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RESEARCH ARTICLE

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Characterization of biobased epoxy resins to manufacture eco-composites showing recycling properties

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Abstract

The present study details the composite fabrication, characterization, and full recyclability of the biobased polymer resins with flax fiber as the reinforcement. Two different biobased resins are selected by varying the resin hardener combination and curing parameters for the comparison. The optimum parameters for the composite preparation are finalized by evaluating the neat resin's mechanical and glass transition temperature (T_g) values. According to the results obtained, a biobased epoxy resin cured by a cleavable hardener displayed the highest T_g (i.e., 93.5°C) upon a two-step cure cycle, guaranteeing full recyclability. Subsequently, eco-composite flat panels are manufactured using the selected formulation reinforced with commercial flax-based fabric. These panel's flexural strength, modulus, and interlaminar shear strength are measured after each curing step. The recyclability yield of the composite is tested, through a specific chemical process, demonstrating the possibility of separating and recovering both the constituent fibers and resin from the composite panels.

KEYWORDS

biocomposite, mechanical properties, post-curing, recycling, thermomechanical, thermosetting resin

1 | INTRODUCTION

Natural fiber reinforced composites (NFRC) are a potential topic of research in sustainable materials owing to their lightweight, environmentally friendly nature good, specific strength, and stiffness. However, most NFRCs are made from the non-renewable thermoset polymer derived from fossil resources. The escalating awareness of fossil fuel depletion and the environmental concerns incited the research community to emphasize developing new eco-friendly green materials from renewable resources.[1,2] Recent events such as the first major wave of decommissioning the composite made wind turbine blades which reached the end of life by 2019–2020,[3] massive aircraft decommissioning in 2020 on the aspect of the Covid pandemic and the ban of composite landfilling in Germany in 2009[4] alerts the desperate necessity for recyclability solutions or developing sustainable composite materials. Recycling of fiber reinforced polymer composites (FRPCs) is imposed by the properties of polymer resin. The epoxies and unsaturated polyesters are the common thermoset resins opted for 80% of the FRPCs.[5] They have high

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mechanical properties, thermal and chemical resistance, durability, and dimensional stability.[6–8] However, the irreversible and crosslinked state of cured thermoset resin makes them unsuitable for recycling and causes significant effort in recovering the fibers and resins.[9–11]

The existing approaches include the introduction of labile linkages such as carbonate bonds, ester bonds, acetal bonds, Schiff base, and sulfur-containing structures to the thermoset resin.[12–14] Hongwei et al.[15] developed degradable epoxy by introducing an aromatic disulfide bond. However, these transesterification techniques require high-temperature conditions and the presence of metal catalysts, which increases the recycling method's cost and complexity. Various other recycling methods, such as thermolysis and solvolysis, are also attempted to date. Then again, these techniques critically damage the fiber's chemical structure, properties, and size. Such reclaimed fibers cannot be reused for the same high technical applications used before.[16] This reduces the commercial value and use of fiber reinforcement.

Other approaches include developing weak and unstable covalent structures for the carbon fiber reinforced composites (CFRC) that enable recycling under mild conditions. Hashimoto et al.[17] investigated a dynamic acetal (—O—CH(CH_3)—O—) epoxy system that can degrade via acid hydrolysis of the acetal linkages at room temperature. A study by Taynton et al.[18] reported the full recycling of CF using a mild transamination reaction of the dynamic polyamine (—C═N—) system. Similarly, Luzuriaga et al.[19] demonstrated remarkable recyclability of the CF using the thiol-disulfide exchange reaction on the dynamic disulfide (—S—S—) epoxy system. Compared with the available thermoset resins, these epoxy systems help achieve full CF recyclability. Then again, with weak and unstable covalent bonds, these composites can only attain reasonable or low thermal stability, chemical resistance, and mechanical properties.[16]

Mechanical, thermal, and chemical recycling techniques are generally practiced for the recovery of the fiber and the matrix from the composite products. Among them, the mechanical and thermal recycling process either results in partial recovery or fully damaged fiber reinforcement and matrix. Chemical recycling claims to recover the less degraded fibers and valuable monomers from the epoxy.[20,21] Li et al.[12] preheated the thermoset composites in acetic acid in their study. They later oxidized them in a solution containing hydrogen peroxide and acetone, achieving clean carbon fibers (CF) with a strength close to 95% of their original strength. However, chemical recycling uses chemical solvents or strong acids and these conditions might be acceptable only for synthetic fibers. The presence of strong acids inhibits its application for the natural fibers reinforced composites. Additionally, thermoset recycling involves severe conditions such as high temperature and pressure that can damage the fiber reinforcement. The present study uses biobased resins that are having cleavable ammines which assist the recycling of the thermoset resin to recyclable thermoplastic matrix and clean fibers at mild temperature conditions (80–120°C).[22,23] Until now, an efficient and non-destructive way to extract the fibers from thermoset resin is still a challenge.[20] Thus, developing a recyclable resin and a suitable chemical recycling technique that can recover the reinforcement under mild conditions without damaging the chemical structure and mechanical properties is a demanding requirement.[16] The attempts to recycle thermoset composites using supercritical fluids were found successful. However, they are limited to lab-scale setups and cannot be implemented for commercial needs.[24] The novelty of the present manuscript is about combining together the chemical recovery of the flax fiber reinforcement through the chemical recycling process without affecting the properties of fiber and matrix.

The explicit solutions are replacing synthetic fibers with natural fibers and using recyclable thermoset resins or biobased polymers. The availability, low cost, biodegradability, and relatively high specific strength and rigidity of the natural fibers highlighted their importance as reinforcement materials in composites. A good amount of research on natural fibers such as jute, hemp, flax, kenaf, and so forth, with epoxy resin and unsaturated polyester resins, are reported.[25–29] Among the natural fibers, flax has density values of 1.3–1.5 g/cm^3, interesting mechanical properties, and good stiffness values. The density of flax is much lower than glass (2.5 g/cm^3) and carbon fibers (1.75–1.93 g/cm^3).[30] The flax fibers also registered a 12% reduction in the primary energy consumption values compared with glass and carbon fibers.[31] The sustainable reinforcement material (natural fibers) also gained attention in the automotive industry to ensure lightweight and cheaper transportation, especially for future electric vehicles.[32]

The present research considered the flax fiber reinforced biobased resin composites manufactured using vacuum-assisted resin transfer molding. Two resin systems AMPRO™ BIO and Polar Bear resin, are compared on various curing conditions and resin hardener combinations, targeting high recyclability and glass transition temperature (T_g). The influence of the diluent is also studied to demonstrate how it affects the T_g and the mechanical properties. The flexural and the interlaminar shear strength values of the flax fiber polar resin composites are evaluated, and a detailed investigation of their recyclability is discussed. The recyclamine is cleaved in an aqueous solution with acetic acid under 80°C achieving cleaner and less degraded. The present article reports the mild recycling of the high-strength biobased thermoset resin that can recover clean and reusable natural

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fibers (flax). Furthermore, such a technique finally looks to commercialize a large volume of composites. The study also investigates the composite developed from natural fiber (flax) sources and recyclable biobased thermoset resins. To the best of the author's knowledge, minimal attempts are made in such a combination.

