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Balancing Sustainability and Performance: Bio-Based Epoxy Systems for Durable Flax Fiber-Reinforced Composites

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ABSTRACT

Three bio-based epoxy systems—Polar Bear, Green Turtle, and Plankton—featuring bio-carbon contents of 28% to 70%, were combined with recyclable (Recyclamine R101) and non-recyclable hardeners to produce flax fiber-reinforced biocomposites. The Polar Bear system demonstrated superior mechanical and thermal properties, while Plankton with recyclable hardener exhibited incomplete curing and poor mechanical performance. Accelerated salt-fog aging up to 3 months revealed substantial degradation in flexural and interlaminar shear properties within 30 days for most composites, except Green Turtle with recyclable hardener, which maintained mechanical stability and exhibited marked interfacial strengthening even after prolonged exposure. Low-velocity impact tests up to 7.5 J showed no penetration and comparable energy absorption, with aging causing increased compliance and matrix plasticization. SEM analysis identified significant fiber/matrix debonding in Plankton composites, contrasting with preserved interfacial integrity in Green Turtle systems. Thermogravimetric analysis confirmed negligible changes in thermal degradation post-aging. These findings highlight the interplay between bio-carbon content, recyclability, and durability, underscoring the potential of Green Turtle and Polar Bear epoxy systems for sustainable polymer composites with balanced performance and durability.

1 | Introduction

Escalating environmental concerns, notably the urgent need to reduce carbon dioxide emissions and the finite nature of fossil resources, are intensifying the global pursuit of sustainable materials. This imperative is driving significant academic research and development into bio-based polymers, which offer environmentally benign and economically viable solutions compared to conventional petroleum-derived materials [1, 2]. Consequently, the growing interest in sustainable materials has spurred the development and application of eco-friendly options like bio-based thermoset resins as composite matrices. These resins, derived at least partially from renewable resources such as plant oils and lignin, provide a greener alternative to traditional epoxies, retaining their desirable mechanical properties and chemical resistance while lessening the environmental footprint [3, 4]. Among the diverse class of bio-based polymers, epoxy resins derived from renewable resources have garnered considerable and growing attention [5, 6]. This interest stems from their inherent versatility, which allows for tailored mechanical, thermal, and chemical properties through chemical modification and crosslinking strategies [7–10]. Consequently, bio-based epoxies exhibit significant potential for a wide array of applications, particularly as high-performance matrices in composite materials.

Summary

- Novel bio-based epoxy resins are evaluated for their use as a matrix in NFRPs.
- Flax fiber-reinforced composites are manufactured through vacuum infusion.
- The aging resistance of composites against salt-fog is assessed.
- Aging leads to increased compliance and matrix plasticization in all composites.
- Sustainable biocomposites show promising mechanical performance and durability.

These bio-based composites offer the prospect of a reduced carbon footprint and decreased reliance on non-renewable resources, aligning with the principles of a circular economy [11, 12].

