

Recyclable Vitrimer-Matrix Carbon-Fiber Composites for Aerostructures: Reprocessing Efficiency and Retention of Mechanical Performance

Krupal Pawar^{1,*}, Rameshwar Kadu², Yogesh Pathare³, Rajeshkumar Sambhe⁴,
 Sailee Shelke², Vasudha Patil⁵

Abstract

To begin with, carbon fiber reinforced polymers (CFRP) have become an important part of the structural loading in many modern aircraft; however, due to their permanent cross-linking in thermosets, they cannot easily be restored after being retired. Vitrimerers have been proposed as a solution since their Covalent Adaptable Networks (CANs), rearranged by associative exchange, may be reshaped, welded, repaired or dissolved without damaging the CFRP fibers. This paper reviews whether the “promise” of vitrimers meets the needs of the aerospace industry, using peer reviewed publications from 2021 to 2026 and combining these into a unified analysis format. In addition to separating reprocessing efficiency from mechanical performance retention and normalizing each fiber volume fraction where possible, it was found that continuous reinforcement could be recovered fully under mild processing conditions such that tensile strengths were nearly identical. Additionally, it was determined that approximately 94% of the original short beam shear strength of recycled laminates would be recovered provided that the fiber content is considered. Since mechanical properties are dominated by the matrix rather than the fiber, there are no issues related to recycling, but there are limitations including the very small range between the exchange temperature and the start of thermal degradation in high Tg systems.

Keywords: Vitrimer composites, covalent adaptable networks, carbon-fiber recycling, aerostructures, property retention

*Author for Correspondence

Krupal Pawar

E-mail id: krupalpawar@gmail.com

¹D.Sc. Researcher, Department of Computer, Engineering and Technology, Sikkim Sardar Patel University, Jorethang Road, Namchi, South Sikkim, India

²Assistant Professor, Department of Mechanical Engineering, Pravara Rural Engineering College, SPPU, Loni, Maharashtra, India

³Assistant Professor, Department of Mechanical Engineering, Dr. Vitthalrao Vikhe Patil Collage of Engineering, SPPU, Ahilyanagar, Maharashtra, India

⁴Professor, Department of Mechanical Engineering, Jawaharlal Darda Institute of Engineering & Technology, SGBAU, Yavatmal, Maharashtra, India

⁵Assistant Professor, Department of Artificial Intelligence & Data Science, Shri Chhatrapati Shivaji Maharaj College, SPPU, Nepti, Ahilyanagar, Maharashtra, India

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INTRODUCTION

Fewer material changes in aviation than the transition from aluminum alloys to polymer composites in the primary structure of modern airplanes are evident. The wings, fuselage barrels, empennages, and control surfaces of current wide-body commercial airplanes are made mainly of carbon fiber-reinforced polymers (CFRP). That said, the reasons behind the use of CFRP are well known. Specifically, the combination of high-specific-stiffness and high-specific-strength characteristics exhibited by CFM materials far exceed those of metallic materials, along with good resistance to fatigue failure, and zero risk of galvanic-corrosion due to electrochemical differences in the galvanic series (Figure 1).

It is less commonly stated with equal enthusiasm that the chemical characteristics that provide the superior performance of CFRP make an airplane

nearly impossible to dismantle after service. Once a thermosetting resin – such as epoxy – has cured and formed a three-dimensional molecular network through cross-linking of molecules via irreversible covalent bonding, the network is rigid and has no inherent thermodynamic pathway to reformulate itself into individual components without causing significant damage to either the fibers or the resin matrix [1, 2].



Figure 1. Close-up of a 2/2 twill carbon-fiber reinforcement of the type used in aerostructure skins and secondary structure. The architecture that gives the laminate its specific stiffness and strength is also what makes an irreversibly cross-linked matrix so difficult to separate from the fiber at end of life.

Consequently, several issues related to recycling of aerospace composites are emerging. Approximately eight thousand commercial airplanes are forecasted to reach the end of their operational life in the next ten years. The amount of composite waste generated annually by manufacturing scrap and retirements has been estimated to be tens of kilo-tons [1]. Technologies currently exist for recovering materials from these wastes. For example, pyrolysis and solvolysis can separate the fibers from the matrix; however, each of these processes reduces the quality of the recovered materials. Specifically, fibers removed using pyrolysis or solvolysis are typically shorter, misaligned and lack sizing. In addition, the matrix is destroyed during pyrolysis and solvolysis processes and is released as char or as a solvent stream requiring additional treatment prior to reuse. Furthermore, most recovered materials are used in non-structural or semi-structural applications that are outside of the structural requirements for continued flight [3, 4]. Consequently, down-cycling (i.e., reducing the value or utility of a material by converting it into a lower-value product) represents a true loss of material rather than merely an accounting convenience. As mentioned earlier, virgin carbon-fiber carries an embodied-energy-content of approximately 183–286 MJ/kg [4]. Thus, recycling these materials represents a viable option to reduce the environmental impact associated with their production.

Life cycle assessments of carbon-fiber reinforced polymers (CFRP) in aerospace structures indicate that the method(s) employed for treating CFRP at the end of life significantly affect the overall environmental benefit associated with using CFRP in aerospace structures [5, 6]. Specifically, recycling CFRP in a closed loop system appears to offer better environmental benefits than landfills or energy recovery.

Vitrimers represent a new class of engineering materials that potentially could address many of the concerns associated with recycling aerospace composites. Unlike traditional thermoplastics that melt upon heating, vitrimers maintain their solid-state morphology until heated beyond a temperature known as their “topology freezing” point. At temperatures above this point, vitrimers flow like a viscous liquid instead of melting. Vitrimers achieve this behavior through reversible association/dissociation reactions among their molecular building blocks. More specifically, vitrimers contain cross-links that periodically dissociate while forming temporary associations allowing segments of the molecule to move relative to

each other. These association-dissociation events occur reversibly so long as the vitrimer is maintained at temperatures below its topology freezing point. While maintaining a constant level of cross-link density over time, vitrimers remain dimensionally stable and insoluble at temperatures below their topology freezing points. Upon reaching temperatures greater than their topology freezing points, vitrimers flow in a fashion like that observed with viscous liquids rather than undergoing melting like conventional thermoplastics. From a practical perspective, vitrimers enable laminated composites to be repeatedly molded and shaped, bonded together and/or selectively dissolved using chemicals capable of disrupting network stoichiometry. Additionally, vitrimers may release composite reinforcements intact without resorting to mechanical methods of separation such as grinding/cutting [7, 8].

Substantial amounts of research have established all four reprocessing functions described above at the coupon-scale. However, little evidence exists demonstrating that each function works at scales required by an aerospace manufacturer or regulatory agency. Specifically, chemistry-based demonstrations typically measure tensile strength of neat resins before and after re-molding; whereas aerospace manufacturers require laminate level allowable values under hot/wet conditions, various types of hole configurations (e.g., open-hole vs. filled-hole), inter-laminar fracture toughness values (mode I and mode II), and assurances that none of these properties degrade significantly across multiple reprocessing cycles for which the material is marketed [1]. Furthermore, literature lacks standardization regarding reporting retention measurements. When comparing a composite that has undergone dissolution followed by subsequent manufacture to its original counterpart, the former will often have a different fiber volume fraction and void content from its precursor. Consequently, direct comparisons of strength conflate a chemistry-based effect with a process-based effect [7].

Therefore, this paper attempts to bridge this gap. It synthesizes results from experimental investigations of carbon fiber-vitrimer composite materials conducted between 2021 and 2026 into an integrated theoretical model that decouples two questions that are frequently addressed simultaneously: how effectively can the material be recycled? And how much of its mechanical properties survive recycling?