2 | MATERIALS AND METHOD

2.1 | Materials

AMPRO™ BIO is a biobased epoxy resin for composite processing purchased from Gurit (UK) Ltd. The product is optimized for use at a temperature between 15 and 25°C and has a biobased carbon content between 40% and 60%. It is assigned to the class 2 category under the TUV OK Biobased certification scheme, based on the ASTM D6866 standard.[33] The viscosity of the resin is 3028 cP at 15°C and 979 cP at 25°C. AMPRO™ Fast Hardener and AMPRO™ Slow Hardener were also purchased from Gurit (UK) Ltd. The two hardeners differ in their pot-life value, that is, 30 min for the Fast Hardener and 45 min for the Slow one. Moreover, they vary even for the viscosity values, 1260 and 1100 cP at 25°C for the Fast and Slow Hardeners, respectively. A biobased epoxy (Polar Bear), recyclable hardener, and diluent purchased from R*CONCEPT (Barcelona, Spain) are also selected for the comparison. Polar Bear is a biobased epoxy with the chemical composition of bis-[4-(2,-3-epoxipropoxi)phenyl]propane with a bio content of more than 19%. The Recyclamine™ R*101 is the recyclable hardener with 2,2-Bis(aminoethoxy) propane chemical composition and R*Diluent with the chemical composition oxirane, mono[(C12-14-alkyloxy)methyl] derivs. The diluent (R*Diluent) is added to alter the viscosity of the Polar Bear for the infusion process. The R*Diluent type is selected because its reactive nature makes it possible to co-react it within the epoxy network. The critical factor for the recyclability of this amine is the presence of acid–cleavable groups in its chemical structure. Flax fiber twill fabrics having an average thickness of 0.80 mm and an areal density of 300 gsm is selected as the fiber reinforcement for the composite fabrication. Five layers of flax fibers are selected for the composite fabrication.

2.2 | Epoxy resin system formulation

Different formulations of epoxy resin and amine are altered in this experimental work to find the most suitable combination for the thermoset eco-composite fabrication. Several curing cycles were studied to draw appropriate considerations on the curing state and optimize the glass transition temperature (T_g). Initially, the biobased AMPRO™ BIO was mixed with the slow and the fast hardener individually at the same amine ratio (at 30 phr). The next category was investigated using the biobased Polar Bear resin and the recyclable hardener Recyclamine™ R*101. The resin to hardener ratio was selected as 100:22 phr as recommended by the R*CONCEPT resin manufacturer. To make the resin infusion process more convenient, 10 wt% of R*Diluent to the weight of Polar Bear resin was added. All the formulations and the corresponding curing conditions investigated are shown in Table 1, where the term BB denotes the biobased epoxy system. In comparison, the BBR refers to both biobased and recyclable epoxy systems.

The blend between epoxy resin and amine was achieved through mechanical mixing in all the epoxy systems. Once mixed, all the blends were degassed using a planetary mixing with a vacuum (Thinky mixer ARV310, THINKY USA Laguna Hills). Next, each epoxy system was poured into silicon molds to manufacture samples with geometries suitable for the thermal analysis. Each blend was cured according to the curing cycles shown in Table 1. Once the formulation with the highest T_g was identified, appropriate samples were manufactured for mechanical characterization (tensile and flexural tests). The latter was measured using an Instron 5985 universal testing machine (Instron, Milan, Italy) equipped with a load cell of 10 kN. The Blue Hill 3.61 software (Instron, MA) was used for system control and data collection.

2.3 | Composite fabrication

The composite laminate was fabricated using cold vacuum-assisted resin transfer molding. A mold resealing agent was sprayed over the aluminium mold to remove the prepared laminate easily. Five layers of flax fibers were stacked between the release film and infusion mesh over the mold. Figure 1 represents the vacuum-assisted resin transfer molding technique. The vacuum was then applied, and the resin was infused into the vacuum bag. The prepared composites were kept for curing at 25°C for 24 h. The next set of composites was subjected to two-stage curing at 100°C for 3 h after the initial curing for 24 h at 25°C. The samples were then cut according to the ASTM D7264 and ASTM D2344 standards for the flexural and interlaminar shear strength studies, respectively.

2.4 | Differential scanning calorimetry

A Shimadzu DSC-60 (Shimadzu, Kyoto, Japan) equipment was used to carry out the calorimetric measurements. The DSC analyses were run on uncured formulations (liquid

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TABLE 1 Investigated epoxy matrix formulation and corresponding curing cycles.

Sample ID	Formulation	Mixing ratio by weight	Curing cycle
BB1.0	AMPRO™ BIO + AMPRO™ Fast Hardener	100:30	Uncured
BB1.1	AMPRO™ BIO + AMPRO™ Fast Hardener	100:30	@25°C for 24 h
BB1.2	AMPRO™ BIO + AMPRO™ Fast Hardener	100:30	@70°C for 3 h
BB1.3	AMPRO™ BIO + AMPRO™ Fast Hardener	100:30	@100°C for 3 h
BB2.0	AMPRO™ BIO + AMPRO™ Slow Hardener	100:30	Uncured
BB2.1	AMPRO™ BIO + AMPRO™ Slow Hardener	100:30	@25°C for 24 h
BB2.2	AMPRO™ BIO + AMPRO™ Slow Hardener	100:30	@70°C for 3 h
BB2.3	AMPRO™ BIO + AMPRO™ Slow Hardener	100:30	@100°C for 3 h
BBR1.0	Polar Bear + R*Diluent(10%wt) + R*101	100:22	Uncured
BBR1.1	Polar Bear + R*Diluent(10%wt) + R*101	100:22	@25°C for 24 h
BBR1.2	Polar Bear + R*Diluent(10%wt) + R*101	100:22	@70°C for 3 h
BBR1.3	Polar Bear + R*Diluent(10%wt) + R*101	100:22	@100°C for 3 h

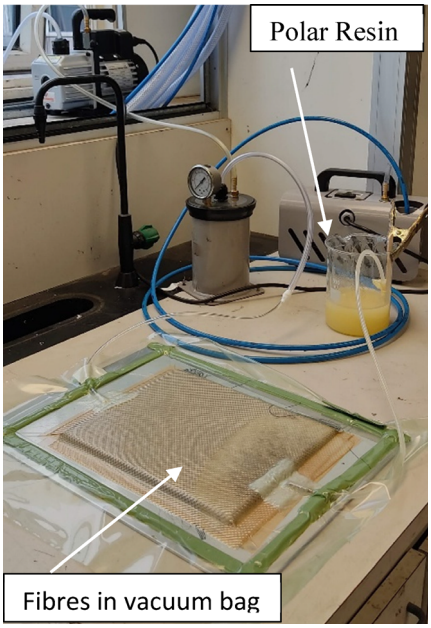

FIGURE 1 Vacuum-assisted resin transfer molding

state) and cured at 25°C (solid state). Samples weighing about 6 mg were placed on the 40-µl sealed aluminium crucibles. Once the analysis started, each sample was heated from room temperature (25°C) up to 250°C at a rate of 20°C/min in air. The total exothermic heat released was calculated from the area of the DSC exothermic peak.