To further enhance the overall sustainability profile of these promising materials, the recyclability and end-of-life management of bio-based epoxy systems are also crucial aspects currently under intense investigation. Traditional thermoset epoxy resins pose challenges in terms of reprocessing and recycling due to their cross-linked network structure. Therefore, the development of novel bio-based epoxy chemistries that facilitate easier depolymerization or reprocessing without significant loss of performance represents a critical advancement in achieving truly sustainable composite solutions [13, 14]. Among various recycling strategies for fiber reinforced composites, thermal and mechanical processes remain limited by fiber degradation, especially in natural fibers, and incomplete matrix recovery [15]. In contrast, chemical recycling offers a promising alternative, as it enables the recovery of clean, undamaged fibers and valuable monomers from epoxy matrices [16, 17]. However, conventional approaches often require harsh solvent systems, limiting their use with natural fiber composites. Cleavable amine-based epoxy systems present a sustainable solution, allowing for the recovery of thermoplastics and intact fibers under mild aqueous conditions at low temperatures. This approach has been the subject of numerous recent studies in the literature, which have demonstrated its technical and environmental effectiveness through life cycle assessment [18–22]. Saitta et al. [19] evaluated two bio-based epoxy resins, Ampro Bio and Polar Bear (R*Concept), to enhance glass transition temperature and recyclability. Polar Bear resin exhibited superior T_g values in a no-diluent formulation post-cured at 100°C for 3 h. Post-curing significantly improved mechanical properties, including tensile and flexural strength. Flax fiber-reinforced composites also benefited from post-curing, showing enhanced flexural and interlaminar performance. A chemical recycling process enabled high-yield recovery of the resin, yielding a reinforced thermoplastic with favorable thermomechanical properties, supporting circular economy strategies through both "design for" and "design from" recycling approaches. Cicala et al. [23] produced hybrid flax/carbon fiber laminates using a bio-based epoxy (SuperSapVR 300, by Entropy Resins) and cleavable amine (Recyclamine 301, by Connora Technologies) via High Pressure Resin Transfer Molding (HP-RTM) with a rapid 8-min precure cycle. Mechanical tests showed that placing carbon fibers on the outer layers offered the best balance of flexural strength and impact resistance. The laminates were also chemically recycled in mild acetic acid, yielding clean fibers and a reusable thermoplastic with properties comparable to engineering polymers. Cicala et al. [24] investigated also a different manufacturing process, that is, resin infusion, to produce hybrid flax/carbon fiber laminates using a bio-based epoxy (SuperSap epoxy monomers CLX(S), by Entropy Resins) and the same cleavable amine (Recyclamine 301) as a hardener. Carbon fibers enhanced mechanical strength, while natural fibers improved damping. The best performance was achieved with carbon fibers on the outer layers, also benefiting durability. Chemical recycling in mild acetic acid yielded clean fibers and a reusable thermoplastic with good properties, especially when reinforced with kenaf and processed via injection molding or fused deposition modeling (FDM). In another study, La Rosa et al. [18] mixed a biobased epoxy SuperSap 300 by Entropy Resins with the Recyclamine 301 hardener, exploring carbon fiber reinforced composites made from a green epoxy resin, suitable for automotive structural use. The key innovation was the recyclability of thermoset composites, allowing recovery of clean carbon fibers and thermoplastic polymers. Preliminary life cycle assessment shows environmental benefits from recycling, mainly by reducing the need for virgin PAN fibers. In a recent study, Taheri et al. [25] compared a recyclable basalt–Recyclamine composite with a conventional basalt–epoxy composite under tensile and compressive loading. While conventional epoxy-based composites showed higher tensile strength (18.6%), the recyclable ones offered comparable stiffness and outperformed the conventional ones in compression, with greater stiffness (14.6%) and ductility (3.9% vs. 1.1%). Overall, recyclable epoxy-based composites combined solid mechanical performance with sustainability, making them a viable alternative for structural applications. While these previous studies have demonstrated the feasibility and effectiveness of incorporating such reinforcements into bio-based and recyclable polymer matrices, they have predominantly focused on a single type of bio-based epoxy resin, typically characterized by a limited bio-carbon content of around 20%. These investigations have primarily addressed the static and dynamic mechanical properties of the composites in the virgin state, without exploring the long-term durability of the systems. In this context, the present study aims to advance the current state of the art by characterizing, for the first time, a set of bio-based epoxy resins with bio-carbon contents ranging from 28% to 70%. These resins were cured using both a conventional hardener and a curing agent capable of enabling recyclability at end-of-life. Additionally, the durability of the resulting composite materials was assessed through salt-fog spray aging tests, marking the first attempt to evaluate their environmental resistance.

Before assessing the aging resistance of the composites, a preliminary investigation was conducted to identify the most promising bio-based epoxy systems based on a thorough assessment of their initial mechanical (e.g., quasi static flexural strength and stiffness, impact strength) and thermal (e.g., glass transition temperature, thermal stability) characteristics. Subsequently, fiber reinforced composites were manufactured using the

TABLE 1 | List of epoxy blend formulations.

Material ID	Epoxy system name (part A)	Part A (phr)	Recyclamine R101 (phr)	Non-recyclable hardener (phr)	Reactive diluent (phr)
PB-R	Polar Bear	100	22	0	8
GT-NR	Green Turtle	100	0	30	0
PL-NR	Plankton	100	0	30	0
GT-R	Green Turtle	100	22	0	0
PL-R	Plankton	100	22	0	0

vacuum infusion technique, with the pre-selected bio-based epoxy systems as polymeric matrices and bidirectional flax fabrics as reinforcement.

