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► **To cite this version:**

Sudhanshu Nartam, Vishal Rautela, Sandip Budhe, Jinu Paul, Silvio de Barros. Strength Retention of Carbon Fiber/Epoxy Vitrimer Composite Material for Primary Structures: Towards Recyclable and Reusable Carbon Fiber Composites. *Applied Mechanics*, 2024, 5 (4), pp.804-817. 10.3390/applmech5040045. hal-04783539

HAL Id: hal-04783539

<https://hal.science/hal-04783539v1>

Submitted on 14 Nov 2024

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Article

Strength Retention of Carbon Fiber/Epoxy Vitrimer Composite Material for Primary Structures: Towards Recyclable and Reusable Carbon Fiber Composites [†]

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[†] This paper is an extended version of our paper published in Nartam S, Budhe S, Paul J. Carbon Fiber/Epoxy Vitrimer Composite Material for Pressure Vessels: Towards Development of Sustainable Materials. In Materials Science Forum 2024; Volume 1123, pp. 89–93. <https://doi.org/10.4028/p-dzac8g>.

Abstract: Recently, the growth of the recyclability of carbon fiber reinforced polymer (CFRP) composites has been driven by environmental and circular economic aspects. The main aim of this research work is to investigate the strength retention of a bio-based vitrimer composite reinforced with carbon fibers, which offers both recyclability and material reusability. The composite formulation consisted of an epoxy resin composed of diglycidyl ether of bioshpenol A (DGEBA) combined with tricarboxylic acid (citric acid, CA) and cardanol, which was then reinforced with carbon fibers to enhance its performance. Differential scanning calorimetry (DSC) and Fourier transform infrared (FTIR) spectroscopy were performed to analyze the chemical composition and curing behavior of the vitrimer. Mechanical testing under tensile loading at room temperature was carried out on epoxy, vitrimer, and associated carbon fiber reinforced composite materials. The results demonstrated that the DGEBA/CA/cardanol vitrimer exhibited thermomechanical properties comparable to those of an epoxy cured with petroleum-based curing agents. It was observed that the maximum tensile strength of vitrimer is about 50 MPa, which is very close to the range of epoxy resins cured with petroleum-based curing agents. Notably, the ability of the vitrimer composite to be effectively dissolved in a dimethylformamide (DMF) solvent is a significant advantage, as it enables the recovery of the fibers. The recovered carbon fiber retained comparable tensile strength to that of the fresh carbon composites. More than 95% strength was retained after the first recovery, which confirms the use of fibers for primary and secondary applications. These research results open up new avenues for efficient recycling and contribute to the overall sustainability of the composite material at an economic level.

Keywords: recyclability; vitrimers; reusability; carbon fiber; sustainability



Citation: Nartam, S.; Rautela, V.; Budhe, S.; Paul, J.; de Barros, S. Strength Retention of Carbon Fiber/Epoxy Vitrimer Composite Material for Primary Structures: Towards Recyclable and Reusable Carbon Fiber Composites. *Appl. Mech.* **2024**, *5*, 804–817. <https://doi.org/10.3390/applmech5040045>

Received: 9 October 2024

Revised: 28 October 2024

Accepted: 29 October 2024

Published: 6 November 2024



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1. Introduction

The use of fiber-reinforced composite materials has been continuously growing for a few decades, and they are currently used in all sectors due to their peculiar properties. The extensive use of composite materials across various industries such as aerospace, automobiles, and sports has raised serious environmental concerns due to challenges associated with recycling and reusability. Carbon fiber reinforced polymer (CFRP) composites based on epoxy are often selected among the different CFRP composites due to its high performance, which is needed for high-end applications such as aircraft components, wind blades, etc. [1–6]. A large quantity of retired composites is generated due to vast application of

CFRP in all sectors for many decades. The majority of composite materials are dominated by synthetic fiber such as carbon, glass, aramid, etc., and petroleum-based adhesive. However, both fiber and petroleum-based adhesives are nondegradable, which has a negative impact on the environment and results in a very low recycling value for such high-value materials. In recent decades, many scientists have worked on recyclability and explored on different methods such as mechanical recycling, thermal process, solvolysis, etc. However, these methods are not only costly but also do not retain the significant mechanical and physical properties of recycled fiber and hence reduce the value of materials. This has led to an urgent need to recycle CFRP waste, driven by both climate change and economic considerations, to recover the fiber effectively with retained properties [7–10].

Conventional recycling methods are not very effective in terms of cost and the retained properties of recycled materials. Numerous researchers are actively pursuing the development of alternative resins to epoxy that can resemble the mechanical properties and, importantly, are recyclable, enabling their reuse in other applications, as the recycling of resin is almost impossible due to an insoluble spatial network. Recent advancements in polymer chemistry have made it feasible to recycle thermosetting polymers due to dynamic bonds that can react reversibly [11–17]. Research work has been carried out on different dynamic bonds, such as ester bonds, imine bonds, acylhydrazone bonds, etc. for the easy depolymerization of thermosetting resin, especially epoxy. A new type of crosslinked polymer, commonly known as vitrimer, was developed by Ludwik Leibler and his team at the French National Center for Scientific Research (CNRS) in the early 2000s, addressing the issue of recyclability [12]. In recent years, vitrimers have emerged as novel polymeric materials because they enable reshaping and reprocessing multiple times upon exposure to external stimuli (heat and pressure). The primary motivation for creating vitrimer-based composites is to maximize the efficiency of fiber recycling and reuse, thereby supporting the circular economy. The recovered fibers can be utilized in the production of composites and various other applications in primary and secondary structures, depending on the retained strength and other properties [9,18].