2.5 | Dynamic mechanical analysis (DMA)

The Dynamic mechanical analysis was run using a dynamic mechanical thermal analyzer (TRITEC 2000 by Triton Technology, Leicestershire, UK). DMA specimens were cast and cured as bars with dimensions equal to 10 mm × 6 mm × 4 mm. The single cantilever deformation mode was selected for the measurement as it is better suited for characterizing material through the glass transition[34] and running the temperature scans. After reaching the equilibration step at 25 °C, the temperature was increased at a rate of 2°C/min up to 150°C. For each test, the displacement was set at 200 µm, and the multi-frequency condition was used with frequencies of 1 to 10 to 50 Hz. The tan δ versus temperature at 1 Hz was plotted for each blend considered.

2.6 | Tensile test

Tensile ASTM D638 type IV samples were fabricated by pouring the epoxy resin into silicone molds. Each test was run in strain control mode at 2 mm/min crosshead displacement and using a clip extensometer with a gauge length of 25 mm. Five samples were tested for each condition to evaluate the ultimate tensile strength (UTS) and Young's modulus (E) values.

2.7 | Flexural and interlaminar shear strength test

The flexural properties of the resin material were evaluated using the ASTM D790 standard. The specimens were prepared with 80 mm length, 10 mm width, and 4 mm thickness by pouring the epoxy resin into a silicon mold. The span length was set at 60 mm, and the crosshead displacement was maintained at 2 mm/min. Five samples

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FIGURE 2 Chemical recycling process schematization

were tested for each condition to attain the flexural strength and modulus values along with the flexural stress versus flexural strain (%) curves. Additionally, the flexural properties of flax fiber reinforced composites samples were analyzed using the ASTM D7264 standard, with a specimen thickness of 3.5 mm and a span to thickness ratio of 32:1. The testing was conducted at a crosshead displacement of 1 mm/min. The interlaminar shear strength studies of the fiber reinforced composites were evaluated using the ASTM D2344 standards. The sample thickness was 3.5 mm, and the span to thickness ratio was 4:1. The test was operated at a crosshead displacement of 1 mm/min.

2.8 | Chemical recycling process

The chemical recycling process was selected to recover the flax fibers and convert the thermoset resin to reusable thermoplastic resin. The present method is a modified version of the author's previously published work.[24] Flax fiber epoxy composite weighing 20 g with 40% fiber volume fraction was solubilized in 300 ml of 75% volume of acetic acid solution at 80°C for 1.5 h. As soon as the thermoset dissolved, the acetic solution was filtered to recover the dried flax fibers. The acid solution was then roto-evaporated at a pressure ranging between 30 and 90 mbar, at 60°C and 3500 rpm rotation speed. About 225 ml of acetic acid was recovered and stocked for the new dissolution procedure for the chemical recycling treatments. Furthermore, into the 75 ml of solution containing the dissolved thermoset matrix, 20 wt% of recovered flax fibers (equal to 2.4 g) was added. Next, this mixture was neutralized in 200 ml of ammonium hydroxide solution containing 50% vol of distilled water and 50% vol of ammonium hydroxide solution (28.0–30.0% NH_3 basis). This way, it was possible to obtain a compound made of recycled thermoplastic (rTP101) filled with 20 wt % of recycled flax fibers directly from the chemical recycling process. Finally, the recycled compound was washed with distilled water and dried at 50°C for 24 h under vacuum conditions. The whole chemical recycling process is schematically represented in Figure 2.

2.9 | Morphology analysis: Scanning electron microscopy

The morphologies of the virgin flax fibers used to manufacture the eco-composite versus the recycled ones were investigated by using a scanning electron microscope SEM EVO 15 (Zeiss, Cambridge, UK), to evaluate possible damage suffered by the fabric during the chemical recycling process performed. The samples were gold sputtered before the analysis. The thin gold film was deposited by a sputtering process, carried out with a sputter coater machine Agar Sputter Coater AGB7340 (Assing Italy). No other additional pre-treatment was applied. The SEM analysis was run at different magnifications, that is, 50× and 150×. The electron source used was a

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Lanthanum Hexaboride (LaB6) emitter, while the electron high tension (EHT) value was set at 20 kV.

3 | RESULTS AND DISCUSSION

3.1 | DSC analysis of biobased and recyclable epoxy

The uncured epoxy resin formulations prepared are tested for the DSC analysis. These analyses are essential to have more knowledge about the crosslinking process. The resin's temperature increases due to the exothermicity, and the crosslinking rate rises in the autocatalytic process.[35] The results obtained are summarized in Table 2. Figure 3 represents the DSC characterization curves for the biobased and recyclable epoxy system in uncured and cured at 25°C conditions. The BB1.0 and BB1.1 samples are denoted as black curves, BB2.0 and BB2.1 in red curves, and BBR1.0 and BBR1.1 in blue. Focusing on the uncured biobased BB1.0 formulation, the plot shows an exothermic peak at 97.55°C with an initial onset temperature of 50.17°C (black curve in Figure 3A). Total exothermic heat released in correspondence to this peak is 105.38 J/g. A similar result is obtained for the uncured BB2.0 resin (red curve in Figure 3A). In this case, an exothermic peak at 98.34°C, which starts at 48.86°C, is identified from the plot. The corresponding total exothermic heat released is 162.14 J/g. The biobased and recyclable uncured epoxy resin BBR1.0 shows an exothermic peak in correspondence with higher temperatures, thus revealing the presence of thermally reactive groups that need to react to start the curing process at higher temperatures. Indeed, the uncured BBR1.0 formulation shows an exothermic peak at 112.3°C, which begins at 72.40°C (blue curve in Figure 3A). The total exothermic heat released in correspondence to this peak is 272.65 J/g, higher than the cured resin system.

The same three formulations are cured at 25 °C for 24 h, and the DSC test is performed. After 24 h, they all presented a solid-state, confirming that the crosslinking process occurred. The DSC results from Figure 3B, BB1.1 (black curve), BB2.1 (red curve), and BBR1.1 (blue curve) explain that all the reactive moieties are reacted, as there are no exothermic peaks in the curves. Nikafshar et al.[36] also reported a similar trend on the effect of curing for the biobased epoxy system derived from vanillin, which can compete with diglycidyl ether bisphenol A (DGEBA).

TABLE 2 Parameters correspond to the DSC characterization run on uncured and cured at 25°C biobased epoxy system BB1, BB2, and BBR1.