These composite materials were then subjected to a comprehensive mechanical characterization protocol, encompassing tensile strength and modulus, flexural strength and modulus, interlaminar shear strength (ILSS) (evaluated through short beam shear [SBS] tests), and impact resistance (assessed via low-velocity impact tests). To simulate the aggressive conditions of marine and coastal environments, mechanical tests were systematically conducted on fiber reinforced composite samples exposed to salt-fog for durations of 1, 2, and 3 months. This systematic analysis will provide critical insights into the long-term performance and durability of these sustainable composite materials. Ultimately, the findings of this research aim to provide essential data for assessing the suitability of these bio-based epoxy flax fiber composites for potential adoption in structural and semi-structural applications where significant exposure to saline environments is a critical design consideration, such as in the marine and construction sectors. In the present study, salt-fog testing was selected over alternative aging methodologies (thermal cycling and ultraviolet exposure) due to its superior capability to simulate the distinctive corrosive conditions characteristic of these environments.

2 | Materials and Methods

2.1 | Bio-Based Epoxy Resins

Three epoxy systems (named Polar Bear, Green Turtle, and Plankton) supplied by R*Concept, each having a different bio-carbon content, were mixed with two different amine-based hardeners to obtain novel bio-based and potentially recyclable epoxy resins useful as a matrix for biocomposites with enhanced aging resistance. Specifically, the reference system (hereafter referred to as PB-R) combines the Polar Bear system (having bio-carbon content exceeding 28%), the recyclable hardener Recyclamine R101 (Connora Technologies), and its own epoxy-based reactive diluent, which reduces the system's viscosity for optimal vacuum infusion. The component weight ratio of the reference epoxy system is 100:22:8 (Polar Bear, Recyclamine R101 and reactive diluent, respectively). Recyclamine R101 is an amine-based hardener designed with cleavable bonds, making it possible to create recyclable thermosets. The presence of acid-sensitive units in its chemical structure is crucial, as it allows the crosslink points in the cured epoxy to be broken down, transforming the material into a thermoplastic-based polymer through a chemical treatment performed in an acetic acid solution [19, 20, 23]. The epoxy system used as a reference in this study was already studied by other authors, both with or without the addition of the reactive diluent [19, 21, 22, 26].

Green Turtle and Plankton systems, which have bio-carbon contents greater than 40% and 70% respectively, were first blended with their specific non-recyclable amine-based hardeners at a 100:30 weight ratio to obtain two "standard" epoxy systems (hereafter referred to as GT-NR and PL-NR, respectively). Furthermore, two additional innovative bio-based and recyclable epoxy blends were subsequently prepared by mixing the Green Turtle and Plankton systems with the recyclable hardener Recyclamine R101 in a 100:22 (resin to hardener) weight ratio. Table 1 lists all the investigated blends, including their nomenclature and relative weight compositions. The epoxy systems studied have viscosities compatible with the following processes: hand laminating, infusion, tooling, and casting. Only in the case of the Polar Bear resin was it necessary to add a diluent to make it suitable for the vacuum resin infusion process. Each formulation underwent a vacuum degassing process after mixing with the hardener for 10 min. All epoxy systems were initially cured at room temperature (25°C ± 2°C) for 24 h, followed by different post-curing: Polar Bear at 100°C ± 2°C for 3 h, whereas Green Turtle and Plankton at 80°C ± 2°C for 4 h.

All the cured systems listed in Table 1 were characterized through quasi-static flexural and Charpy impact tests to assess their main mechanical properties. Furthermore, epoxy samples underwent thermal analysis using differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and dynamic mechanical analysis (DMA) to investigate the curing behavior of the resin systems, their thermal stability, and the temperature-dependent evolution of their storage and loss moduli.