Three contributions are presented in this paper. Firstly, normalization conventions are introduced enabling comparison between retention figures measured independently in different laboratories. Secondly, existing data sets are evaluated to determine which property categories degrade significantly upon recycling and why. Finally, the remaining barriers to widespread aerospace structural adoption of vitrimer composites are identified in logical order, along with suggestions for experimental procedures necessary to overcome each barrier.

SCOPE AND METHOD OF ANALYSIS

Literature Selection

Sources of information included peer reviewed journals published since 2020 that reported experimental data regarding continuous fiber reinforced vitrimers; preference for use of data collected as part of laminate level testing versus neat resin testing when using carbon fibers. Review articles were used for background material as well as for finding sources of primary literature. Data (quantitative) from reviews were not used. Fiber reclamation quality was evaluated through all types of fiber (chopped, non-woven & short) however the strength data obtained from these samples were omitted from comparisons due to the difference in how loads are transferred in each type of laminate.

Definitions

The two efficiency measures employed throughout the research have been clearly stated due to the frequent ambiguity within the literature regarding what constitutes these measures. Mechanical retention is quantified by measuring a particular characteristic of a material prior to processing (first generation), and then subsequently upon subsequent processing (second generation). This measure of mechanical retention is typically calculated as a percentage. The measure of process efficiency describes the thermal, temporal and chemical costs associated with the achievement of that mechanical retention. These include temperature and residence time requirements necessary for the network

rearrangement, solvent volumes and solvent recovery burdens, and the amount of fiber and/or matrix that is recovered in a state suitable for reuse. It should be noted that an individual system may excel in one of these areas yet fail to achieve acceptable levels in the other area. For example, a matrix that is easily dissolved in excess amounts of glycol at 180°C will likely provide high values of mechanical retention, but it may lack industrial attractiveness. Conversely, a fast low-cost method that provides a low temperature processing route could potentially leave residue on the surface of filaments that compromises the integrity of the second-generation composite.

Normalization Convention

To account for differences in the fiber volume fractions (FVF) of the laminate, all results were normalized to FVF when comparing matrix dominated mechanical properties; this was done based on approaches used previously with filament winding [7]. The void content was considered a confounding variable as opposed to being inherent within the chemical make-up as it has been noted historically that there can be an approximate decrease in the flexural strength of carbon-epoxy laminates of up to a couple percent per percent voids. When the FVF was not reported by the primary source for a given sample, then the measured values were included and indicated appropriately. Normalizing FVF is important because simply comparing a first generation prepreg derived part to a second-generation part manufactured via wet winding will punish the recycling process for what may well have been a difference in manufacturing methods.

DYNAMIC NETWORK CHEMISTRIES RELEVANT TO AEROSTRUCTURES

Not every vitrimer chemistry is a plausible airframe matrix, and it is useful to separate the candidates before discussing performance. Four families dominate the recent literature [9, 10].

What unites these families, and separates them from thermoplastics, is that exchange is associative: a replacement bond forms before the original one is released, so the crosslink density and the load path through the network are preserved throughout reprocessing (Figure 2). The families differ chiefly in the temperature at which that exchange becomes fast, in whether a catalyst is required, and in how readily the network can be taken fully apart – and it is precisely those differences, rather than novelty of chemistry, that decide aerospace relevance.

Transesterification Networks

Epoxy networks cross-linked using anhydrides (e.g., glutaric anhydride) or carboxylic acids (e.g., acrylic acid) form β -hydroxyl ester bonds which can be exchanged via transesterification reactions, often facilitated by the presence of a metal catalyst (e.g., $\text{Zn}(\text{acac})_2$). Anhydride-based systems are still dominant in this area due to their use of pre-existing commercially available materials to create the monomer units. The effects of varying catalyst loadings have been demonstrated for a system consisting of diglycidyl ether of bisphenol A cured with glutaric anhydride. Catalyst loadings provide control over network dynamic behavior, and the measured activation energy for stress relaxation was ~ 60 kJ/mol; these results were associated with controllable reprocessing/repair characteristics exhibited by carbon-fiber laminates [11]. When applied to an industrially relevant prepreg system, a polyethersulfone modified epoxy vitrimer has been synthesized and used to produce laminates possessing a $T_g = 86.2^\circ\text{C}$ and a storage modulus of 16 GPa. Stress relaxation $> 200^\circ\text{C}$ was observed, and the relaxation activation energy was determined to be approximately 65.43 kJ/mole. Additionally, upon exposure to ethylene glycol at 180°C, the laminate returned to both the fibers and the matrix [12].

Aromatic Disulfide Networks

Disulfide metathesis proceeds at lower temperatures compared to transesterification and this is attractive from a processing point of view. Until now, the most comprehensive assessment of any vitrimers for aviation used epoxy formulation with aromatic disulfide hardener processed by resin transfer molding and subjected to campaign that included tension, compression interlaminar shear, in plane shear, tension and compression with open holes and compression with filled holes and toughness for Mode I and Mode II interlayers at room temperature and at 70 degrees Celsius and 120 degrees Celsius after saturation with moisture [13]. Results compare favorably with incumbent RTM systems and lower compression and shear scores trace to toughening veils rather than dynamic chemistry itself.

Depolymerization solvent free of bio networks using cystamine has also been demonstrated on carbon-fiber composites recovering amine terminated oligomers that can be recross-linked recently [8].

