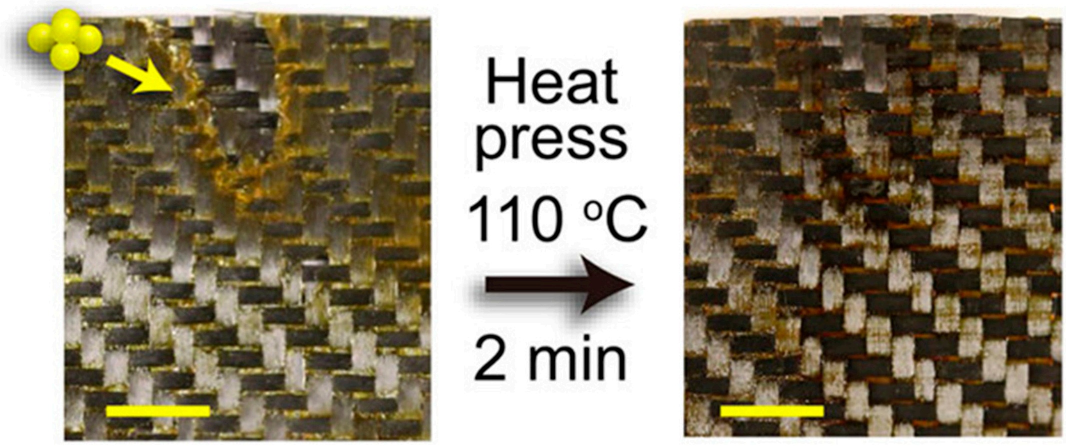

Figure 24. Repair of polyimine carbon fiber composites [173].

Koratkar et al. [174] demonstrated the ability to repair fatigue damage in carbon fiber/vitrimer composites by applying repeated heating. Microcracks, often invisible to the naked eye and caused by service-related fatigue, can degrade the composite's mechanical properties over time. Their study showed that simple thermal treatment effectively reversed fatigue-induced failures, significantly extending the composite's service life and durability. However, it is crucial that the vitrimer materials used possess sufficient thermal stability to prevent irreversible chemical changes from repeated heating.

7. Pultrusion of Fiber-Reinforced Vitrimer Composites

The limited research on the pultrusion of vitrimer composites can be attributed to the emerging nature of vitrimer technology and its specific processing challenges. The temperature-dependent bond exchange reactions and the need for precise viscosity control make vitrimers difficult to integrate into pultrusion processes, which inherently require strict temperature regulation and accurate curing conditions. Most current pultrusion research focuses on well-established thermosets and thermoplastics, while vitrimer applications remain largely within academic research, with limited practical implementation. Key challenges such as the scalability, mechanical efficiency, and thermal stability of vitrimers during pultrusion remain unresolved. However, the recyclability and self-healing properties of vitrimers align with sustainability goals, presenting a promising avenue for future research to bridge this gap.

Aranberri et al. [74] conducted one of the pioneering studies on the pultrusion of carbon fiber-reinforced epoxy vitrimer composites. Initially, they synthesized and characterized the vitrimer resin by combining EPIKOTE resin and 4-aminophenyl disulfide hardener and curing it in a mold under controlled conditions. To ascertain the resin's viscosity-temperature profile and curing kinetics, which are critical factors in pultrusion, these preliminary procedures were implemented. The viscosity of the vitrimer resin at 45 °C was found to be within the optimal range for successful pultrusion, as illustrated in Figure 25a, which compares the viscosity of the vitrimer resin to a reference resin at both 25 °C and 45 °C. The resulting carbon fiber composite fabricated with vitrimer resin is shown in Figure 25b. In terms of mechanical and thermal properties, the epoxy and vitrimer resin composites demonstrated similar performance. However, a key advantage of the vitrimer composite was its ability to undergo complete recycling. Through mechanical grinding and reprocessing of the powdered pultruded composite, a second-generation composite was successfully fabricated, incorporating short fiber reinforcement. This demonstrates the potential of vitrimer-based composites for closed-loop recyclability, marking a significant advancement in sustainable composite manufacturing.