Zhou et al. [19] synthesized a vitrimer system using an epoxy monomer with disulfide bonds and 4,4'-disulfanediyl dianiline (4-AFD). This vitrimer demonstrated impressive mechanical properties, including a high storage modulus of 2.2 GPa and a glass transition temperature above 130 °C. In addition, it was fully degraded in dimethylformamide (DMF) at 50 °C. Similarly, Yu et al. [8] developed vitrimer-based carbon fiber composites with ester bonds, where the resin matrix was degraded and dissolved using an ethylene glycol solution under heating, allowing for the non-destructive recovery of carbon fibers. However, disulfide- and acid-based vitrimer reactions typically require metal catalysts to facilitate dynamic bond exchanges. These catalysts raise environmental concerns, as they can leach harmful metals during degradation, potentially contaminating the ecosystem. In contrast, imine-based and catalyst-free bio-based vitrimers offer safer and more environmentally friendly alternatives, maintaining the dynamic properties needed for reprocessing and recycling while being more cost-effective and sustainable.

Narubeth et al. [20] studied a carbon fiber/epoxy vitrimer composite cured with a bio-based cross-linker, citric acid (CA), and cashew nutshell liquid (CNSL) to patch-repair metallic sheets. Wenbin Li et al. [21] synthesized a bio-based epoxy vitrimer using tung oil and a zinc catalyst. The vitrimer exhibited high thermal stability and tensile strength. It could be used to prepare CFRP composites with fully recyclable carbon fibers, using a mixture of ethanol and NaOH as the solvent. Jianrong Xia et al. [22] developed a fully bio-based vitrimer using saturated cardanol, vanillyl alcohol, and citric acid (CA). Vitrimers derived from cardanol demonstrate excellent network rearrangement properties and can be efficiently depolymerized with ethylene glycol, achieving complete degradation in 1 h at 180 °C. However, these examples highlight that limited research has been conducted on the synthesis and structural characterization of bio-based vitrimer composites and their recycling behaviors. While vitrimer resins offer the potential for developing recyclable

CFRP composites, the retained properties of the recycled composites still constrain their wider application.

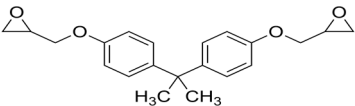
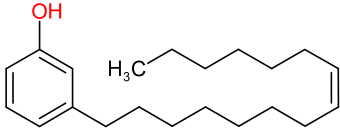
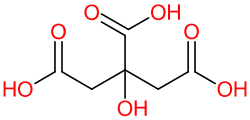
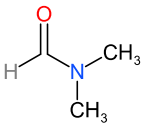
To ensure the reusability of recycled fibers, it is essential to study the retention properties of composite materials, especially fiber, as the properties change during the recycling process. In this research work, the vitrimer was prepared using DGEBA (Bisphenol A diglycidyl ether), citric acid, and cardanol as the primary components. The application of the cardanol component for the vitrimer preparation is due to renewable resources from natural cashew nutshell oil. The thermal degradation and curing behavior of the vitrimer were analyzed. Recycling was performed by dissolving vitrimer-based carbon-fiber composites in dimethylformamide (DMF) solvent. The mechanical properties of both the vitrimer-based carbon fiber composites and recycled composites were evaluated to assess the retention properties of the recycled fibers.

2. Experimental Details

2.1. Materials

The epoxy resin used in this study was Bisphenol A diglycidyl ether (DGEBA), which is known for its superior mechanical, adhesion, and thermal stability properties. A specific brand of DGEBA epoxy resin D.E.R 332 was purchased from Sigma Aldrich, Darmstadt, Allemagne, with an epoxy equivalent weight (EEW) of 174 g eq^{-1} . Cardanol obtained by vacuum distillation of the spongy mesocarp of cashew nutshell liquid (CNSL) was used as a curing agent and was purchased from Kumarasamy Industries, Cuddalore Tamilnadu, India. Citric acid monohydrate, a tricarboxylic acid with an equivalent weight of 70 g/eq , was also used as a curing agent for the epoxy resin. Dimethylformamide (DMF), an organic solvent capable of dissolving both the epoxy resin and curing agent, was used during the recycling process. The chemical structures of the components used in this research work are presented in Table 1. Plain-weave carbon fibers were chosen to reinforce the composite material.

Table 1. Chemical structure of the chemical components.

Name	Chemical Structure
Bisphenol A diglycidyl ether (DGEBA)	
Citric Acid	
Cardanol	
Dimethylformamide	

2.2. Specimen Fabrications

2.2.1. Vitrimer Preparation

The preparation of the vitrimer involved a complex sequence of steps, as illustrated in Figure 1. Initially, the equivalent ratios of the acid equivalent weight (AEW) to epoxy equivalent weight (EEW) were established. The procedure began with citric acid (CA)

dissolved in ethanol using a magnetic stirrer at room temperature for 30 min, yielding a white paste. Subsequently, cardanol (10 wt. % of DGEBA) was incorporated into the citric acid paste. This mixture underwent heating and stirring at 80 °C for 20 min. Simultaneously, the DGEBA was heated to decrease its viscosity before being added to the solution. The resulting blend was then subjected to heating and stirring for 40 min at 80 °C. The blending mixture was strained into an aluminium mold lined with a mylar sheet and subjected to a two-stage curing process in an oven. The first stage involved heating at 120 °C for 2 h, followed by the second stage at 160 °C for 4 h. The epoxide groups in DGEBA are likely to react with the carboxyl groups of citric acid, leading to esterification. Citric acid has multiple functional groups, and that can act as a trifunctional cross-linker. Each citric acid molecule can react with three DGEBA molecules, creating a network structure. The phenolic hydroxyl group in cardanol can react with the epoxide groups in the DGEBA to form ether bonds. Figure 2 shows the tensile specimen of a vitrimer polymer sample.