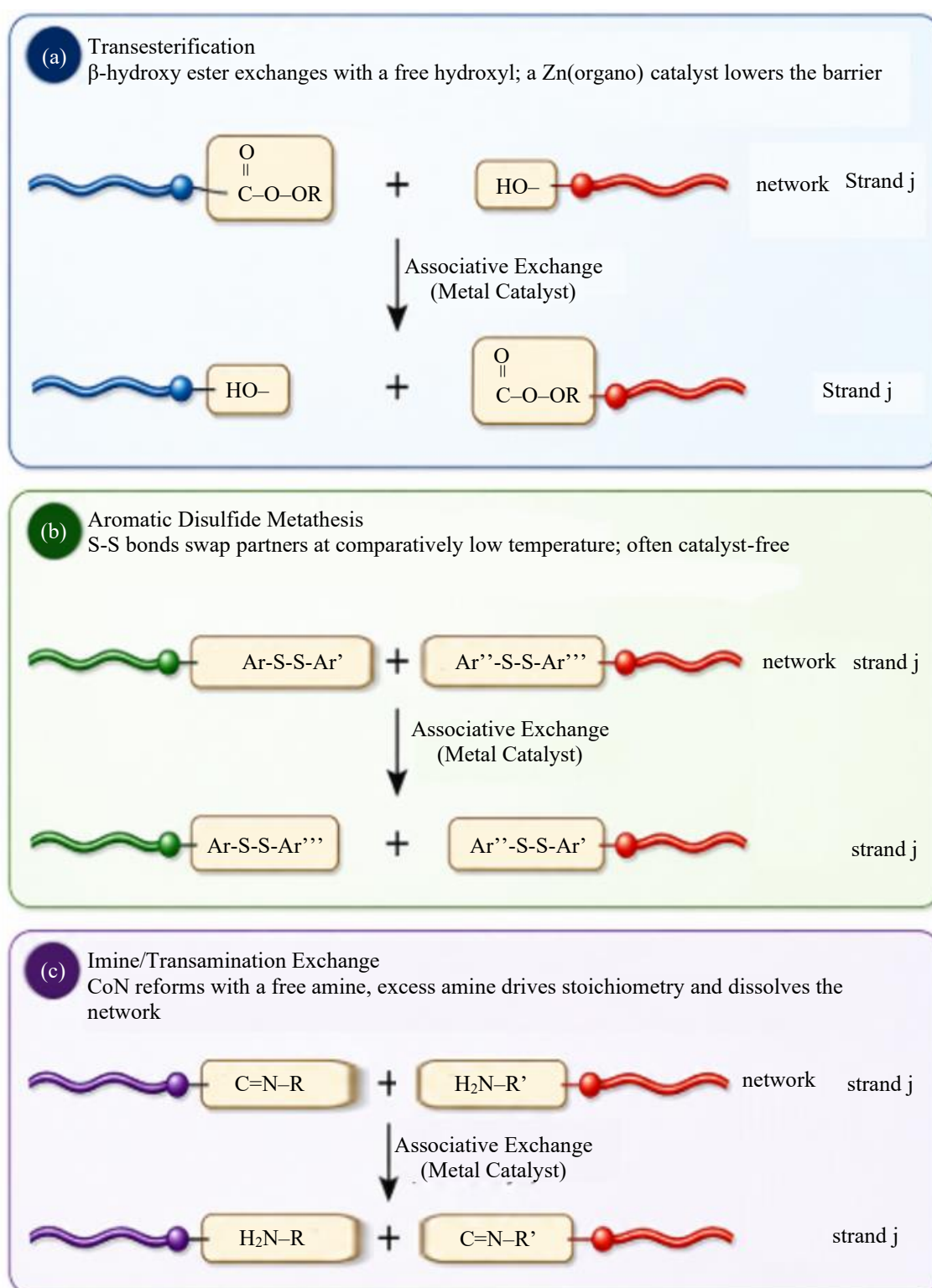


Figure 2. Associative bond-exchange mechanisms of the covalent adaptable networks relevant to aerostructures: (a) transesterification of β -hydroxyl esters, (b) aromatic disulfide metathesis, and (c) imine/transamination exchange. In each case a new bond forms, like the old one breaks, conserving network connectivity.

Imine and Polyimine Networks

The advantage of imine chemistry over other is its ability to dissolve under very mild conditions; this is due to an excessive amount of amine disrupting the stoichiometric relationship between the aldehyde and the amine causing it to break down into individual polymers. Carbon fiber reinforced polyimines were dissolved using a 20/80 Diethylenetriamine (DETA)/xylene solution at 80°C; Scanning Electron Microscopy (SEM) images confirmed the removal of the resin matrix as well as good bonding to the fibers when the recovered carbon fibers were redeposited within new composites made from the original materials [14]. Secondary amine hardeners containing imines have also been shown to allow for chemical recycling of conventional Bisphenol A based epoxy composite materials [15].

Dual-Dynamic and Hybrid Networks

Two types of degradation mechanism can be combined into one type of composite material to separate the temperature at which repair becomes possible, from the temperature at which full depolymerization of the composite takes place. An example of this concept was a composite developed by combining bisphenol-A epoxy with acrylated epoxidized soybean oil as the matrix. This composite was cured with dithiodibenzoic acid and was catalyzed for transesterification. It provided an adequate tradeoff between mechanical properties (stiffness & flexibility) and recycled end-of-life use as well as being both cost competitive with and having similar thermal performance to some conventional thermoset resins – such as the Bisphenol F resin – used in carbon fiber reinforced polymers (CFRP) [16]. Other related bio-based transesterification composite materials have shown tensile strength and flexural strength values of 543 MPa and 414 MPa, respectively [17]. These are within the range of tensile strength and flexural strength values for commercially available petroleum derived carbon fiber reinforced polymers (CFRP), however these values degrade when placed into ethylene glycol with the surface morphology of the recovered fibers preserved. Although there may be potential interest in aerostructures based on the principles of this technology, no dual-dynamic system has ever undergone a testing campaign that could provide certification data.

Table 1 sets the four families against the criteria that actually gate an airframe application rather than against laboratory novelty and read this way no single chemistry is dominant. Transesterification inherits the deepest formulation base and the highest glass-transition temperatures but pays for exchange with a catalyst and with temperatures that crowd the onset of degradation. Aromatic disulfides exchange at the lowest temperature and underpin the only qualification-style campaign in the corpus, yet their reduced compression and shear traced to toughening veils rather than to the dynamic bond – a reminder that the matrix chemistry is not always the property-limiting element. Imine networks dissolve under the mildest conditions of any family, which makes them the strongest fiber-reclamation candidates, but their low glass-transition temperatures and hydrolysis sensitivity push them toward secondary rather than primary structure. Dual-dynamic hybrids are the only route that deliberately decouples a repair temperature from a depolymerization temperature, but none has yet been carried beyond coupon-scale feasibility.

Table 1. Comparative assessment of the dynamic network chemistries against aerospace-relevant criteria.

Chemistry family	Exchange mechanism/typical condition	Catalyst requirement	Principal strength	Principal limitation for primary structure
Transesterification (anhydride/acid-cured epoxy) [11, 12]	β -hydroxyl ester exchange; flow >200°C; glycolysis at 180°C	Usually metal catalyst (e.g., Zn)	Highest T, (up to ~86–120°C reported); mature epoxy base; full fiber + matrix recovery	Exchange window crowds degradation onset; β -hydroxy esters more hydrolysable.
Aromatic disulfide [8, 13]	S–S metathesis at comparatively low T; RTM-processable	Often catalyst-free	Only qualification-style hot/wet campaign to date; low-temperature exchange	Softer compression/ILSS (traced to toughening veils, not the bond).

Imine / polyimine [14, 15]	C=N transimination; dissolves at 80–90°C with excess amine	Catalyst-free	Mildest reclamation of any family; continuous-tow recovery feasible	Low T _g ; imine hydrolysis sensitivity limits hot/wet service.
Dual-dynamic / hybrid [16, 17]	Two exchange chemistries with separated repair/depolymerization temperatures	Mechanism-dependent	Decouples repair from full recycling; bio-based, cost-competitive variants	No certification-level testing; behavior beyond one cycle unknown.

REPROCESSING EFFICIENCY

Thermal Reconsolidation and Reshaping

The simplest re-processing technique involves heating a cured laminate to a temperature higher than the “topology-freezing” temperature with applied pressure, allowing for the re-arrangement of the molecular structure (network) of the polymer. Since fiber reinforcement remains intact, this method is essentially a consolidation issue as opposed to a chemical issue, and thus the overall effectiveness of this approach will depend upon whether the matrix can flow sufficiently to allow closure of the inter-laminar voids prior to the onset of thermally induced degradation. Process windows reported in the literature have been shown to be smaller than some of the industry’s hype would indicate. For example, in the commercial prepreg referenced above, pressure, temperature and time (dwell), were optimized using an orthogonal experimental design. However, there was no meaningful flow in the prepregs until they had reached temperatures greater than 200°C, which was significantly above the glass transition temperature of 86.2°C for the matrix resin [12]. This large difference between these two temperatures is why the re-heating process works; however, it is also why materials that exhibit large differences in their glass transition temperatures may be very difficult to work with (Figure 3).