Sample ID	Peak (°C)	Onset (°C)	Heat (J/g)
BB1.0	97.55	50.17	105.38
BB2.0	98.34	48.86	162.14
BBR1.0	112.33	72.40	272.65

3.2 | DMA analysis of biobased and recyclable epoxy

Dynamic mechanical analysis (DMA) is also performed on the resin combinations (as mentioned in Table 1) to obtain the glass transition temperature (T_g) values. Figure 4 illustrates the DMA plots for different biobased resins BB1, BB2, and BBR 1 under different post-curing conditions, where 25°C is represented in black, 75°C in blue, and 100°C in the red curve. A common trend is

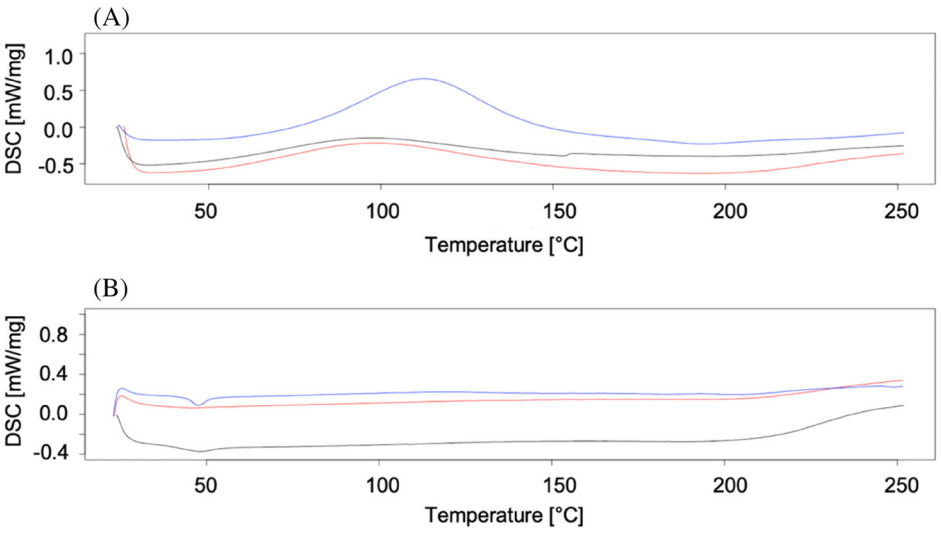

FIGURE 3 Plots obtained from DSC characterization of biobased and recyclable epoxy system in (A) uncured and (B) cured at 25°C for 24 h. BB1.0 and BB1.1 (black curves), BB2.0 and BB2.1 (red curves), BBR1.0 and BBR1.1 (blue curves)

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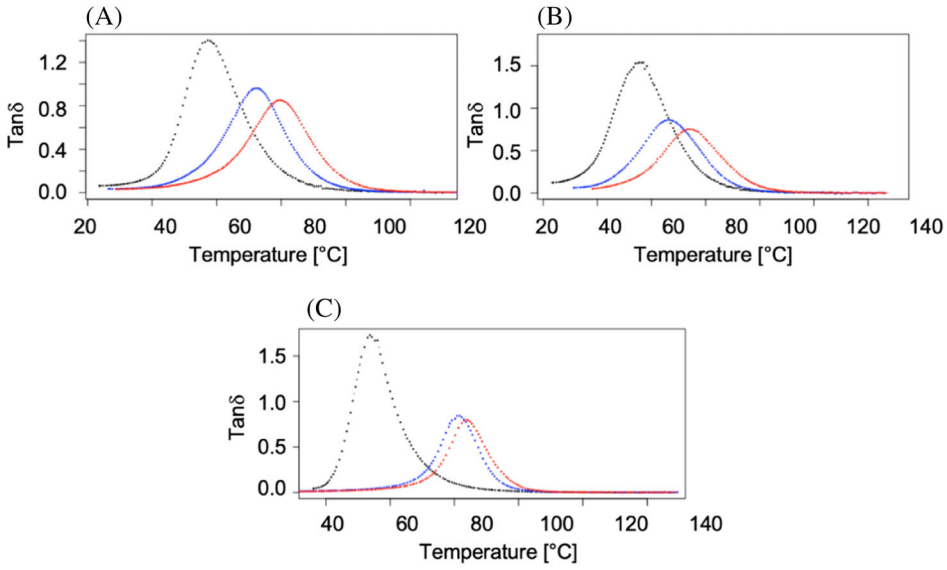

FIGURE 4 Plots obtained from DMA of different biobased epoxy systems: (A) BB1, (B) BB2, and (C) BBR1. Each of them is cured at different temperatures: 25°C (black), 75°C (blue), and 100°C (red)

noted for the DMA test results on each formulation cured at different temperatures. The results infer that the T_g values are enhanced as the curing temperature is raised. Figure 4 explains that the T_g values of the samples BB1.1 and BB1.2 increase by 27% when the curing temperature is raised from 25°C up to 70°C. In contrast, a slight increase of 10% is noted when the curing temperature is changed from 70°C to 100°C. A similar trend is observed for BB2.1, BB2.2, and BB2.3 formulations. The increase in the T_g value is 18% when the curing temperature is altered from 25°C to 70°C, whereas an 11% increase when the curing temperature is changed from 70°C to 100°C. The glass transition temperature increases when curing the resin at a temperature below its T_g.[37] Thus, for the present comparison, when the post-curing temperature was raised to the glass transition temperature values of the polymer, the T_g values tend to show significantly less improvement. The resin formulations BBR1.1, BBR1.2, and BBR1.3 (with diluent) also reported a rise in their T_g value with the curing temperature. The BBR1.1, BBR1.2, and BBR1.3 signify the biobased recyclable polymer with 10% diluent post-cured at 25, 70, and 100°C, respectively (as mentioned in Table 1). The T_g value improved by 52% when the post-curing temperature is maintained at 70°C, whereas the T_g value shows minimal improvement by 3% when the curing temperature is moved to 100°C. The similarity in the trend or the shift in the T_g peak values implies the increase in the crosslink density of the resin system on account of the thermal curing. Moreover, it is noted that the bio-based and recyclable epoxy system BBR1.3 shows the highest T_g value in accordance with its higher heat of reaction, causing the increase in the crosslink density of the matrix.[38]

3.3 | Effect of diluent and curing on the T_g values

The influence of diluent in the resin system and its effect on the T_g value is analyzed by comparing two resin combinations with diluent (BBR1) and without diluent (BBR2). The BBR2 resin composite is prepared by mixing polar biobased and Recyclamine R*101 (hardener) at the mixing ratio of 100:22 phr. The DSC test also analyzes the effect of curing cycles to observe its influence on the T_g values. Figures 5A illustrate the DSC curve of the uncured BBR1.0 and BBR2.0 samples, whereas Figure 5B shows curves for the cured samples (25°C for 24 h) BBR1.1 and BBR2.1. In Figure 5, the samples BBR1.0 and BBR1.1 are presented as blue curves, whereas BBR2.0 and BBR2.1 are indicated in green color. Table 3 shows the exothermic peak values, onset peak value, and exothermic heat released during the DSC analysis of the uncured and post-cured samples without the diluent. The results explain that the curing process for 24 h at 25 °C can increase the crosslinking among the polymer chains, which is evident from the increase in the T_g values.