Five prismatic samples of each epoxy resin (57.6 mm long, 12.7 mm wide, and 3 mm thick) were subjected to three-point bending tests according to ASTM D790 standard using a Zwick-Roell Z005 universal testing machine equipped with a 5 kN load cell. The crosshead speed and the support span distance were set to 1.3 mm/min and 48 mm, respectively. Impact tests were performed in accordance with ASTM D6110 on rectangular specimens measuring 90 mm in length, 10 mm in width, and 3 mm in thickness, using a CEAST 9050 Charpy pendulum by Instron, equipped with a 5 J instrumented impactor. DSC analysis was performed under nitrogen flow by using a DSC model 214 Polyma by Netzsch. Three specimens (around 10 mg) for

each epoxy system were placed in a Concavus aluminum crucible with a pierced lid and analyzed in accordance with the following thermal program: heating from −20°C to 210°C (5 min hold interval), cooling to −20°C (5 min hold) and second heating to 210°C, all steps carried out with a rate of 10°C/min.

DMA tests were conducted in a three-point bending configuration under dynamic conditions from 30°C to 75°C, with a heating rate of 2°C/min and a frequency of 1 Hz. The tests were performed in triplicate by using a DMA 242E Artemis by Netzsch and the storage modulus (E'), loss modulus (E'') and damping factor ($\tan\delta = E''/E'$) were measured. The thermal stability of the bio-based epoxy systems was investigated by TGA, which was carried out by heating the samples (around 20 mg) in alumina crucibles from room temperature to 800°C under a nitrogen atmosphere and with a heating rate of 10°C/min by using a TG209 F1 Libra by Netzsch.

2.2 | Biocomposites

All the investigated biocomposites were produced using six layers of a balanced twill weave flax fabric, having areal weight of 318 g/m^2, as reinforcement (Lineo). The polymer matrix varied by employing the previously characterized bio-based epoxy systems. Specifically, four composite laminates were exposed to salt-fog spray conditions to assess their behavior under aggressive environments. Indeed, due to its limited mechanical properties revealed in the preliminary characterization of bio-based epoxy resins, the PL-R epoxy system was excluded as a matrix for the bio-based composites, as will be discussed in Section 3. For simplicity, the composites investigated are identified by the following codes: F-PB-R, F-GT-R, F-GT-NR, and F-PL-NR. The prefix "F" denotes the flax reinforcement, while "PB," "GT," and "PL" represent the bio-based epoxy system used as the matrix (Polar Bear, Green Turtle, and Plankton, respectively). The suffixes "R" and "NR" denote whether the epoxy matrix is "recyclable" or "non-recyclable," a distinction determined using the recyclable hardener Recyclamine R101 in the polymer blend. All as-manufactured biocomposites exhibited a nominal thickness of 4 mm and a fiber volume fraction of 0.32.

To evaluate the durability of the bio-based composites, a CC1000iP climatic chamber (Ascott analytical) was used to expose all composite panels to salt-fog spray (5 wt% NaCl solution) at 35°C ± 1°C, in accordance with ASTM B117. Exposure times were set at 1, 2, and 3 months (corresponding to 30, 60, and 90 days). With regard to the selection of the 3-month period, previous studies [27–30] indicated that 2 months are generally sufficient, in the case of composites reinforced with natural fibers, to observe significant variations in mechanical behavior. Building on this evidence, the aging period was further extended to enable a more in-depth investigation into the durability of the developed composites.

Mechanical testing was performed on five specimens of each bio-based composite, including three-point bending tests, SBS tests, and low-velocity impact tests. To further characterize the composites, scanning electron microscopy (SEM) and TGA were employed to assess their microstructural configuration and thermal stability.

Three-point bending tests were performed in displacement control mode using a Zwick-Roell Z005 universal testing machine, equipped with a 5 kN load cell, according to ASTM D790. The support span length was maintained at 64 mm, and the crosshead speed was set to 1.7 mm/min. Tensile tests, following ASTM D3039, and SBS tests, following ISO 1430, were conducted in displacement control mode using a WANCE ETM-C universal testing machine, equipped with a 50 kN load cell. The crosshead speed was 2 mm/min for tensile tests and 1 mm/min for SBS tests.