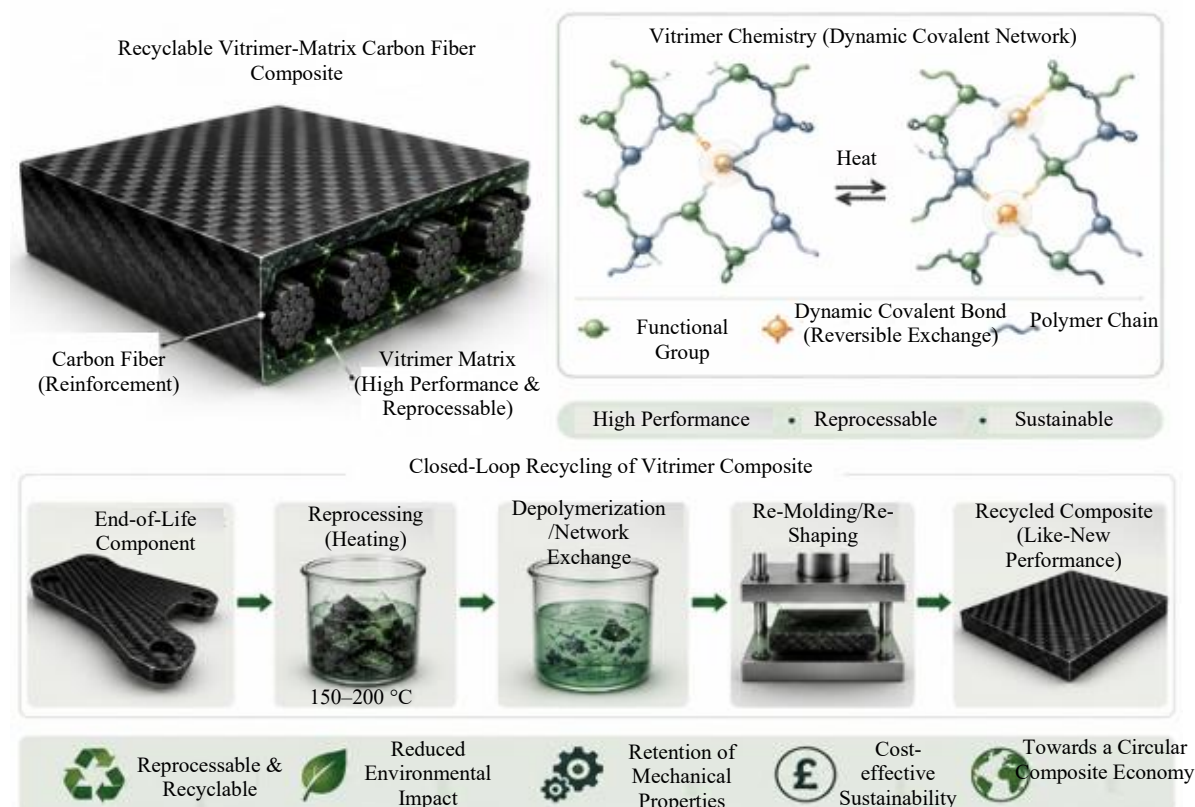


Figure 3. Recyclable vitrimer-matrix carbon-fiber composite and its recycling process.

Welding as a Reprocessing Route

Joining should be treated as a separate category from other vitrimer characteristics, in that it is likely to have the most direct application for an aerospace use environment. Resistance welding of vitrimer composites was done by simply placing a carbon-fiber heating element saturated with matrix between adherends and driving current through it. This produced single-lap joints in vitrimer composite materials with shear strength values of 12.0 ± 2.6 MPa (versus 8.4 ± 0.6 MPa values for samples consolidated using the manufacturer's recommended press cycle) [18]. In that the welded sample had a higher shear strength than the sample consolidated by the manufacturer's method, there should be some pause here; it suggests that localized, current-driven heating provides improved local rearrangement of networks than does a bulk press process, likely due to the fact that heat is being supplied to areas of the joint where bond rearrangement is required. This study also demonstrated that repeated application of resistance welding resulted in increased shear strength of the joints and that they could support at least five complete welding cycles.

Solvent-Assisted Dissolution and Fiber Reclamation

If the goal is to recover fibers as opposed to repairing individual parts, then the polymer matrix will be depolymerized in a reagent (solvent) that takes part in an exchange reaction. There are three factors that affect how efficient this method will be:

- Conditions under which the method occurs;
- Extent to which all the matrix material has been removed;
- Whether or not the fiber architecture remains intact.

The first two areas have provided good results. Polyether sulfone modified epoxy vitrimer matrices can be degraded using ethylene glycol at 180°C resulting in the complete separation and recovery of both the matrix and reinforcement components [12]; Diethylene triamine – xylene solutions dissolve polyimine matrices at 80°C and have also been scaled up for use on larger structure [14]; Cystamine has been shown to enable solvent free disulfide depolymerization of vitrimers such that the oligomer products generated can be cross-linked again [8].

It is here that vitrimers show one of the largest differences when compared to conventional methods of recycling. Tubes fabricated using a polyimine based vitrimer were subjected to dissolution at 90°C in a mildly acidic aqueous solution. The continuous tows produced by the dissolved tubes were then rewound into a second-generation tube [7]. Since filament winding never separates the fiber from its surrounding matrix it was possible to maintain the continuous tow configuration throughout the process. No other pyrolysis or solvolysis route can provide this level of continuity. A comparison of this route to alternative routes was made by the authors. A steam pressurized decomposition route resulted in retention of 94% of original fiber strength but utilized temperatures of approximately 400°C [7, 19]; Pyrolysis used for recovery from hydrogen tanks produced segments of 0.5–0.8 m which could then be down-cycled. The energy requirements for these processes are significantly different. The energy required to recover fibers is quoted as 2.03 MJ/kg whereas virgin production requires approximately 200 MJ/kg [7].

Plotted on a logarithmic axis (Figure 4), the contrast is stark: solvent-assisted reclamation of a vitrimer matrix is reported roughly two orders of magnitude below virgin-fiber production and well below thermal recovery routes. The caveat the primary sources rarely state is that these are process-energy estimates obtained at coupon scale under favorable conditions; they exclude solvent manufacture, recovery and reconditioning, and they have not been reconciled against a common system boundary. The environmental case is therefore directionally strong but not yet auditable, which is itself one of the clearer research gaps identified below.

Handling and Storage as a Hidden Efficiency Term

In addition to retention considerations with respect to manufacturing efficiency, an equally important consideration regarding process efficiency is what occurs after a product is manufactured, i.e., during delivery to the customer. The conventional aerospace prepreg is a refrigerated material and also has an

expiration date. The refrigeration requirement (cold chain) is a significant economic burden. Schiff-base vitrimer prepreps that were stored at ambient temperature for three months experienced loss of tensile strength and elastic modulus (versus fresh) of only 1.8% and 0.9%, respectively. This was due to the ability of the reversible dynamic bonds in the resin to keep the resin in a “stable” semi-cured condition until the high-temperature compression molding step completes the formation of the final network structure [19, 20]. A similar advantage for B-staged vitrimer films consolidated into organosheets as preform sheets has been noted. These organosheet preforms can be stored at room temperature and then laminated [21]. While removal of refrigeration from the supply chain does not represent a mechanical property improvement; this represents a true increase in processing efficiency and may ultimately become economically compelling.