The peak for the uncured BBR2.0 formulation is identified at 124.58°C, with the start point at 79.36°C and the total exothermic heat released equal to 228.05 J/g (green curve in Figure 5A). The two uncured formulations have quite similar behavior. The cured resin BBR2.1 (green curve in Figure 5B) presents an exothermal peak and onset values at 133.74 and 104.02°C, respectively. The exothermic heat generation value, in this case, is 24.16 J/g. Some moieties are still unreacted even after completing the curing process at 25°C for the BBR2.1 epoxy system. At the same time, the BBR1.1 system did not show any exothermal reaction in the DSC analysis after the

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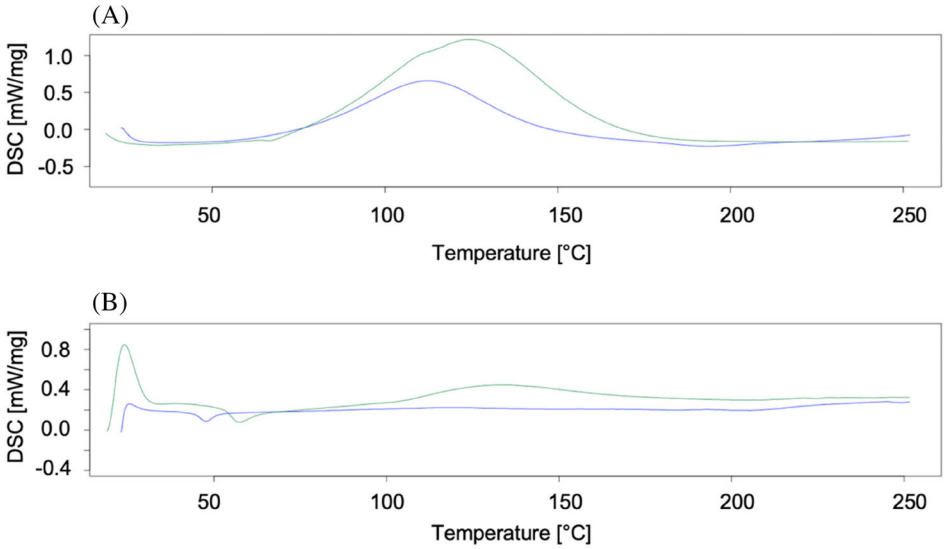

FIGURE 5 Plots obtained from DSC characterization of biobased and recyclable epoxy system at different curing conditions (A) uncured and (B) cured at 25°C: BBR1.0 and BBR1.1 (blue); BBR2.0 and BBR2.1 (green)

TABLE 3 Parameters correspond to the DSC characterization run on uncured and cured at 25°C biobased epoxy system BBR2.

Sample ID	Peak (°C)	Onset (°C)	Heat (J/g)
BBR2.0	124.58	79.36	228.05
BBR2.1	133.74	104.02	24.16

curing process under the same conditions. The DSC curves generally show the variation in the heat flow versus temperature. The change in the heat flow vs. temperature is associated with the absorption energy indicating the changes in the physical state of the polymer. Curing the resin at a temperature below the upper bound value of the resin ($T_{g\infty}$) can shift the heat flow peak vs. temperature towards the right, as presented in Figure 5.[37] Based on these findings, further comparison between the two systems is carried out through DMA analysis. The two curing cycles presented in Table 4 were investigated, targeting the maximum T_g values by focusing on the full crosslink density of the epoxy system as an outcome of the thermal curing. Figure 6 illustrates the DMA test results for the sample types, as mentioned in Table 4.

3.4 | DMA analysis of the biobased and recyclable epoxy under curing conditions

From the experimental analysis, it is inferred that the diluent (10 wt%) does not significantly alter the T_g values for the curing conditions maintained at 25°C for 24 h. Cicala et al.[39] also reported a similar finding using epoxy-reactive diluents. A similar result was obtained by Delevar et al.[40] in studying the effects of reactive diluents in quantity <15% by weight added in DGEBA epoxy resin. Maiorana et al.[41] got similar results with epoxy-modified eugenol when the diluent content was kept below 15 wt%. In our study, the addition of diluent slightly reduces the T_g value when the epoxy is post-cured. This is due to the diluent introducing higher microstructural heterogeneity in the network compared to the sample without diluent.[42] It is possible to achieve a higher T_g value without the reactive diluent in the blend. Comparing some of the polymer combinations as mentioned in Table 4, it is noted that the T_g value related to the BBR2.2 system is 36% higher than the T_g obtained for the BBR1.5. Here, BBR2.2 signifies the polar resin sample without diluent, and BBR1.5 denotes the Polar resin samples with diluent. Both these systems were post-cured for 3 h at 100°C to compare and analyze the influence of diluent. In conclusion, the biobased and recyclable system (BBR2) shows the best T_g value (i.e., 96.20°C).

3.5 | Tensile and flexural properties of the biobased and recyclable epoxy

Further, both the BBR2.1 and BBR2.2 systems are analyzed to determine the effect of post-curing on the tensile and flexural properties. Table 5 presents the polymer sample's tensile and flexural properties of the BBR2.1 (cured at 25°C for 24 h) and BBR2.2 (post-cured at 100°C for 3 h). The post-curing significantly improved the tensile and flexural properties. The tensile strength values of the post-cured resin increased by 33% (from 39.87 to 52.94 MPa), while the tensile modulus value increased by 6% (from 2.44 to 2.58 GPa). A similar trend is also observed for the flexural properties. The flexural strength values for the post-cured samples were also enhanced by 72% (from 40.18 to 69.28 MPa) and the flexural modulus

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by 33% (1.33–1.77 GPa). The curing and post-curing temperature can significantly contribute to the structural performance of the polymer in terms of the T_g and the mechanical properties.[43] The tensile and flexural properties tend to increase until the post-curing temperature is raised up to the resin's upper bound value ($T_{g\infty}$). The ($T_{g\infty}$) corresponds to the T_g value of the fully cured network at which the maximum cross-linking of the resin is reached. Post curing below the $T_{g\infty}$ value promotes crosslinking most effectively. Quite the reverse, thermal degradation can reduce mechanical properties when the post-curing temperature is raised above the $T_{g\infty}$.[37] The curing temperature selected for the biobased and recyclable resin BBR2.2 is below the $T_{g\infty}$ value of the resin. Thus, the mechanical properties attained an improvement. However, if this curing temperature is raised above the $T_{g\infty}$ value of the polymer, it can lead to porosity in the matrix and eventually reduce the mechanical performance. Thus, the optimized curing cycles for the fully recyclable and partial biobased epoxy system allowed to obtain a high value of T_g and increased mechanical responses compared with results previously found for the partial biobased epoxy system containing soybean oil[44] and Bisphenol A (BPA) type epoxy thermosetting resin.[45] The stress–strain plot from the tensile and flexural tests for the BBR2.1 and BBR2.2 formulations is presented in Figure 7. A more brittle behavior is witnessed for the epoxy system without post-curing. In contrast, an apparent increase of both tensile stress and tensile strain (%) values (Figure 7A), as well as flexural stress and flexural strain (%) values (Figure 7B), are obtained for the samples that are post-cured. Moreover, the post-cured epoxy system (BBR2.2) shows ductile behavior. The polymer resins cured at room temperature may not achieve complete crosslinking. The post-curing can activate the crosslinking faster and improve the composite's mechanical properties.[37]

TABLE 4 Different curing cycles for the biobased and recyclable epoxy system BBR1 and BBR2.

Sample ID	Formulation	Mixing ratio by weight	Curing cycle
BBR1.4	Polar Bear + R*Diluent(10%wt) + R*101	100:22	25°C for 24 h
BBR1.5	Polar Bear + R*Diluent(10%wt) + R*101	100:22	25°C for 24 h + 100°C for 3 h
BBR2.1	Polar Bear + R*101	100:22	25°C for 24 h
BBR2.2	Polar Bear + R*101	100:22	25°C for 24 h +100°C for 3 h

3.6 | Flexural and interlaminar shear strength properties of flax fiber biobased recyclable resin composite

The fiber-reinforced composites are prepared using the biobased recyclable resin BBR2.1 and BBR2.2 to evaluate the flexural and interlaminar properties of the composites. The sample FBBR2.1 denotes the flax fiber

TABLE 5 Tensile strength, Young's modulus, flexural stress, and flexural modulus values of the BBR2.1 and BBR2.2 biobased and recyclable epoxy resin.