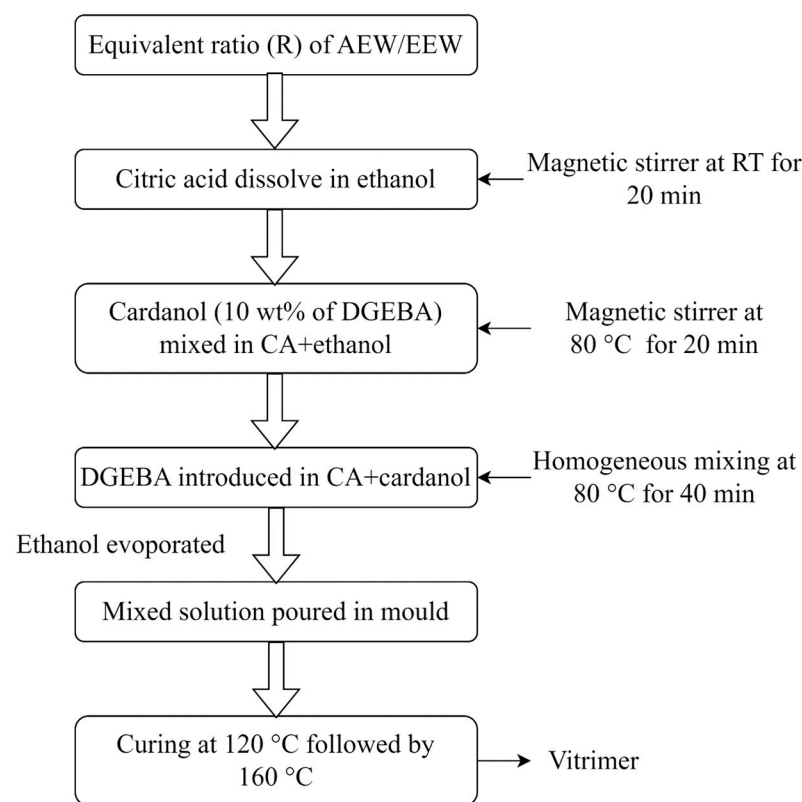


Figure 1. Step-by-step procedure for vitrimer.



Figure 2. DGEBA/CA/cardanol vitrimer polymer specimen.

2.2.2. Carbon Fiber–Vitrimer Composite

Two types of composites were prepared using the hand layup method: (1) carbon fiber-reinforced epoxy composites and (2) carbon fiber-reinforced vitrimer composite specimens. The curing cycle for the vitrimer-based composite was followed as previously mentioned in Section 2.2.1, while the epoxy-based composite material was cured at room temperature. This thermal treatment enables the chemical reactions within the matrix to progress, resulting in the cross-linking of the epoxy resin molecules and the formation of

a robust and durable composite structure. Figure 3 shows the tensile specimens of the composite materials.



Figure 3. (a) Epoxy CF composite and (b) vitrimer CF composite.

2.2.3. Recycling of Carbon Fiber

The recycling process of the vitrimer composite involved several steps for the effective recovery and reuse of the carbon fibers, thereby promoting sustainability and reducing waste. Once prepared, the vitrimer composite components underwent a dissolution process using dimethylformamide. The DMF was capable of dissolving the vitrimer matrix while leaving the carbon fibers intact. After carbon fiber separation, the composite specimen was again prepared using recycled carbon fiber to verify the retained mechanical properties.

2.3. Functional Group Characterisation

Fourier Transform Infrared Spectroscopy (FTIR-ATR)

A FTIR Spectrometer (JASCO-4700 LE, Tokyo, Japan) was used to identify the various functional groups of the materials. On an FTIR spectrometer, Fourier transformation infrared (FTIR) spectra were captured with a 4 cm^{-1} resolution. The sample was placed inside the apparatus, where an infrared detector measured the infrared rays that struck it. Peaks were generated in the Fourier transform infrared (FTIR) spectra depending on the frequency at which each bond vibrated. These peaks occurred when resonance occurred, or when the natural frequency of the bond vibration coincided with the frequency of the infrared rays emitted. The intensity of the peaks indicated the presence of various functional groups in the sample. The blends of FTIR spectra between 400 and 4400 cm^{-1} were measured.

2.4. Thermal Characterisation

2.4.1. Differential Scanning Calorimetry (DSC)

The method of differential scanning calorimetry (DSC) was employed to study the thermal transitions that occurred when the polymers were heated. DSC utilized independently controlled elements to heat the sample and is referenced in different manners. In this study, the bio-based vitrimer was analyzed using the DSC Q20 TA instrument (Bangalore, India). Vitrimer samples, each weighing 10 mg , were prepared. The experiment was conducted under isothermal conditions while maintaining the samples at ambient temperature.

2.4.2. Thermogravimetric Analysis (TGA)

The thermal characteristics of the cured vitrimer were assessed using a thermogravimetric technique. The heating range was set from $25\text{ }^{\circ}\text{C}$ to $700\text{ }^{\circ}\text{C}$ at a rate of $10\text{ }^{\circ}\text{C}$ per minute, with nitrogen serving as the preferred environment. TGA was carried out using a TA Instruments TGA Q50 V20.13 (Bangalore, India), following the ASTM standard E1868 [23]. Each sample weighed 10 milligrams. Thermal properties such as T_i , the temperature at which deterioration or decomposition began; T_p , the temperature at which the

rate of deterioration increased; and T_e , the temperature at which the process ended, were calculated from thermograms.