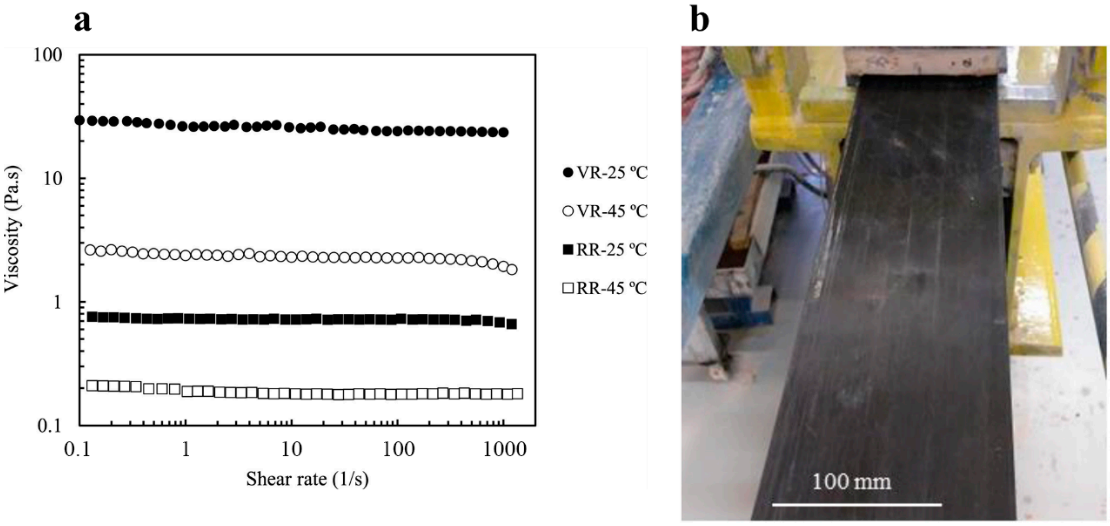

Figure 25. (**a**) Viscosity of the vitrimer and reference resin @ 25 and 45 °C, and (**b**) pultruded vitrimer carbon fiber composite [74].

8. Applications of Pultruded Fiber-Reinforced Composites—Vitrimer

Pultruded fiber-reinforced composites are polymer-based materials strengthened with fibers through the pultrusion process, producing materials with high strength, lightweight properties, and exceptional corrosion resistance. Due to these advantageous characteristics, pultruded composites are widely used across various industries for both structural and functional applications. Pultrusion is primarily used to manufacture beams, channels, and rods for structural elements in bridges, buildings, and platforms, as well as in concrete reinforcement, offering superior corrosion resistance and durability compared to traditional materials like steel. Additionally, pultruded composite gratings and decking are extensively utilized in corrosive environments such as wastewater treatment plants and chemical processing facilities. Their robustness and lightweight nature also make them ideal for use in railway sleepers, platforms, and various other structural applications. In the automotive sector, pultruded composites are utilized in products ranging from roof supports to bumpers due to their lightweight yet strong composition. In the marine industry, these composites are valuable in shipbuilding, dock construction, and as marine fenders, where resistance to seawater corrosion is crucial. In the energy sector, pultruded composites are used for components such as turbine blades, nacelle covers, power poles, and crossarms, offering a sustainable and efficient alternative to traditional materials. Their ability to replace conventional materials like steel and aluminum provides both economic and environmental benefits, making them favorable for modern engineering and manufacturing applications.

Key application areas for vitrimer composites include:

Vitrimer composites have diverse applications across various industries due to their recyclability, self-healing capabilities, and mechanical strength. In the aerospace industry, lightweight and durable carbon fiber vitrimer composites are used for components that enhance sustainability by reducing waste. To meet aerospace service conditions, vitrimer matrices must achieve Tg and Tv values above 200 °C, ensuring thermal stability under high operational loads. Recent cost models suggest vitrimer resins could reach ~USD 2.28/kg for bio-based systems when scaled [175], placing them within competitive range for secondary aerospace structures if performance advantages are realized. In the automotive sector, they are applied in manufacturing bumpers, panels, and interior components, offering extended lifespans through self-healing and recyclability. Because the automotive industry is highly cost-sensitive, vitrimer monomer prices must remain close to epoxy baselines (USD 1.5–2.0/kg [176]). Current pilot vitrimer systems are more expensive (~USD 5.37/kg), but recyclability and repairability could offset lifecycle costs by reducing part replacement. Hybrid fiber reinforcement strategies (e.g., glass/carbon blends) are particularly promising for lowering cost without sacrificing mechanical performance. In electronics, they improve thermal stability and facilitate easier recycling for circuit boards and enclosures, aiding in efficient electronic waste management. Their versatility in additive manufacturing makes them ideal for 3D printing complex, customizable components with high precision. In medical applications, their flexibility and biocompatibility make them suitable for devices and implants. For energy storage, they are used in battery casings and supercapacitors due to their mechanical strength and thermal stability. In construction, they are used in reinforced concrete and modular elements for their durability, reparability, and sustainability. Scaling challenges include the limited industrial production of vitrimer resins and the cost of retrofitting pultrusion equipment for new curing/monitoring regimes. However, the potential for closed-loop recyclability could significantly reduce material waste in infrastructure projects, aligning with sustainability-driven policies. Sports equipment like tennis rackets, bicycle frames, and helmets benefit from their lightweight and robust characteristics. In the marine industry, these composites are employed in corrosion-resistant, lightweight components for boats and offshore structures. Cost modeling frameworks developed for fiber-reinforced composites [176] can be adapted to estimate the total cost of vitrimer pultrusion in these sectors, accounting for resin, fiber, labor, and equipment adaptation. Such models are essential to demonstrate feasibility for large-scale marine and packaging applications. Additionally, their reusability and reprocessability align with the principles of the circular economy, making them ideal for sustainable packaging solutions. Despite their promising potential, research on pultrusion of vitrimer-based composites remains scarce due to the emerging nature of vitrimer technology and the specific challenges related to processing. Currently, there are no substantial studies exploring the pultrusion of vitrimer composites for industrial applications. Given their recyclability, self-healing capabilities, and alignment with sustainability goals, there is a clear need for in-depth research to investigate their potential for industrial scale pultrusion. Advancements in this area could revolutionize composite manufacturing by offering durable, eco-friendly materials suitable for diverse sectors.