Sample ID	Tensile strength (MPa)	Young's modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)
BBR2.1	39.38 ± 2.66	2.41 ± 0.66	39.72 ± 1.43	1.35 ± 0.17
BBR2.2	52.75 ± 1.59	2.62 ± 0.19	69.34 ± 2.08	1.75 ± 0.07

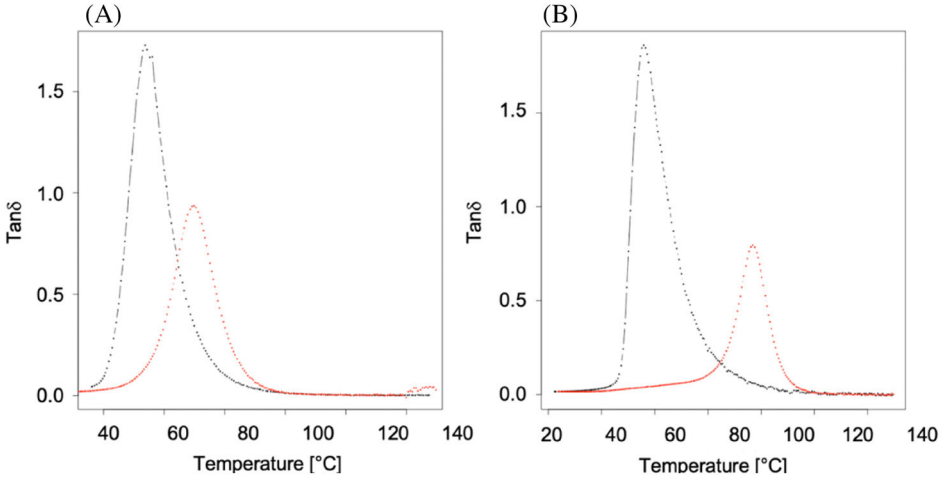

FIGURE 6 Plots from DMA of different biobased and recyclable epoxy systems: (A) BBR1.4 - BBR1.5 and (B) BBR2.1 - BBR2.2. All are cured at different temperatures: At 25°C for 24 h (black) and at 25°C for 24 h +100°C for 3 h (red)

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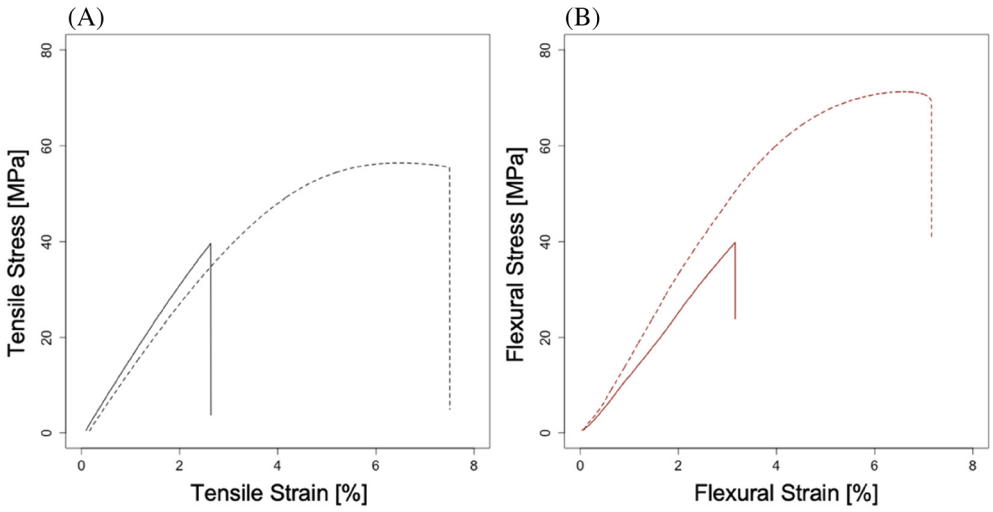

FIGURE 7 (A) Tensile stress versus strain (%) and (B) flexural stress versus flexural strain (%) plot obtained for the samples tested for BBR2.1 (continuous lines) and BBR2.2 (dotted lines) epoxy system.

Sample ID	Flexural strength (MPa)	Flexural modulus (GPa)	Elongation (%)
FBBR2.1	65.09 ± 1.74	5.31 ± 0.12	2.06 ± 0.03
FBBR2.2	73.32 ± 1.49	5.28 ± 0.12	2.05 ± 0.02

TABLE 6 Flexural strength, flexural modulus, and percentage elongations of the flax fiber reinforced biobased resin composites for the two curing cycles (FBBR2.1 at 25°C for 24 h, FBBR2.2 at 25°C for 24 h and post cured for 100°C for 3 h).

reinforced biobased composites cured for 24 h at 25°C, whereas FBBR2.2 denotes the fiber reinforced composites subjected to post-curing for 3 h at 100°C after the 24 h of curing at 25°C. The flexural strength, modulus, and percentage elongation of the flax fiber reinforced sample (FBBR2.1) are 65.09 MPa, 5.31 GPa, and 2.055%. The post-curing improved the flexural strength values of the flax fiber reinforced composite (FBBR2.2) by 16% (73.32 MPa), whereas the modulus (5.28 GPa) and percentage elongation (2.051%) values did not notice any significant improvement. Table 6 represents the flexural strength, modulus, and percentage elongation for the fiber-reinforced composites FBBR2.1 and FBBR2.2. A similar trend is followed for the short beam shear test (ILSS) for the fiber reinforced composites. The FBBR2.1 sample reported the interlaminar shear strength value of 12.03 MPa, and the post-cured sample FBBR2.2 noted an improvement in the values by 33% (16.01 MPa). Both the tests show that the post-curing significantly improved the fiber-reinforced composites bending and interlaminar shear properties. The crosslinking is achieved faster for the post-cured samples, possibly enhancing the composite's strength against the bending and shear loading conditions. Table 7 shows the interlaminar shear strength values of the FBBR2.1 and FBBR2.2 composite samples. A similar trend is noted for the neat resins, and the same discussion applies to the fiber-reinforced composites. The post-curing at a temperature closer to the $T_{g\infty}$ improved

TABLE 7 Interlaminar shear strength values of the flax fiber reinforced biobased resin in two different curing conditions (FBBR2.1 at 25°C for 24 h, FBBR2.2 at 25°C for 24 h and post cured for 100°C for 3 h)

Sample ID	Interlaminar shear strength (MPa)
FBBR2.1	12.03 ± 1.03
FBBR2.2	16.10 ± 0.81

the flexural strength and the interlaminar shear strength values. The flexural modulus and percentage elongation did not achieve a notable improvement. A research study by Jahani et al.[37] also reported that the elastic modulus tends to have a constant value upon post-curing at temperatures below the resin's upper bound value ($T_{g\infty}$).