2.5. Mechanical Testing

The mechanical properties of the vitrimer and epoxy polymer, as well as the CFRP composite with both polymers, were evaluated at room temperature using a Shimadzu AG-X UTM (Mumbai, India) under tensile testing. The tensile tests were conducted in accordance with the ASTM D638 standards for plastics in all blends and composites [24], with a strain rate of 1 mm/min and a gauge length of 60 mm. The elongation of the composites was determined using an extensometer. Four specimens were tested for each material.

2.6. SEM Characterization

To investigate the morphological characteristics of the recycled carbon fiber, scanning electron microscopic (SEM) images were acquired using the scanning electron microscope HITACHI, SU6600 (Bangalore, India) with an accelerating voltage of 5 kV. After reclaiming the fiber from depolymerizing, it was washed with acetone to ensure that the samples were free from any residual dust or debris.

3. Results and Discussions

3.1. Fourier Transform Infrared (FTIR) Spectrometry

The FTIR spectra of the vitrimer were analyzed before and after curing to assess the degree of curing. Figure 4 shows the FTIR spectra of the DGEBA, revealing an absorption band at 911 cm^{-1} , characteristic of the epoxide ring. During the curing process of the DGEBA/CA/cardanol vitrimer, the epoxide group underwent ring-opening, evidenced by a reduction in the intensity of the 911 cm^{-1} peak. The presence of peaks at 1740 cm^{-1} , corresponding to the ester groups, and 1110 cm^{-1} , corresponding to the ether groups, confirms the successful synthesis of the epoxy vitrimer. The 1740 cm^{-1} peak, assigned to the C=O stretching of the ester, indicates the esterification reaction between the DGEBA and carboxyl groups. The 1110 cm^{-1} peak, attributed to the C–O–C stretching of ether, suggests etherification between the DGEBA and the hydroxyl groups in cardanol.

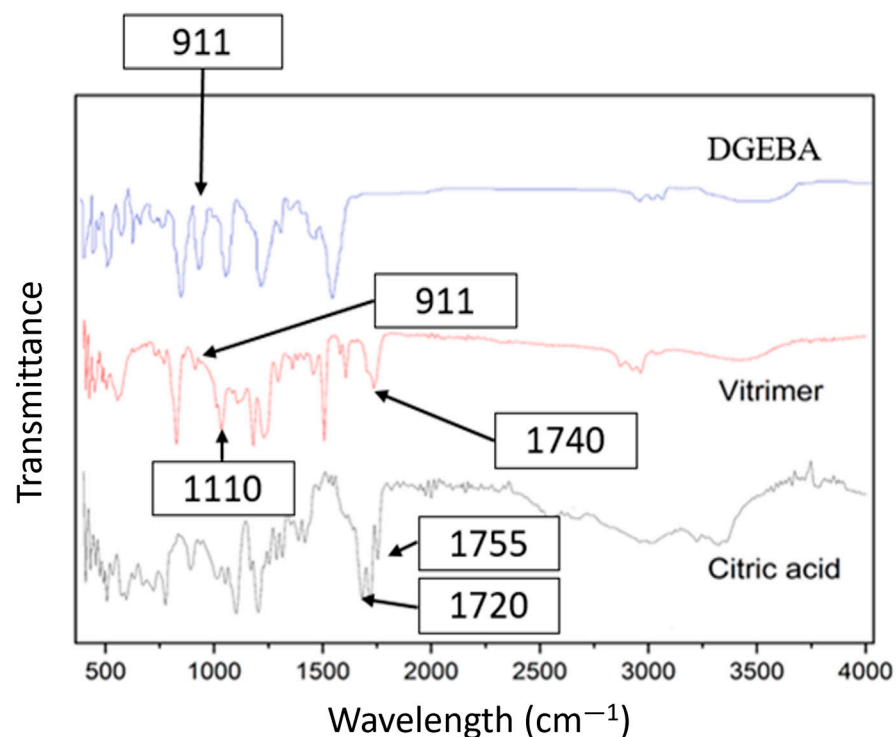


Figure 4. FTIR spectra of the DGEBA vitrimer.

3.2. Differential Scanning Calorimetry

Figure 5 displays DSC thermograms of the DGEBA/CA/cardanol vitrimers of the uncured and cured samples. The curing reaction of DGEBA/CA/cardanol began at approximately ~ 120 °C, while the exothermic peak temperature was observed between 160 °C to 165 °C, as observed from DSC curve. This change in heat flow suggests a phase transition or a transformation occurring within the reaction mixture at that temperature. It could be associated with a chemical reaction, such as the curing or crosslinking of the epoxy resin due to the presence of cross-linker. Hence, 120 °C was selected as the initial curing temperature, followed by 160 °C. The exothermic peak of the cured samples dissipated after curing, indicating the completion of the curing reaction, as confirmed by the FTIR results (Figure 4).