Low-velocity impact tests were performed on each bio-based composite using an instrumented drop weight impact tower (Instron/Ceast 9340) according to ISO 6603-2. Specifically, the specimens were clamped between two steel plates, leaving a circular unsupported area with a 40 mm diameter. A 12.7 mm diameter hemispherical impactor, weighing 3.05 kg, was used. Square samples (7 cm × 7 cm) were impacted at room temperature at two energy levels (5 and 7.5 J) by adjusting the impactor's release height. The fracture surfaces of the bio-based composites were examined using SEM with a Tescan Mira3. Prior to SEM, the specimens were sputter-coated with gold to ensure electrical conductivity. TGA was performed on approximately 20 mg of biocomposite samples placed in alumina crucibles and heated from 25°C to 500°C at 10°C/min under a nitrogen atmosphere, using a Netzsch TG 209 F1 Libra.

3 | Results and Discussion

3.1 | Bio-Based Epoxy Resins

The main quasi-static flexural and impact properties of the bio-epoxy resins investigated are shown in Figures 1 and 2, respectively. First, this preliminary investigation evidenced that all the innovative systems (those obtained using Green Turtle or Plankton as bio-epoxy) showed lower flexural strength and modulus, and higher deformation at break compared to the reference (i.e., PB-R).

In particular, GT-NR and GT-R systems (obtained mixing Green Turtle with its non-recyclable hardener and with Recyclamine R101, respectively) showed flexural strength decreased by 25% and 59% and flexural modulus by 32.7% and 61.2%, respectively, compared to the reference system (i.e., PB-R). Conversely, the deformation at break of these innovative bio-epoxy systems was found to be 58% and 91% greater than that of PB-R.

Different results were achieved by using Plankton as a bio-based epoxy system. Indeed, the differences between PL-NR and the reference system (PB-R) were less pronounced than those observed with the GT-NR system: −14.1%, −2.4%, and +18.5% for flexural strength, flexural modulus, and deformation at break, respectively. Conversely, combining Plankton (which, as previously mentioned, exhibits the highest bio-carbon content among the investigated bio-based systems) with the recyclable hardener, intended to enable recyclability in this innovative epoxy system, led to dramatic decreases in flexural properties. In particular, the PL-R epoxy system exhibited negligible flexural strength and modulus values (2.0 MPa and 0.021 GPa, respectively), making it completely unsuitable as a polymeric matrix

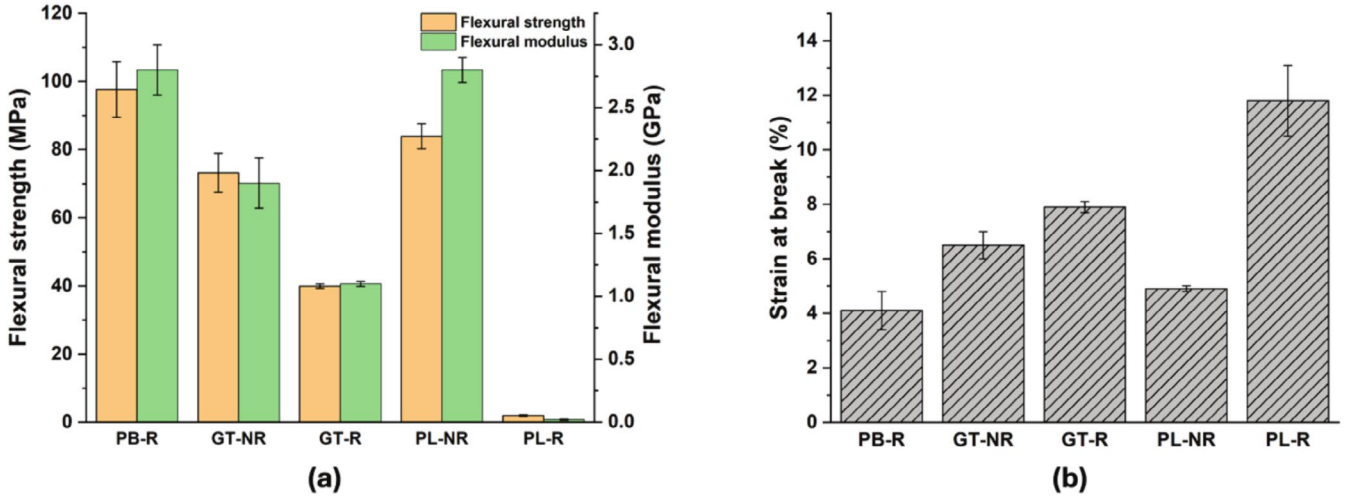

FIGURE 1 | (a) Strength, modulus, and (b) strain at break during bending tests on the different neat epoxy systems.