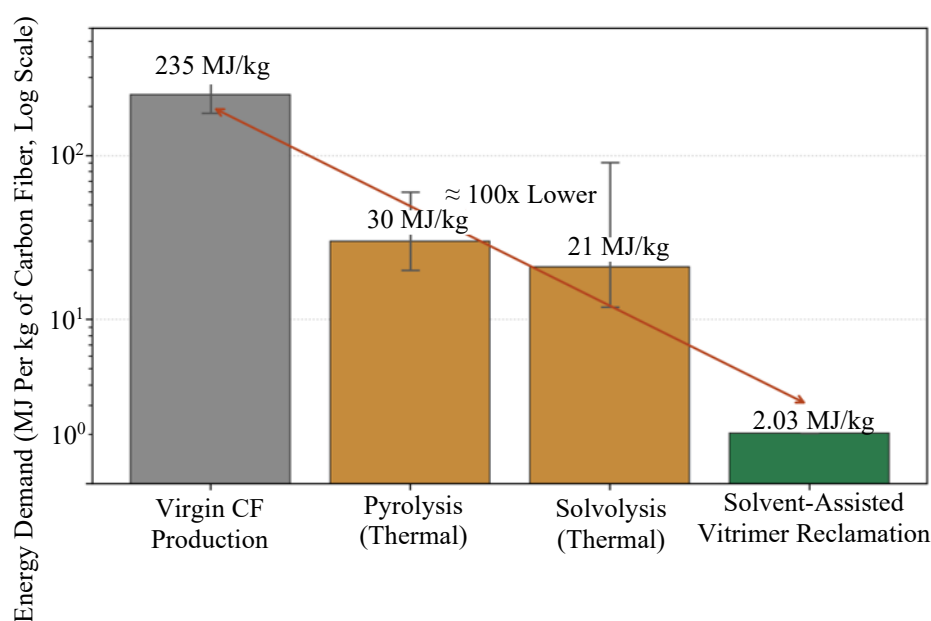


Figure 4. Reported energy demand of carbon-fiber recovery routes compared with virgin-fiber production, on a logarithmic scale. Solvent-assisted reclamation of a vitrimer matrix (2.03 MJ/kg) is roughly two orders of magnitude below virgin production (183–286 MJ/kg).

RETENTION OF MECHANICAL PERFORMANCE

Matrix-Dominated Properties

The two main properties to be affected by re-manufacturing are inter-laminar (interlayer) and short beam shear (SBS). This is since both are directly dependent upon the matrix and fiber/matrix interface as opposed to the fibers themselves. As such the losses have been shown to be very minor; in the case of the filament wound study there was an initial drop in SBS from the re-manufactured specimen compared to its original counterpart, however, this was attributed to process rather than chemical differences. The recycled tows were re-processed wetly (as no pre-pregging facility was available), resulting in higher resin contents and greater than 1 mm² sized resin voids/pockets within the laminate; these would act as sites for stress concentrations. When normalized for fiber volume fraction, the second-generation material maintained 94% of the original strength of the first generation [7]. Reprocessed polyimine composites demonstrated a moderate reduction in interlamination adhesion strength. A possible reason for this is likely to be due to the presence of residual solvents, as well as a loss in glass transition temperature for the nonwoven samples and an increase for the unidirectional samples [14].

Fiber-Dominated Properties

Longitudinal tension has a completely different viewpoint based on how the load is applied. It is hard to see why this would occur when you consider the load path. Provided that the filament does not suffer any damage during the recovery process and the interface is sufficient then tensile strength will remain

relatively unchanged for fibers loaded along their longitudinal axis. There were no statistical differences noted (though there was greater variability) between single-filament tests conducted using tow filaments recovered from a dissolved vitrimer matrix than those produced as virgin fiber [7]. Composites made via bio-based transesterification demonstrated both tensile strengths at 543 MPa and flexural strengths at 414 MPa yet remained biodegradable, with retained fiber morphology and surface chemistry post-recovery [17]. The disulfide systems developed for aerospace applications performed similarly to existing commercial RTM products throughout the entire test suite, including hot/wet [13].

Repair and Healing Efficiency

Between no repair (full re-processing) and repairing material lies the potential for “repair” in Vitrimers. It appears that vitrimers have a high value to cost ratio when it comes to damage or defect repair; specifically, inter-layer cracking within an industrial grade carbon-fiber laminate was repaired through localized heat application and demonstrated both recovery of original shape as well as repair capabilities [12]. The same research also demonstrated catalyst concentration could be used to dictate trade-offs between rapidity of chemical exchange and potential for increased creep susceptibility due to increased rate of chemical exchange for improved crack closing ability [11]. More recently, another study examined high glass transition temperature disulfide matrix laminates and developed a framework for determining safe window boundaries for repairs based upon thermal degradation rates and rates of chemical exchange at bonds, thereby providing a means to quantify those combinations of time and temperature for which healing processes will occur prior to further degradation of the network [22]. That type of framework is exactly what a certification authority wants, i.e., not a demonstration that repair is feasible, but rather a certified window for performing repairs.

Behavior Across Multiple Cycles

Most single cycle tests are well represented within literature. The biggest evidence-based gap at present is to understand what happens with multiple cycle testing. There are examples where weld joints demonstrate improvements after several repetitions of welding and were able to survive five cycles [18], and fatigue damage in carbon-fiber vitrimer laminates has been reversed repeatedly by thermal healing [23]. Conversely there are many examples of neat vitrimer resins being subjected to several remolding operations which lead to loss of mechanical properties due to changes in the network structure and accumulation of damage from thermal oxidation. To date very little has been published on a continuous fiber composite undergoing three or more full processing and test cycles under aerospace conditions. Therefore, all claims regarding service life resulting from the ability to reprocess material must be made by using some form of extrapolation (Table 2 and Figure 5).

Table 2. Reported reprocessing routes and performance retention for vitrimer-matrix fiber composites.

Matrix chemistry	Reprocessing route	Key conditions	Reported outcome
Polyimine (filament wound CF tube) [7]	Acidic aqueous dissolution and rewinding	90°C, pH 1.9, 2 h for tow recovery	94% of first-generation short-beam shear strength after normalization to fiber volume fraction; single-filament strength statistically unchanged.
PES-modified epoxy, transesterification [12]	Hot compression reprocessing; glycolysis	Flow above 200°C; matrix degradation in ethylene glycol at 180°C	T _g 86.2°C, storage modulus 16 GPa, E _r 65.43 kJ mol ⁻¹ ; full recovery of fiber and matrix; interlayer cracks repaired.
DGEBA-glutaric anhydride, Zn catalyzed [11]	Thermal reprocessing and interlaminar repair	Catalyst loading tunes exchange kinetics	Relaxation activation energy up to ~60 kJ mol ⁻¹ ; reprocessability and repair scale with catalyst content.
Polyimine, non-woven and UD CF [14]	Solvent dissolution and re-lamination	DETA-xylene 20:80 at 80°C	Effective matrix removal with minimal residue; interlaminar adhesion slightly reduced; T _g shift attributed to residual solvent.
Vanillin-derived Schiff base [20]	Closed-loop recovery; ambient prepreg storage	High-temperature compression molding completes cure	Matrix and fiber both reused; after three months of ambient storage, prepreg lost 1.8% tensile strength and 0.9% modulus.

Epoxy vitrimer, CF adherends [18]	Resistance welding of single-lap joints	Current driven through CF heating element	Weld shear strength 12.0 ± 2.6 MPa vs 8.4 ± 0.6 MPa for press consolidation; up to five welding cycles sustained.
Epoxy-aromatic disulfide, aeronautical grade [13]	RTM manufacture; full qualification-style campaign	Room temperature, 70°C and 120°C after moisture saturation	Comparable to incumbent aeronautical RTM systems; lower compression and ILSS attributed to thermoplastic toughening veils.
Bio-based transesterification epoxy [17]	Chemical degradation and fiber reclamation	Ethylene glycol solvolysis	CFRP tensile 543 MPa and flexural 414 MPa; recovered fiber surface morphology and chemistry preserved.
High-Tg disulfide GFRP [22]	Thermal repair of matrix-dominated damage	Time-temperature-pressure envelopes derived from degradation kinetics	Framework proposed for defining safe repair windows and quantifying degradation limits in structural composites.