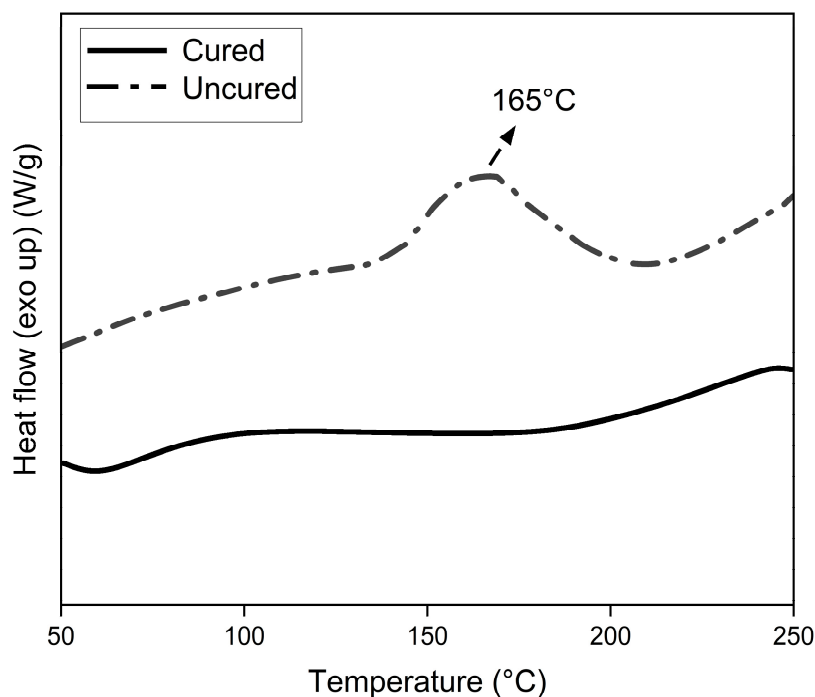


Figure 5. DSC curve for the uncured DGEBA/CA/cardanol blend and cured vitrimer.

3.3. Thermogravimetric Analysis (TGA)

The weight loss observed during the analysis indicates the thermal degradation and breakdown of the crosslinked polymer network in the vitrimer. The decomposition process began at a higher temperature of 250 °C (Figure 6). Approximately 20% decomposition occurred at approximately 370 °C, and further weight loss occurred from 370 °C to 470 °C. The TGA graph for the DGEBA/CA/cardanol vitrimer exhibits a single dip, indicating that the reaction has occurred, and the material has transformed into a polymer. Through the reversible exchange reaction between crosslinks, the vitrimer material has the special feature of releasing deformation stress at high temperatures [11,12,25]. These results suggest that both vitrimers underwent thermal degradation, with the ester linkages being the primary sites of decomposition. The derivative thermogravimetric (DTG) curve shows that the T_{\max} of the sample is ~ 425 °C. The amount of weight loss and the associated breakdown temperature range can reveal information about the vitrimer's thermal stability and degradation mechanism.

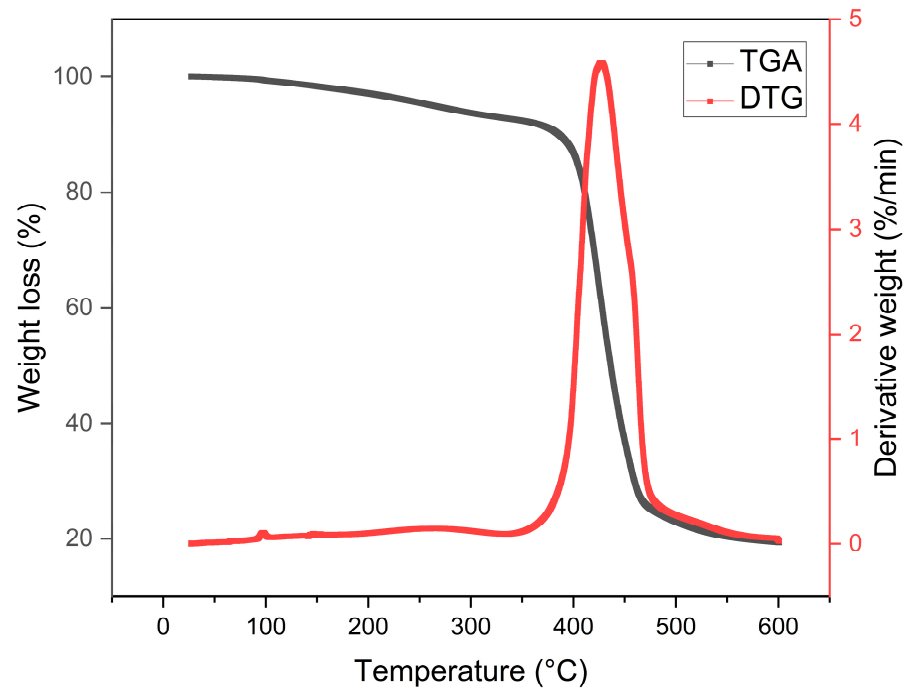


Figure 6. Thermogravimetric analysis (TGA) and derivative thermogravimetric (DTG) curves of the vitrimer polymer.

3.4. Mechanical Behavior

3.4.1. Vitrimer Polymer

The vitrimer specimen exhibited a tensile strength of 50.01 ± 1.25 MPa, with a corresponding tensile modulus of 860 ± 80 MPa, as shown in Figure 7. The maximum tensile strength of the vitrimer polymer (50.01 MPa) is comparable to that of epoxy resins cured with petroleum-based curing agents, which typically exhibit maximum tensile strengths in the range of approximately 44–60 MPa [26].