3.7 | Recycling of flax fiber reinforced biobased recyclable resin composites

The recycled compound and the flax fibers obtained from the chemical recycling process are adequately dried. The recycling yield for both the fiber and the compound is evaluated. The fully drying is verified by monitoring its weight with respect to the drying time. Once the compound's weight is stable over time, it is considered fully dried. The weight versus drying time curve is presented

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in Figure 8. The recycled flax fibers and the compound (rTP101 + 30%wt flax fibers) are weighed and reported in Table 8. Figure 9 displays the results from the chemical recycling process.

The following aspects are taken into account to evaluate the process yields:

- The recycled sample's weight is equal to 20 g, and it has a content of flax fibers equal to 40 wt% (12 g of epoxy matrix and 8 g of flax fibers);
- The recycled compound has a content of flax fibers equal to 30 wt%;

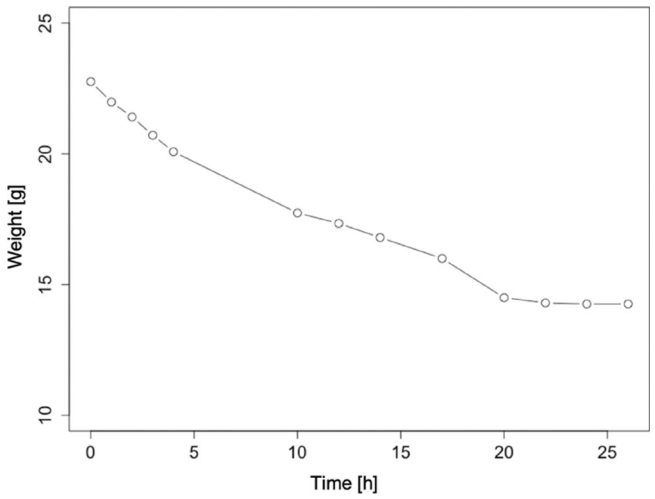

FIGURE 8 Weight versus time plot: monitoring of the drying process.

The results show that 100% of the flax fibers are recovered. The total amount of the fibers added to the neutralized solution is 3.6 g, and the final weight of the re-compound is 15.48 g. The process yield for the rTP101 recycled is 99%, that is, 11.88 g. So, in conclusion, the process yield is higher than the results achieved in the author's previous work.[46] The use of Recyclamine® 101 as the hardener for the Polar bear biobased epoxy resin can transform the recyclable thermoset into a usable thermoplastic. The hardener tailored with acid cleavable group is responsible for this achievement. The polyamine structure with the amino end group connected with the central cleavable group is the chemical structural design of the recyclable epoxy hardener. The yield for the chemical recycling process analyzed is determined by considering the Equation 1 as mentioned below.

$$Yield\,(\%) = \frac{W^{(E)}}{W^{(rTP)}} \times 100 \tag{1}$$

where, the $W^{(E)}$ indicates the initial weight of the epoxy resin recycled and $W^{(rTP)}$ is the final weight of the recycled thermoplastic which is obtained from the implemented chemical recycling process. In the present analysis, the value of $W^{(E)}$ is 12 g and $W^{(rTP)}$ is 11.88 g and the process yield is 99%.

The recycled reinforced thermoplastic (re-compound) obtained from the chemical recycling process is characterized by DMA. The tan δ versus temperature curve is measured at a frequency of 1 Hz, starting from 0°C up to 120°C. The results (Figure 10) clearly show the presence of a single transition region, due to the softening of the

TABLE 8 Recycled flax fibers and compound weight.

Sample (−)	Final weight (g)	Flax fibers weight (g)	rTP101 weight (g)
Re – Flax fibers	8	8	—
Re – Compound	15.48	3.6	11.88

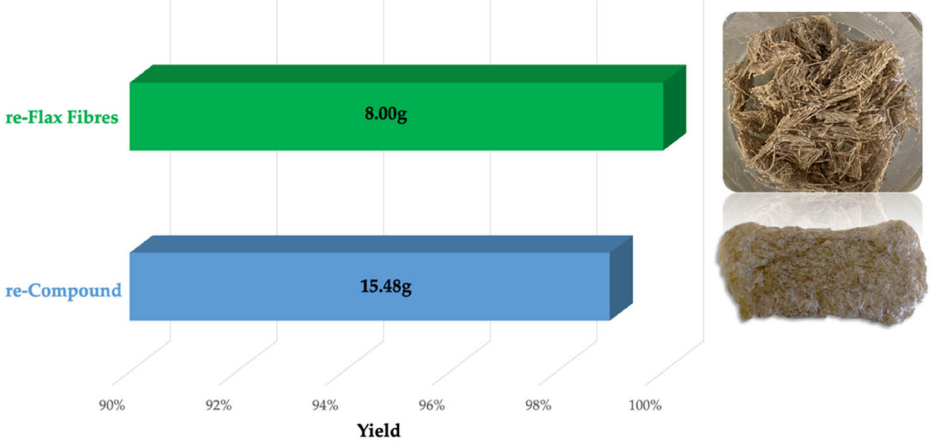

FIGURE 9 Chemical recycling process yields for the re-flax fibers (green bar) and the re-compound (blue bar).

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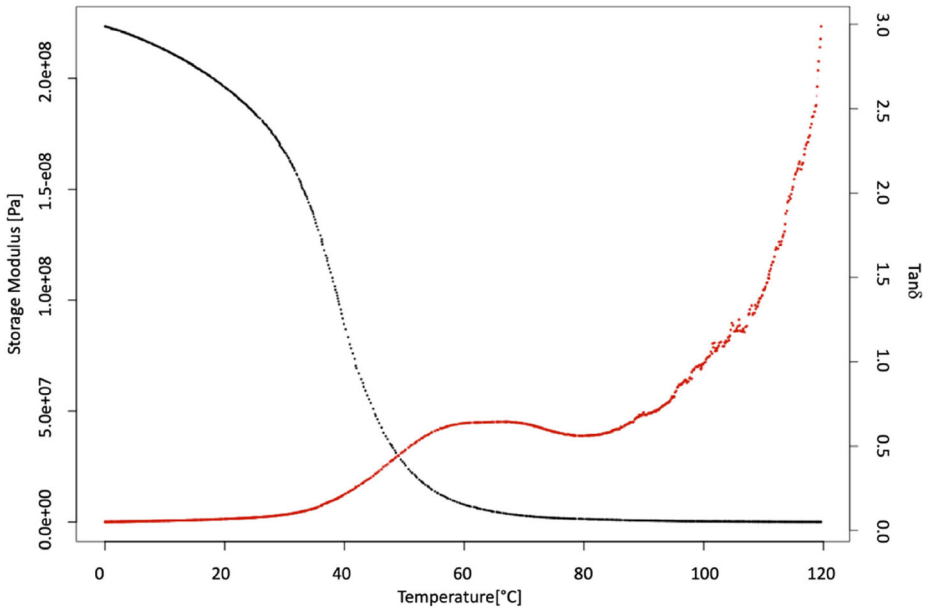

FIGURE 10 Tan δ versus temperature (red curve) and storage modulus versus temperature (black curve) plots were obtained from the DMA test on the recycled reinforced compound.