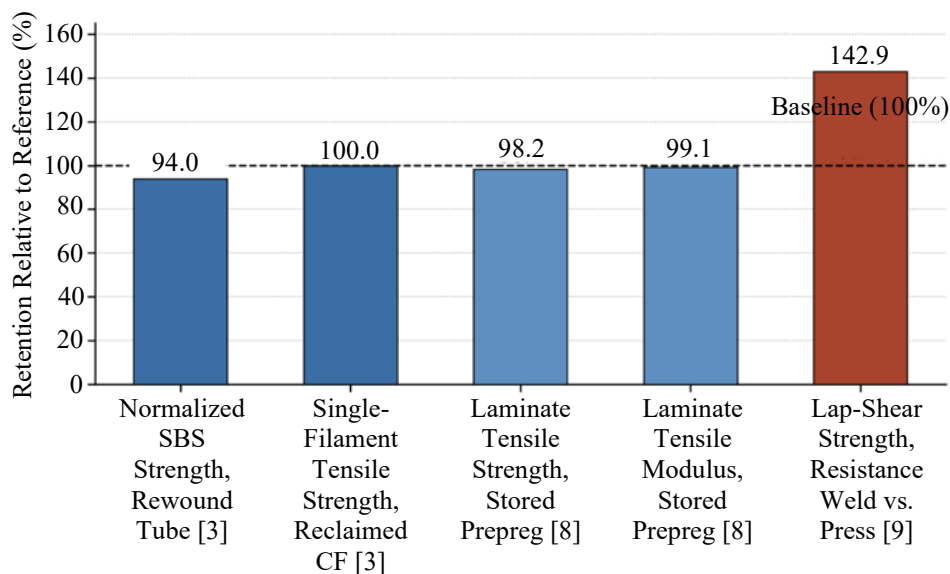


Figure 5. Reported property retention for vitrimer-matrix composites after one reprocessing or recycling operation. Note: Values compiled from the primary sources indicated. The single-filament figure denotes no statistically significant difference from virgin fiber rather than an exact measured ratio.

There are two aspects that stand out in Figure 5. First, fiber dominated measurements cluster around (or exactly) one. This confirms that very mild dissolution chemistries do not degrade the carbon backbone. Second, the weldability measurement exceeds the base line entirely. This is a reminder that reprocessing does not always result in degradation of the material being processed. Where the process results in improved consolidation quality, it will also likely provide improvement of the property being measured. In addition to the two obvious points made by the lack of data presented above, the scarcity of the amount of similar data for other properties required by an aerostructure allowables document is itself indicative.

CRITICAL COMPARATIVE ASSESSMENT

The preceding sections catalogue what has been demonstrated; this section weighs those demonstrations against one another and against the requirements they must ultimately meet. Three cross-cutting observations follow from reading the corpus as a whole rather than study by study.

First, most reported strength losses are attributable to processing rather than to chemistry. The most complete data point – the filament-wound polyimide tube – lost short-beam shear strength only until fiber volume fraction was normalized, after which recovery reached 94% and the residual gap mapped onto wet-rewinding voids larger than one square millimeter rather than onto the dissolution step [7]. The same pattern recurs in the aeronautical disulfide campaign, where the softer compression and interlaminar-shear numbers followed the thermoplastic toughening veils, not the exchangeable bonds [13]. The implication is uncomfortable for the field's own headline claims: a comparison of first-generation prepreg parts with second-generation wet-wound parts measures the change in

manufacturing route, not the recyclability of the matrix. Until retention is reported at matched fiber volume fraction and void content, cross-study comparison of absolute strengths is not meaningful.

Second, the corpus divides cleanly along a load-path line that the reviewed studies rarely draw explicitly. Fiber-dominated properties – longitudinal tension and single-filament strength – are essentially conserved because mild dissolution does not touch the carbon backbone; the reviewed single-filament data show no statistically significant difference from virgin fiber, only greater scatter [7, 17]. Matrix- and interface-dominated properties – interlaminar and short-beam shear – carry almost all the risk, because they depend on the very bonds being reshuffled and on any solvent residue or glass-transition shift left behind [14]. This split is the most useful organizing principle the literature offers, yet it is obscured whenever a study reports a single “retention” figure without stating which property class it belongs to.

Third, the three reprocessing routes are not competing options but a hierarchy of increasing intervention, and their evidence bases are markedly uneven. Thermal reconsolidation is the least invasive but is boxed in by a narrow window between matrix flow and degradation. Welding is the only route that has repeatedly produced a property improvement rather than a loss – resistance-welded joints exceeded press-consolidated ones and survived at least five cycles [18] – and it carries the clearest near-term airframe use case, yet it attracts the least systematic mechanical characterization. Solvent-assisted reclamation is the most transformative route and the best evidenced for fiber recovery, but almost all its data are single-cycle. The field has, in effect, optimized the demonstration nearest to publication novelty rather than the one nearest to certification.

The through-line of these observations is a single dominant gap. Almost the entire quantitative corpus rests on one reprocessing cycle. Continuous-fiber laminates carried through three or more full process-and-test cycles under representative aerospace conditions are essentially absent, so every service-life claim founded on reprocessability is presently an extrapolation from single-cycle coupons and from neat-resin remolding studies that already show property drift with cycle count. Figure 6 summarizes how the four families compare across the criteria that matter for adoption; the shape of

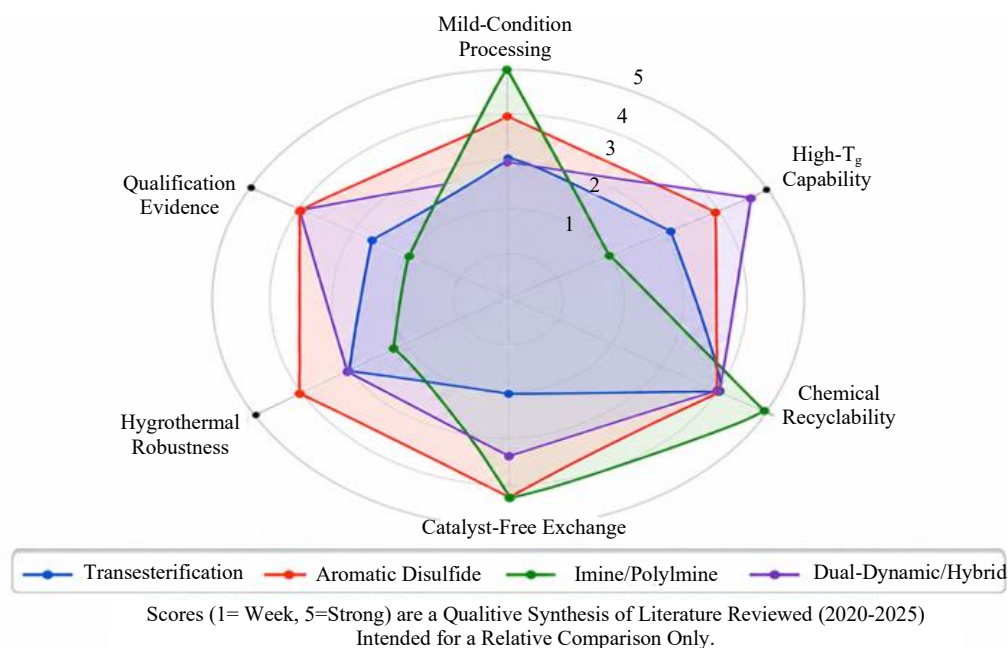


Figure 6. Radar comparison of the four vitrimer chemistry families across six aerospace-relevant criteria. Each profile summarizes the qualitative balance of strengths and weaknesses drawn from the reviewed literature; no family dominates all axes.