9. Challenges and Future Directions

The pultrusion of vitrimer composites presents significant potential for advanced applications yet faces several scientific and practical challenges that must be addressed to unlock their full potential.

9.1. Challenges

The primary challenge lies in the dynamic covalent bond exchange mechanisms of vitrimer resins, which require precise temperature control and curing conditions. Achieving an optimal balance between flowability for effective fiber impregnation and network formation for structural integrity is critical. Another hurdle is ensuring strong interfacial adhesion between fibers and the vitrimer matrix, particularly in dynamic systems where bond exchanges occur. Elevated temperatures required for these reactions can degrade certain fibers or compromise their mechanical properties. Additionally, the curing time for vitrimers is often longer than that of traditional thermosets, potentially reducing production efficiency and increasing costs. Scaling up for high-volume, cost-effective pultrusion also remains technically challenging due to the higher costs of vitrimer monomers and catalysts, coupled with the need for precise temperature control and potential post-processing for reshaping or recycling. Furthermore, the lack of standardized testing protocols and certification frameworks for vitrimer composites hinders their adoption in regulated industries such as aerospace and automotive. Addressing these challenges through material innovation, process optimization, and the development of industry standards will be essential to fully harness the potential of vitrimer composites in advanced applications.

9.2. Future Directions

Vitrimer composite pultrusion development requires targeted innovation to overcome current limitations and bring out their best for industrial applications. Next-generation research needs to be application-focused vitrimer resins with disulfide, imine, and vinylogous urethane-linkage functional groups to enable the tuning of bond-exchange rates, decrease processing temperatures, and enhance the mechanical properties to facilitate successful pultrusion. Hybrid fiber composites (carbon/glass or carbon/natural fibers, for instance) must be optimized through design of experiments (DOE), response surface methodology (RSM), and finite element modeling (FEM) to determine the fiber volume fractions and stacking sequences that maximize toughness and stiffness, while minimizing the cost of the product. Some possible methods for online monitoring and automatic control include the incorporation of fiber Bragg grating (FBG) sensors to track strain and temperature, infrared thermography to measure die wall and composite surface temperature distributions, and ultrasonic probes to monitor resin penetration and cure evolution. 1–10 Hz data collection frequencies are usually required to capture rapid-changing curing, and predictive optimization of pulling speed, die temperature, and resin feed can be facilitated by advanced machine learning-based models. Novel online monitoring systems for viscosity further offer avenues for closed-loop feedback to ensure stable process conditions. At the level of sustainability, recycling technologies scalable upward such as thermal depolymerization or chemical selective recycling would be benchmarked against life cycle assessment (LCA) to quantify environmental and economic gains against conventional materials. Blending nanomaterials or smart additives would provide supplementary functionality such as conductivity, fire resistance, or self-sensing, and opening new fields of application. Finally, the making of these strategies will be a product of interdisciplinary research between materials science, chemical engineering, and industrial process control. Continuous innovation and investment will be the drivers to further solidify pultruded vitrimer composites as next-generation sustainable, high-performance materials.

10. Conclusions

Pultruded vitrimer composites represent a significant advancement in the field of composite materials, offering a promising pathway toward sustainable, high-performance solutions. These materials combine the inherent mechanical strength of fiber-reinforced composites with the dynamic covalent bonding properties of vitrimers, enabling recyclability, self-healing, and extended material lifespans. Their unique ability to undergo reversible bond exchange under specific conditions allows for reshaping, repair, and reprocessing without compromising mechanical or structural integrity. Beyond their robustness and thermal stability, these composites align with sustainability goals, offering a significant advantage over conventional thermosets, which are difficult to recycle due to their permanent cross-linked structures. The potential for closed-loop recycling makes vitrimer-based composites highly relevant for industries such as aerospace, automotive, construction, and renewable energy, where reducing environmental impact and improving material efficiency are increasingly vital. However, challenges remain in the pultrusion process of vitrimer composites. Managing temperature-dependent bond exchange reactions during processing—particularly concerning resin viscosity, cure cycles, and fiber impregnation—remains a significant hurdle. Achieving strong fiber–matrix adhesion is critical for ensuring the composite's mechanical performance. Additionally, issues such as longer processing times and temperature sensitivity hinder the scalability of vitrimer-based composites for mass production. Addressing these challenges through continued research and innovation will be essential to fully harness the potential of pultruded vitrimer composites in industrial applications.

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