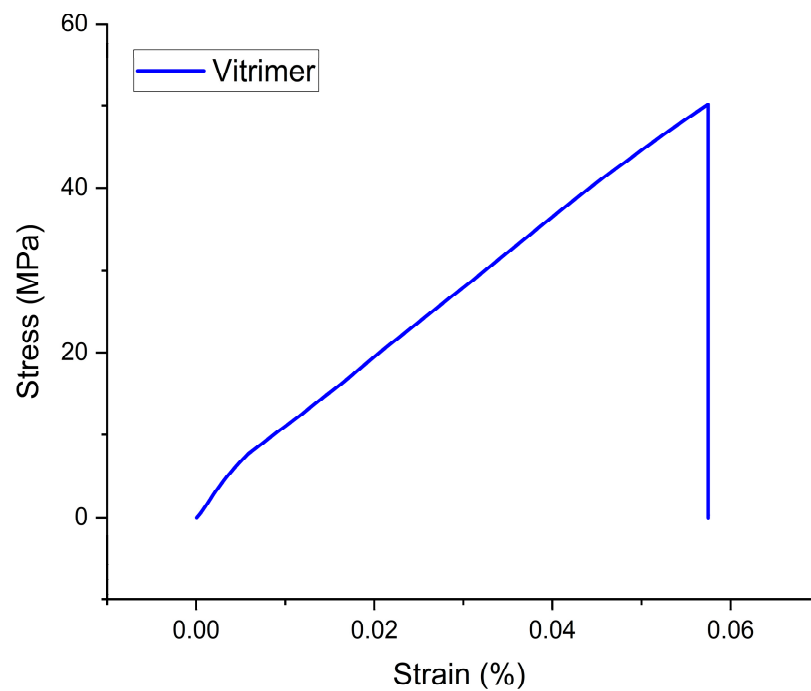


Figure 7. Representative stress–strain curve.

3.4.2. Composite Materials

The tensile test properties of the vitrimer polymer and composite materials are summarized in Table 2.

Table 2. Mechanical properties of the vitrimer polymer and composites.

Material	Tensile Strength (MPa)	Tensile Modulus (MPa)	% Elongation at Break
Vitrimer Polymer	50.01 ± 1.25	860 ± 80	0.058 ± 0.005
Epoxy Composite	446 ± 16.91	$10,878 \pm 93$	0.042 ± 0.04
Pristine Vitrimer Composite	422.31 ± 18.5	9380 ± 82	0.045 ± 0.03
Recycled Vitrimer Composite	402 ± 17.5	8995 ± 64	0.045 ± 0.04

In comparison, the epoxy composite displayed a significantly higher tensile strength of 446 ± 6.91 MPa, accompanied by a higher tensile modulus of 10878 ± 93 MPa. However, the epoxy composite exhibited a slightly lower percentage of elongation at break of $0.042 \pm 0.04\%$, as compared to the pristine vitrimer composite. The pristine vitrimer composite showed a very close tensile strength of 422.31 ± 18.5 MPa, with a tensile modulus of 9380 ± 82 MPa. The failure strain values of both the pristine and recycled vitrimer composites were slightly higher than those of the epoxy composite, indicating the ductile behavior of the vitrimer polymer. Furthermore, the recycled vitrimer composite demonstrated a maximum tensile strength of 402 ± 17.5 MPa, with a tensile modulus of 8995 ± 64 MPa. This $+8/-5$ indicates a strength retention of up to 95% as compared to that of the pristine composite. Si et al. [27] observed similar behavior in the vitrimer–disulphide epoxy composite, which achieved a strength retention up to 90%. These results also suggest the possibility of using vitrimer instead of epoxy polymer for secondary structures and high-end applications.

Figure 8 illustrates the stress–strain curves representing the behavior of the epoxy composite, vitrimer composite, and recycled vitrimer composite. All the composites exhibited similar trends in behavior; in particular, both the pristine and recycled vitrimer composites showed almost the same strength. However, it is noteworthy that the vitrimer specimens reported a higher percentage elongation at the break, suggesting improved ductility and flexibility as compared to the epoxy composite. The recycled vitrimer exhibited even greater ductility as compared to the fresh vitrimer composite, with a slight reduction in strength. Overall, these results highlight the mechanical characteristics of the various specimens and indicate the potential of vitrimer composites as viable substitutes for epoxy composites, offering advantageous strength and elongation capabilities.

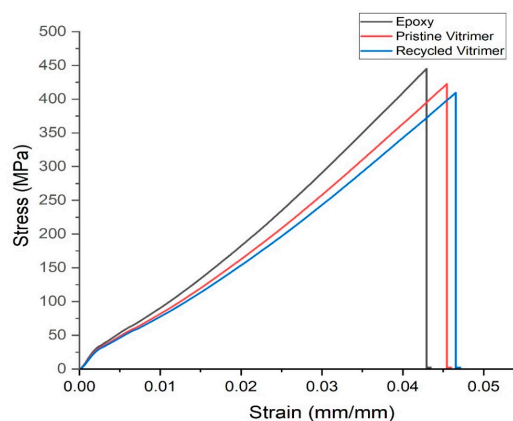


Figure 8. Stress–strain curve of the epoxy composite, pristine vitrimer composite, and recycled vitrimer composite.

3.5. Recycling of Vitrimer Composite

The complete procedure, from composite preparation to the recycling of the carbon fiber, is illustrated in the flowchart (Figure 9). In addition to carbon fiber, there are possibilities for reusing the resin after depolymerization. Quanyi Mu et al. [28] reported the decomposition of resin; the depolymerized resin and residual solvents were efficiently separated through reduced pressure distillation at a temperature of 80 °C. This method opens scopes for a completely sustainable composite material by enabling the reuse of adhesive components and carbon fiber. This will not only promote sustainability by reducing waste and conserving resources but also support a circular economy across various industries.