Note: Scores (1–5) are a qualitative synthesis of the 2021–2026 corpus intended for relative comparison only, not absolute measurement.

each profile, more than any single axis, shows why the choice of chemistry is application-specific rather than absolute.

Taken together, the evidence supports a measured conclusion: the chemistry works, the fiber survives, and the matrix-dominated penalty is small once processing is controlled for – but the data are neither deep enough nor standardized enough to distinguish a genuinely durable material from a well-executed single-cycle demonstration.

BARRIERS TO AEROSTRUCTURE ADOPTION

The Glass Transition and Reprocessability Trade-off

The main issue of material science in aerospace is structural, as opposed to incidental. The glass transition temperature for aerospace composite matrices should be sufficiently higher than the highest operating temperature of the system (with a typical margin of about 28°C) and hot-wet performance should be determined after saturation with water [2, 13]. Only a few of the vitrimer matrices used in composites have T_g values greater than 120°C. However, this allows them to perform well in secondary structures and interior applications; however, they will not be suitable for use in primary structure components that require to operate at or near the exterior surface of an aircraft. Another constraint on the formulations that can achieve the necessary range of T_g values is that the crosslinking process must occur above the glass transition temperature of the matrix. Therefore, the lower the T_g value of the matrix, the closer to the onset of thermolysis degradation the matrix must be reprocessed. Additionally, if the time available to complete the crosslinking reaction does not allow sufficient time for full rearrangement of all polymer segments within the network, then chemical side reactions can initiate prior to completion of the desired crosslink density [1, 22]. Figure 7 makes that squeeze explicit. The solvent-assisted reclamation routes reported to date operate at 80–90°C, comfortably below the glass transition demanded of an aerospace primary structure, whereas the purely thermal routes require 180–200°C, which is at or above the temperature at which degradation of the same matrix has been reported.

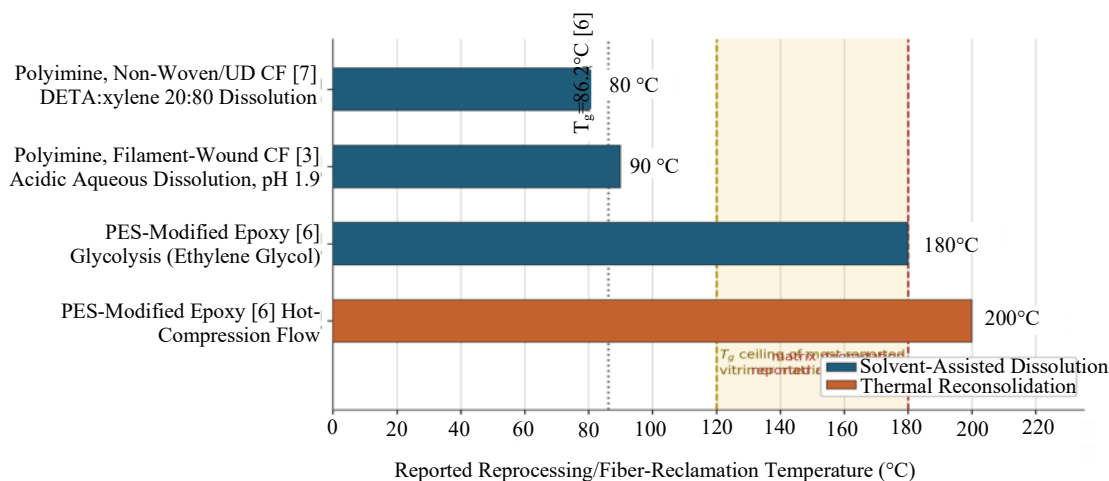


Figure 7. Reported reprocessing and fiber-reclamation temperatures for the vitrimer-matrix systems compiled in Table 2, shown against the glass transition ceiling of most reported vitrimer matrices and the matrix degradation temperature reported for the transesterification system.

Note: Temperatures are those stated in the primary sources compiled in Table 2. The bars indicate the thermal conditions under which each route was demonstrated, not a measure of performance.

Creep and Long-Term Dimensional Stability

It is the exact same property that causes a vitrimer to be recyclable that also allows a vitrimer to deform. The dynamic bonds within a vitrimer allow for continued and slow relief of stress even at temperatures significantly below (and often far below) the topology-freezing temperature and many

researchers have found a significant amount of creep behavior in vitrimers compared to their commercial counterparts at temperatures as low as those near the topology-freezing temperature [1]. Creep behavior over tens of thousands of flight hours for an airframe component held under sustained loads would not be negligible. Some mitigation strategies are beginning to emerge with the obvious one being the inclusion of some permanent crosslinking along with exchangeable crosslinks that have demonstrated they can raise the extrapolated topology-freezing temperature by reducing creep behavior at service temperatures up to approximately 150°C [24] but will necessarily slow down bond exchange rates and thus limit recycling efficiency. No experimental data has yet addressed whether there may exist some acceptable trade-off at the level of structural loading.

Hygrothermal Durability

The β -hydroxy ester linkages formed during cure of anhydrides on epoxies are more hydrophilic than their fully reacted counterparts. Further, esters have been shown to be hydrolysable. Carbon-epoxy laminates already suffer loss of interlaminar shear strength through plasticization and degradation of the interphase when exposed to hygrothermally sustained conditions. As such, accelerated aging studies remain a fundamental basis for storage-life predictions in the aerospace supply chain [25] (vitrimers introduce an additional concern that hydrolysis compete with exchange chemistry itself. Thus, aging may not only alter the strength of vitrimers but also their ability to reprocess later in life.)

Manufacturing Compatibility

Automated fiber placement, Automated tape laying, Autoclave curing, Resin Transfer Molding (RTM) and a matrix which cannot be placed on these machines will not get selected for use even though it may meet the end-of-life needs. There has certainly been some progress made. The formulation of vitrimers have shown they can be processed using RTM [13, 26] and by vacuum-assisted resin infusion [27], or via prepregs in an industrial setting [12]; they have also shown they can be processed by filament winding either in their wet or towpreg forms [7], and finally through B-staged film consolidation into a storable sheet format [21]. However, compatibility with automated fiber placement has yet to be shown – but only identified as an open research area and not demonstrated capability [2]. Therefore, the tack, drape, out life and deposition rate of the vitrimer formulations being deposited under the head of the machine is different than what the formulations have been optimized against.

Absence of a Qualification Pathway

Even if a material meets all the criteria for aerospace material, there will still be an issue with the process. The basic premise of aerospace qualification is based on a material which has constant properties after curing. A reprocessable matrix cannot meet this requirement; therefore, any welding, repairs, etc., done during use could result in properties that are different than when they were first qualified. To date, no consensus or acceptable method for assessing whether or how a material's state can be altered by past or future process history (and/or how one would inspect a material to verify that a previous weld was satisfactory) has been identified. It is also recognized that establishing industry standards for testing procedures is likely to be a critical step toward gaining acceptance of these new types of matrices and that it may be the most difficult challenge related to development of the necessary new technologies from a materials perspective [2, 10].

Table 3 consolidates these barriers, separating those that are fundamentally chemical from those that are, at root, a shortage of characterization. The distinction matters for prioritization: a formulation problem needs new molecules, whereas a characterization problem needs disciplined, repeated measurement – and most of the entries below fall into the second category, which is why the recommendations that follow emphasize measurement over synthesis.