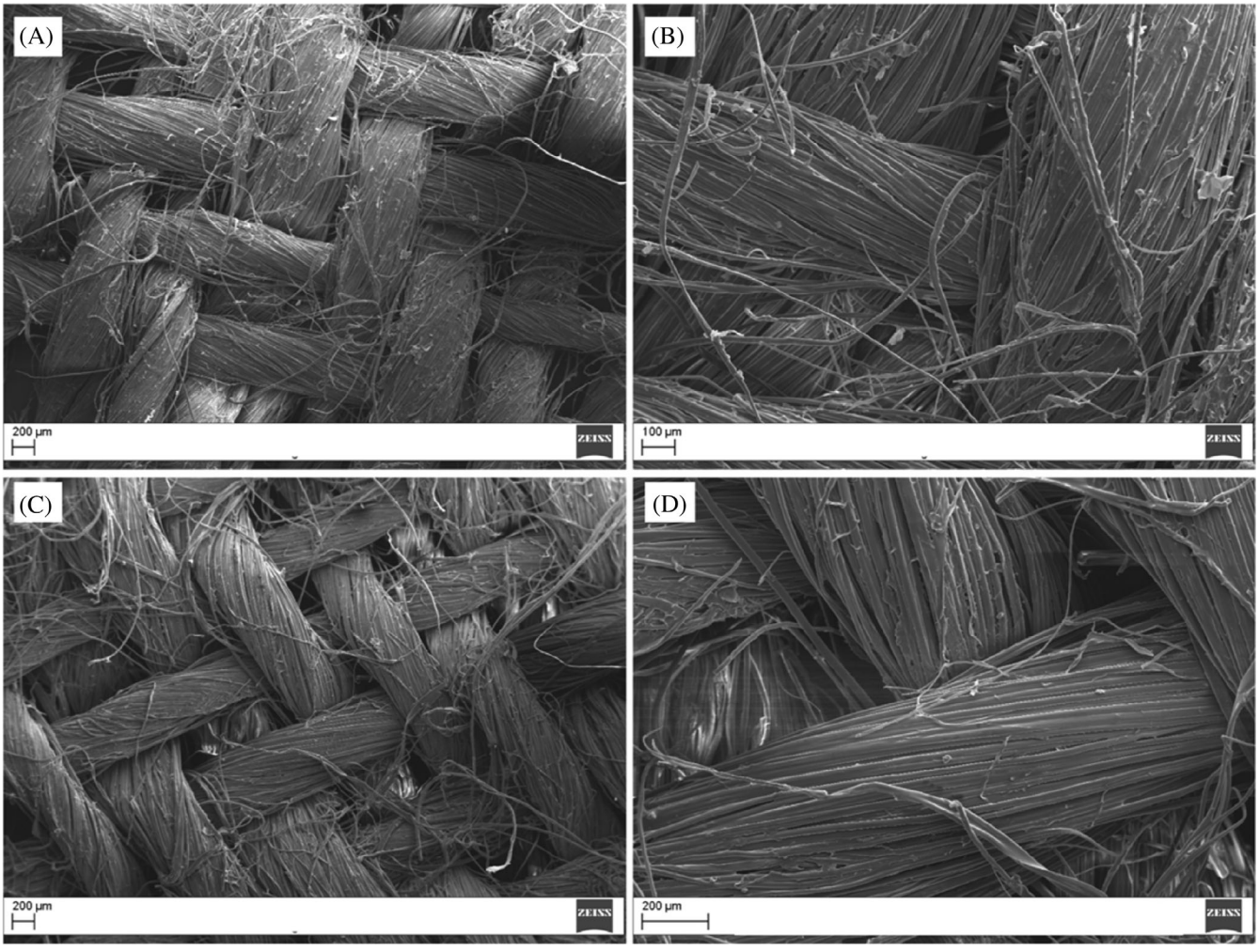

FIGURE 11 SEM morphology of: (A) virgin flax fibers with magnification 50× and (B) 150×; (C) recycled flax fibers with magnification 50× and (D) 150×.

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recycled thermoplastic, with a peak for the tan δ centered at about 63°C. Similar results are obtained by recycling thermoplastic polymers via a chemical process when cleavable amines were used.[46] The storage modulus values resulted in the range between 100 and 200 MPa for lower temperatures, less than 40°C (where a storage modulus drops occurred), Thus ensuring the suitability of the reinforced compound for a potential application as core material for sandwich structure.

The microscopic inspection is carried out using SEM analysis to evaluate any potential damage caused on the surface of the fiber due to the chemical recycling process. There are no visible effects when compared with the virgin fabric (Figure 11). Moreover, the diameter of the flax fibers is not affected by the chemical recycling proposed and implemented, which remained equal to 23.13 ± 2.13 μm before and after the chemical treatment performed in acetic acid solution.

4 | CONCLUSIONS

Two different types of biobased resins, the AMPRO™ BIO and Polar Bear from R*CONCEPT, were experimentally analyzed to target the higher glass transition temperature and improve recyclability. Four different types of curing conditions were attempted to optimize the T_g values. The diluent effect was also analyzed to achieve the best possible T_g value. The DSC results reported that the biobased resin from Polar bear polymer registered better glass transition temperatures among the two sets studied. Further investigations were focused only on the Polar bear polymer resins on the effect of diluent and post-curing, aiming to enhance the glass transition temperature of the polar resin. The post-curing for 3 h at 100°C (after the initial curing for 24 h at 25°C) increased the glass transition temperature of polar resins. Out of the Polar resin samples, the BBR2.1, polar resin without diluent, which was post-cured for 3 h at 100°C, claimed the maximum T_g value as 96.2°C. The flexural and tensile properties of the Polar resins BBR2.1 and BBR2.2 (without diluent) were characterized. The post-cured resin's tensile strength and modulus values (BBR2.2) improved by 32% and 5% compared to the BBR2.1 (uncured) sample. Similarly, the flexural strength and the modulus of the post-cured sample (BBR2.2) improved by 72% and 33%, respectively. The post-curing positively influenced the enhancement of the crosslinking of polymer chains, which was reflected in the improvement in the mechanical properties. The flexural and the interlaminar properties of the flax fibers reinforced polar resin composites also improved with post-curing. For the post-cured composite, the flexural strength values increased by 16% (73.32 MPa), and the modulus and percentage elongation values were 5.28 GPa and 2.051%, respectively. The interlaminar shear strength value of the post-cured sample FBBR2.2 also projected an improvement of 33% (16.01 MPa). The chemical recycling process used as a disposal strategy for the epoxy composite allowed to recover of the initial material with a high recovery yield. Thus, a reinforced thermoplastic composite was obtained, which showed good thermomechanical properties in T_g and storage modulus in a wide range of temperatures (from 0°C up to 40°C) to be potentially used as core materials for sandwich structures. In this way, achieving a twofold goal consistent with the circular economy approach is possible. By following a "design for recycling" path, manufacturing an eco-composite (made of fully-recyclable biobased epoxy resin and natural flax fibers) is attained with good thermomechanical properties; and a "design from recycling" path as a good strategy for reusing the recycled material obtained from the chemical recycling process.

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CONFLICT OF INTEREST

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

The raw/processed data required to reproduce these findings can be shared by the authors upon motivated request.

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