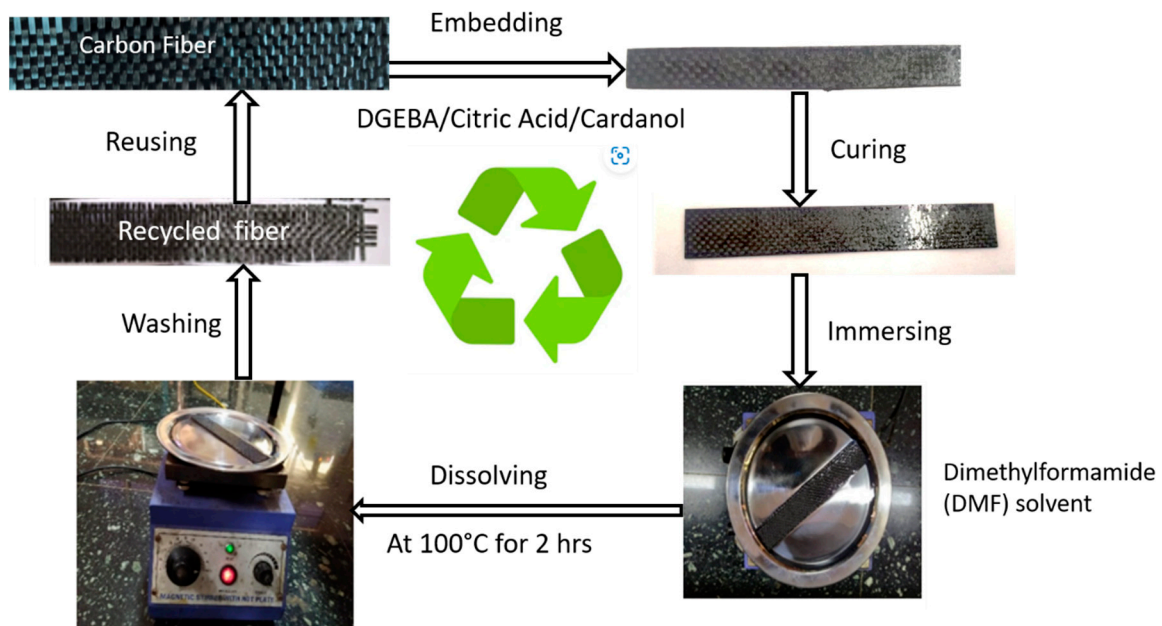


Figure 9. Recycling process of the DGEBA/CA/cardanol vitrimer composite.

The thermosetting nature of the epoxy matrix and its cross-linked networks pose challenges for recycling carbon fiber from CFRP-epoxy composites. However, the DGEBA/CA/cardanol vitrimer exhibited efficient dissolution in DMF, enabling the recycling of carbon fibers from the CFRP composites. The vitrimer composite was immersed in a DMF solution at a temperature of 100 °C for 2 h, resulting in the successful recovery of all layers of carbon fiber from the composite [29]. As the DMF solvent is strong, it did not affect the sizing agent as the dissolution occurred at a relatively low temperature, and the recycled fibers were treated with a chemically resistant sizing agent. To analyze the successful recovery and effectiveness of the recycling, the microscale morphology of the original fiber and the recycled fiber was observed by SEM. The recycled carbon fiber showed a smooth and clean surface that was almost similar to the virgin fiber (Figure 10). Minimal surface damage or resin residue was observed, reflecting the effectiveness of the recycling process through solvent dissolution, preserving the fiber's integrity for potential reuse in new composites based on its retention properties. As a result, 95% of the strength of the recycled fiber composite was retained, indicating that the sizing agent remained almost intact. For more clarity on the effects of the sizing agent, a fiber pull-out test or fiber-matrix adhesion can be performed on the recycled fiber.

The tensile strength of the vitrimer composite was 422 MPa, which is closer to the epoxy composite of 446 MPa, and the reformed vitrimer composite's strength declined marginally (Figure 11). The epoxy composite exhibited an impressive ultimate tensile strength of 446 MPa. However, the vitrimer composite showed maximum tensile strengths of 422 MPa and 402 MPa for the pristine and recycled vitrimer composites, respectively (Figure 11), which is close to the epoxy composite's strength. When evaluating the maximum tensile

strength in relation to the epoxy composite, it is noteworthy that the pristine vitrimer composite achieved approximately 94.61% of the epoxy composite material's strength. Similarly, the recycled vitrimer composite demonstrated a considerable maximum tensile strength of about 90.14% as compared to the epoxy composite. Furthermore, in terms of a direct comparison between the pristine composite and the recycled vitrimer composite, the recycled vitrimer composite showed a commendable performance at approximately 95.26% of the epoxy composite's maximum tensile strength.