Table 3. Critical assessment of the principal barriers to aerostructure adoption.

Barrier	Underlying cause	Nature of the problem	Priority action/severity
T ₉ vs. reprocessability trade-off	Exchange must occur above T ₉ , so low-T ₉ matrices reprocess close to their degradation onset	Chemical + process	High for primary structure; raise usable T ₉ without closing the process window.
Creep / long-term dimensional stability	The dynamic bonds that enable recycling also permit slow stress relaxation under load	Chemical (intrinsic)	High; needs laminate-level creep data at service stress ratios, not neat resin.
Hygrothermal durability	Ester/imine linkages are hydrolysable; hydrolysis competes with exchange chemistry	Chemical	High; hot/wet ageing of reprocessed (not only virgin) laminates required.
Manufacturing compatibility	Tack, drape, out-life and deposition rate unproven for automated fiber placement	Characterization	Moderate; RTM/VARI/prepreg shown, AFP still an open demonstration.
Multi-cycle durability	Almost all laminate data are single-cycle; property drift seen in neat-resin remolding	Characterization	Highest / most tractable; ≥3 full process–test cycles on long-fiber coupons.
Absence of a qualification pathway	Qualification assumes fixed post-cure properties; a reprocessable matrix has process history	Standards / regulatory	High; inspection and re-certification framework for altered material state.

RECOMMENDATIONS FOR FUTURE WORK

There are four key priorities because of the previous assessment. First, there needs to be multi cycle lamina test data (at least three full cycles of processing and testing) using long fiber coupons. This will include data regarding interlaminar shear; in plane shear; Mode I and Mode II fracture toughness of the coupon; along with fiber volume fractions and void contents determined at each stage of testing. Additionally, this will provide an accurate representation of how well the lamina performs throughout its life span.

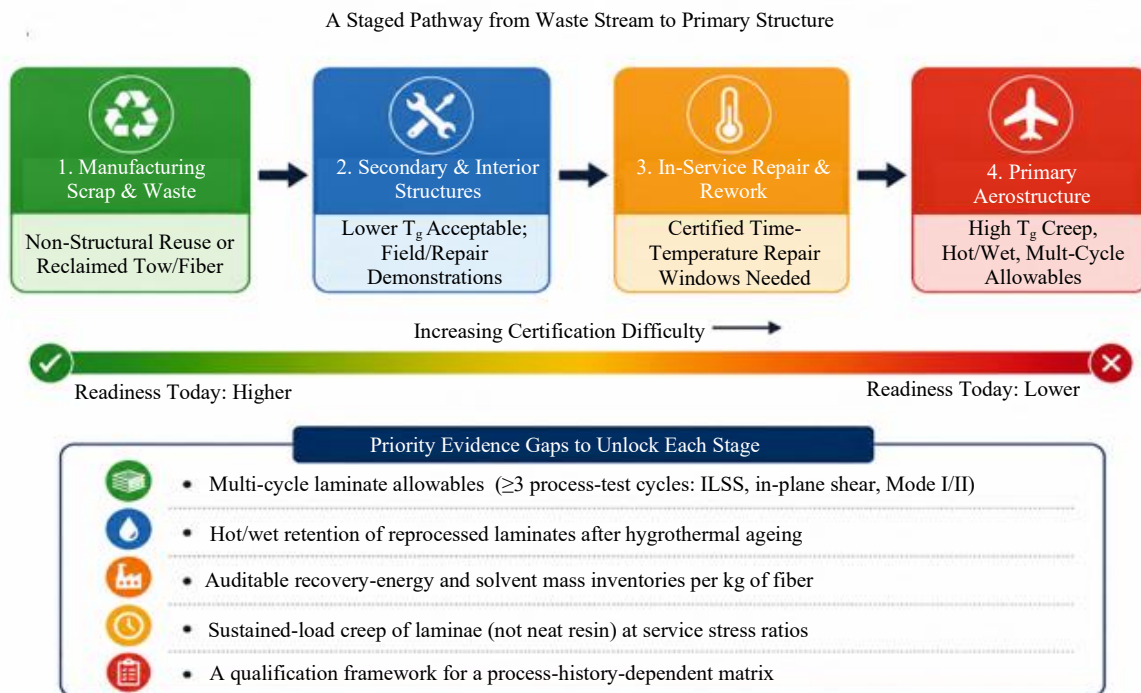


Figure 8. A staged adoption pathway for vitrimer-matrix composites, from reuse of manufacturing waste through secondary structure and certified in-service repair to primary aerostructure, with the priority evidence gaps that must be closed to unlock each stage.

Secondly, the purpose of reprocessing should be assessed by determining if it influences the ability of the lamina to withstand hygrothermal effects. As stated previously, virgin materials have been tested under hygrothermal conditions. It would be beneficial to test the reprocessed material to determine if reprocessing negatively affects performance. Third, consistent measurements need to be taken regarding the amount of energy used during the recycling process. Specifically, measurements should be made regarding the amount of energy required to recycle one kg of fiber. The same measurement should also be taken to measure the mass of solvents removed per kilogram of matrix. With these numbers, companies can make environmental claims that can be audited instead of just being asserted. Fourth, sustained load creep testing should be conducted using representative stress ratios and service temperature on laminas rather than neat resins. When a lamina is subjected to sustained loads, the presence of fibers will constrain the creep behavior of the matrix and will affect the way it behaves when subjected to continued loading.

These priorities map onto a staged adoption pathway rather than a single leap to primary structure (Figure 8). The lowest-burden entry points – reuse of manufacturing waste, then secondary and interior parts, then certified in-service repair – can accumulate the multi-cycle and hot/wet evidence base that a wingbox application will eventually demand, while deferring the barriers that remain genuinely unsolved.

CONCLUSIONS

There is some legitimate hope based upon the body of evidence presented above. Carbon fiber-reinforced polymer (CFRP) laminates made of vitrimer matrices can be resintered or weld bonded or repaired or even solubilized using techniques and temperatures that are mild enough to preserve the integrity of the fibers, allowing continuous tows to be reclaimed and rewound. Fiber dominated properties have been retained when sufficient rigor was applied to measure retention; and matrix dominated properties have generally returned to approximately 94% of their original values after accounting for fiber volume fraction. Welding processes – such as resistance welding – have resulted in joints that were significantly stronger than those obtained by other methods of consolidation. Elimination of the need for ambient temperature prepreg storage of materials also represents a reduction in cost within the supply chain.

There are, however, significant barriers remaining to further development of this technology. These are located directly in areas where aerostructures are most demanding. The relationship between T_g and RP temperature reduces the process window to an area where the T_g 's of typical aerospace formulations is too low. In addition, creep resistance and recyclability rely on mutually exclusive features of the network, and there does not exist any scientifically proven and valid compromise at aerospace loads. Durability over multiple thermal cycles of reworked material has received little attention. Data on multi cycle behavior of laminates is limited, and currently no formal qualification framework exists for evaluating a material whose mechanical properties depend on its processing history.

Although none of these may appear insurmountable, many represent efforts in characterization, versus problems of chemical formulation. Therefore, it appears that the greatest benefit to advancing this area would be a shift in emphasis. This would involve fewer demonstrations of feasibility of reprocessing, and more measurements of how much each demonstration costs to make. This should be done frequently enough and in a consistent manner to establish the statistical basis necessary to obtain certification. Vitrimer composites will likely begin entering aerospace manufacturing via secondary parts or manufacturing waste before they appear in a wingbox; therefore, this appears to be the logical way to introduce them.

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