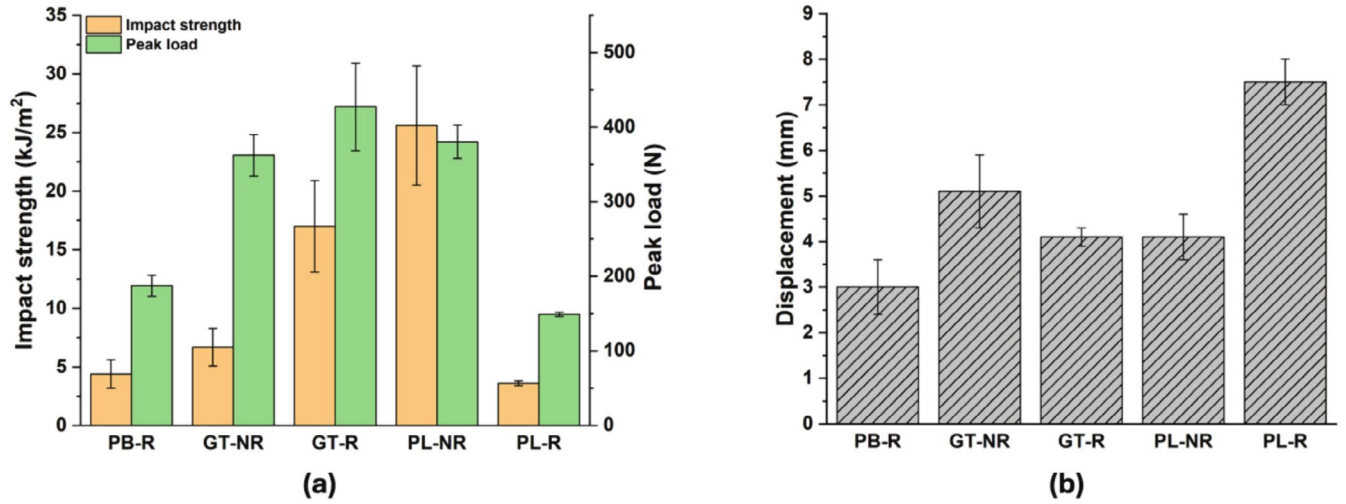

FIGURE 2 | (a) Impact strength, peak load, and (b) displacement during Charpy impact tests on the different neat epoxy systems.

for biocomposites. Furthermore, this epoxy system displayed a strain at break of 11.8%, a value notably exceeding the typical range for fully cured thermoset polymers [31].

These findings can be ascribed to the incorrect or incomplete cure between Plankton and Recyclamine R101, mainly due to the chemical structure of this epoxy system. Plankton, unlike the Polar Bear and Green Turtle systems, is bisphenol-free and likely possesses more than two epoxy rings within its polymer chain. In contrast, both Polar Bear and Green Turtle are DGEBA-based epoxies, characterized as diepoxies by the presence of two terminal epoxy rings as functional groups. Since the recyclable hardener is specifically designed to cure with DGEBA monomers, it likely did not react properly with a different epoxy monomer, such as Plankton, resulting in a thermoset epoxy resin with a relatively low cross-linking density.

The quasi-static mechanical behavior exhibited by the different bio-epoxy systems is in full agreement with the results obtained from the Charpy impact tests. Indeed, all the innovative bio-based epoxy systems showed higher impact strength compared to the reference (i.e., PB-R system), except for the PL-R system, as reported in Figure 2. In more detail, the impact strength values of the GT-NR, GT-R, and PL-NR systems were 34.7%, 74.4%, and 83% greater than the reference, respectively. The increased impact strength exhibited by these bio-epoxy systems, compared to the PB-R system, is related to improvements in their peak load and maximum displacement (i.e., displacement at peak load).