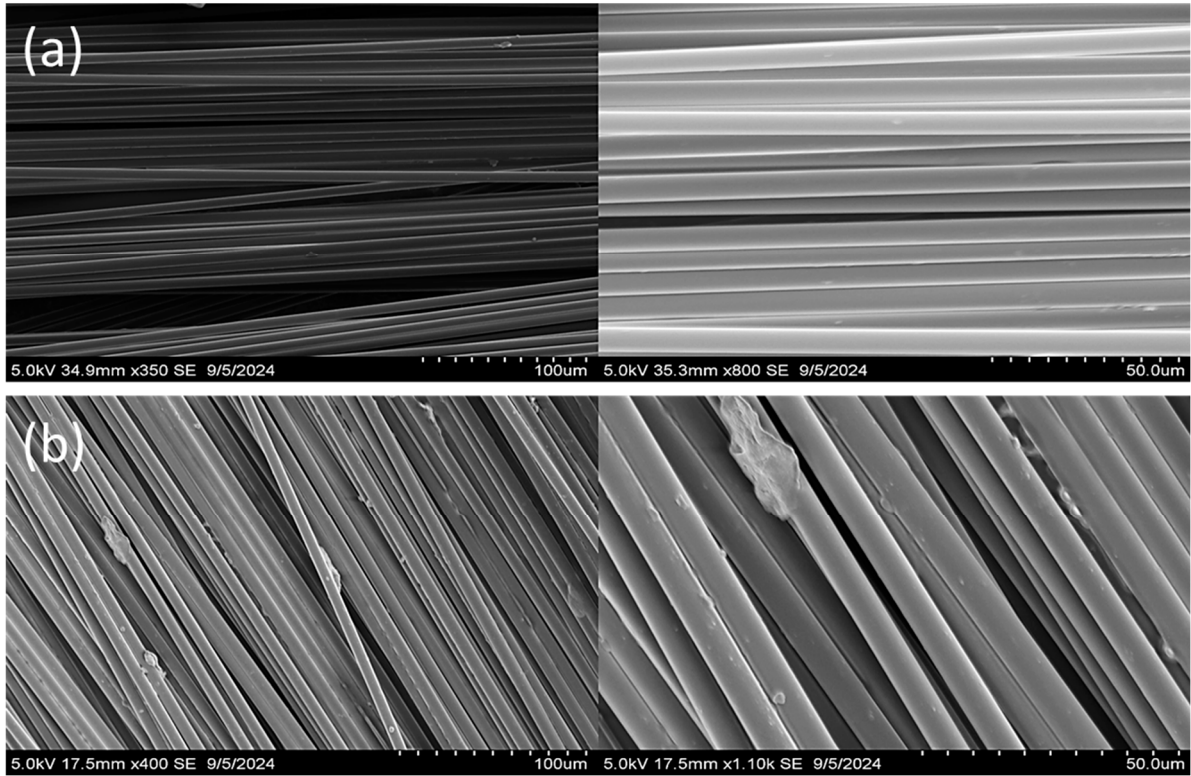


Figure 10. SEM images of the original CF (a) and the recycled CF (b).

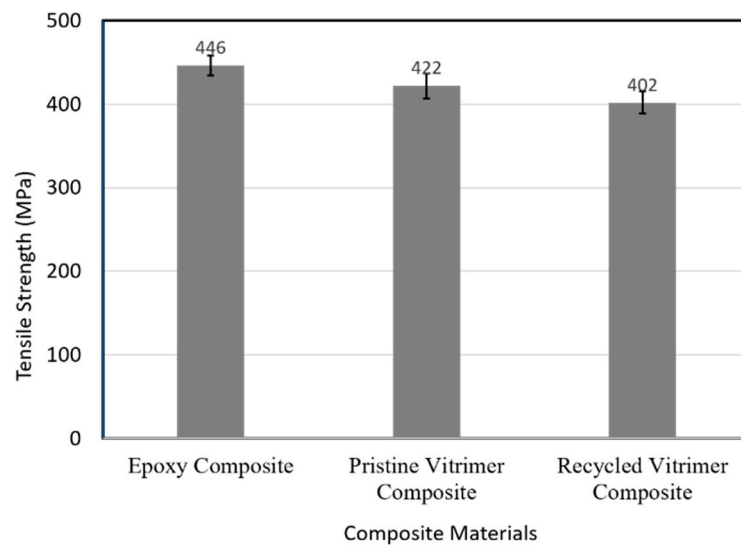


Figure 11. Tensile strength of the composite material and the retained strength of the vitrimer-based composite.

These findings underscore the remarkable potential of the recycled vitrimer composite, which demonstrates promising mechanical properties comparable to both the pristine vitrimer composite and the epoxy composite. The effective integration of the recycled carbon fibers into the vitrimer matrix highlights the feasibility of sustainable composite fabrication while maintaining a significant level of strength. This advancement holds considerable significance for various applications where the utilization of recycled materials is desired, contributing to environmental preservation and resource efficiency.

4. Conclusions

This research study on the resulting DGEBA/CA/cardanol-based epoxy vitrimer provided important insights into its behavior and properties. The DSC analysis revealed a significant change in heat flow at 160 °C, indicating a phase transition/reaction occurrence. FTIR confirmed the successful curing of the vitrimer composite and esterification between the DGEBA and the citric acid. The thermal gravimetric analysis demonstrated a one-stage thermal degradation process, with ester linkage segment decomposition observed between 270 °C and 450 °C. A maximum tensile strength of 50.01 MPa was exhibited for the vitrimer polymer, which is close to that of the regular epoxy polymer, which assures the possibility of replacement of the epoxy with the vitrimer for particular applications. The CFRP composites based on the DGEBA/CA/cardanol (epoxy vitrimer) found outstanding mechanical properties, with a tensile strength of 422 MPa and a tensile modulus of 9.38 GPa. Carbon fibers were successfully recovered from the vitrimer composite using DMF solvent dissolution, and SEM analysis revealed that the carbon fiber surface was clean without any epoxy content. The recycled CFRP composites retained almost 95% of their mechanical strength properties, with a tensile strength of 402 MPa and an elastic modulus of 8.2 GPa. The findings of this study can aid in the creation of effective recycling procedures and the use of recycled carbon fibers in a variety of industries, encouraging the principles of a circular economy and lowering the need for new materials. In order to comprehend the effects of recycling on the composite's performance over time, long-term aging studies can also be carried out.

Author Contributions: Conceptualization, S.B.; methodology, V.R.; formal analysis, S.N. and S.B.; investigation, V.R.; writing—original draft preparation, S.N.; writing—review and editing, J.P. and S.d.B. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Acknowledgments: The authors would like to acknowledge the Ceramics and Composite Lab, NIT Calicut and support of the CESI LINEACT and Brazilian research agencies.

Conflicts of Interest: The authors declare no conflict of interest.

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