On the other hand, the bio-epoxy system obtained by mixing Plankton with the recyclable hardener (PL-R) was the only one to show a reduced impact strength (−20%) compared to the reference system. Despite its higher maximum displacement (7.5 vs. 3 mm), the PL-R system exhibits a lower average peak load (149.1 N) in comparison to the PB-R reference system (182.4 N), leading to reduced impact strength. Consistent with the quasi-static flexural testing results, this finding confirms that, from a mechanical point of view, the PL-R system cannot be considered suitable as a polymeric matrix for biocomposites reinforced with natural fibers.

The DSC analysis highlighted how the combination of different epoxy systems with their respective curing agents significantly influenced the glass transition temperature values (T_g),

which are summarized in Table 2. Both standard epoxy systems, namely GT-NR and PL-NR, exhibited comparable glass transition temperatures in the range 60°C–75°C, which are in line with those of the reference system (PB-R), though the Plankton system displayed a slightly higher temperature. On the other hand, the use of recyclable hardener (Recyclamine R101) induced a significant reduction in T_g for both systems (GT-R and PL-R), especially for the Plankton epoxy system, confirming the results of bending and impact tests. Indeed, the calorimetric tests conducted on the epoxy systems obtained from the mixing of the Plankton system and recyclable hardener show a glass transition temperature for the cured material equal to about 19°C, which is noticeably lower than typical values for epoxy resins cured at room temperature (e.g., exceeding 40°C).

This trend is in line with the results achieved through DMA. Typical trends of the storage modulus (E') and damping factor ($Tan\delta = E''/E'$) are shown in Figure 3, while specific parameters are reported in Table 2. Due to the significant reduction in glass transition temperature below room temperature for PL-R, they were so compliant that it was impossible to test them in a three-point bending configuration. Furthermore, also in these tests, the use of a recyclable hardener negatively affected the glass transition temperature and the ability of the GT-R resin to withstand relatively high temperatures. In any case, standard epoxy systems (GT-NR and PL-NR) show comparable glass transition temperatures and storage moduli, with a slightly higher performance offered by the Plankton-based system (PL-NR).

The thermal stability was investigated by TGA. The main results are reported in Table 3 while representative curves are shown in Figure S1. It was found that Green Turtle and Plankton displayed different degradation patterns, with a higher thermal stability offered by Green Turtle, while in this case, the presence of the recyclable hardener only results in a

TABLE 2 | Glass transition temperatures and storage modulus (E') at 50°C for all the epoxy resin formulations as evaluated by DSC and DMA.

Material ID	T_g (°C)—DSC	T_g (°C)—DMA	E' (MPa)—DMA
PB-R	67.9 ± 0.2	72.1 ± 0.1	2340.3 ± 21.4
GT-NR	63.7 ± 0.2	54.7 ± 0.3	722.6 ± 10.9
GT-R	31.0 ± 0.2	41.2 ± 0.3	125.7 ± 8.4
PL-NR	76.3 ± 0.2	57.8 ± 0.4	823.2 ± 10.7
PL-R	18.6 ± 0.3	N/A	N/A

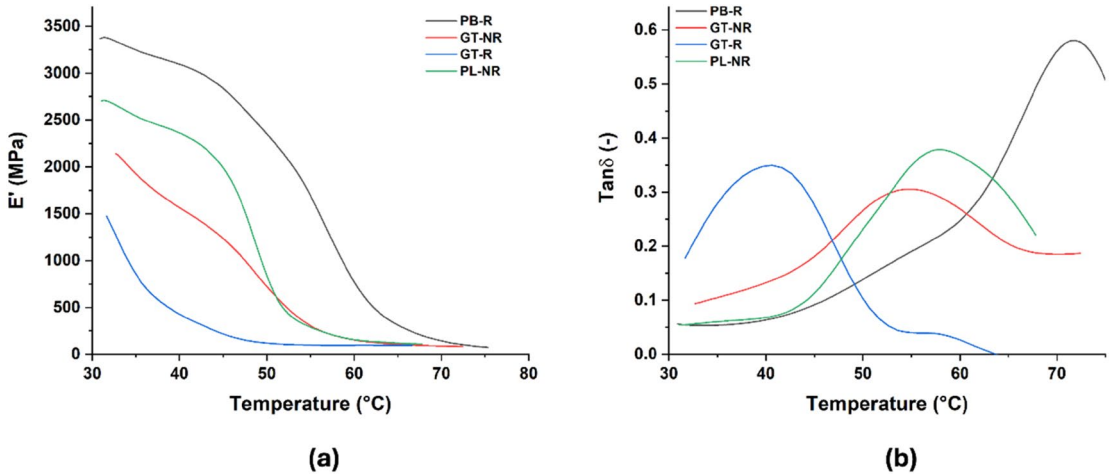

FIGURE 3 | Evolution of (a) storage modulus (E') and (b) $Tan\delta$ as a function of temperature as obtained from DMA tests.

TABLE 3 | Onset, peak degradation, and 5 wt% loss temperatures, and residual mass for all the epoxy resin formulations as evaluated by TGA.

Material ID	Onset temperature (°C)	Peak temperature (°C)	$T_{5\%}$ (°C)	Residual mass (%)
PB-R	355.8 ± 0.4	378.2 ± 0.5	337.4 ± 0.4	10.6 ± 0.3
GT-NR	311.9 ± 0.4	331.9 ± 0.5	305.9 ± 0.4	7.3 ± 0.2
GT-R	321.3 ± 0.5	358.3 ± 0.4	251.6 ± 0.5	6.7 ± 0.4
PL-NR	269.3 ± 0.3	272.5 ± 0.4	270.4 ± 0.4	7.2 ± 0.4
PL-R	268.3 ± 0.5	274.2 ± 0.3	260.6 ± 0.4	11.1 ± 0.3

reduction of the initial degradation temperature. Specifically, all Plankton samples showed an earlier and steep degradation step, while the degradation peak is much broader in Green Turtle samples.

Except for the Plankton systems, which exhibited lower thermal stability, the other epoxy systems showed onset degradation temperatures and maximum rates of decomposition consistent with values reported in the literature for both conventional [32–34] and bio-based epoxy resins [35, 36].

Overall, based on the main findings from this preliminary investigation, the PL-R system is the only bio-based epoxy system among those compared that will be excluded from biocomposite manufacturing due to its very low mechanical and thermal performance. This is likely attributable to an incomplete curing reaction between the recyclable hardener (Recyclamine R101) and Plankton epoxy system, resulting in a thermoset resin with limited cross-linking.

3.2 | Biocomposites

As already stated, among the bio-based epoxy systems investigated, only PL-R was not considered as a polymeric matrix for bio-composites. Therefore, the mechanical behavior and the thermal stability of four bio-based composites exposed to salt-fog spray conditions for up to 3 months (F-PB-R, F-GT-NR, F-GT-R, F-PL-NR) were investigated.

3.2.1 | Three-Point Bending

Figure 4 illustrates the evolution of the flexural properties (i.e., strength, modulus, and strain at break) of the compared biocomposites as a function of exposure time to salt-fog.

Starting from the flexural response of the unaged biocomposites, it was found that F-GT-R samples, manufactured with the recyclable polymeric matrix having intermediate bio-carbon content (> 40%), exhibited approximately half the mechanical properties (both flexural strength and modulus) compared to other investigated biocomposites. Furthermore, the F-PL-NR laminates, manufactured with a non-recyclable polymeric matrix containing the highest bio-carbon content (> 70%), demonstrated the best unaged performance among the investigated bio-based composites (i.e., 83.5 MPa and 4.4 GPa in terms of flexural strength and modulus, respectively).

Overall, bio-based composites derived using standard epoxy resins (GT-NR and PL-NR) as the matrix exhibited mechanical performance in their unaged state (i.e., prior to salt-fog exposure) comparable to those manufactured with the reference epoxy matrix (PB-R). These results indicate that, despite the increased bio-carbon content in the polymeric matrix (exceeding 40% and 70% for the Green Turtle and Plankton systems, respectively) compared to over 28% in the reference system, the mechanical response of the unaged biocomposites did not exhibit any significant reduction.

On the other hand, the use of Recyclamine R101 as a recyclable hardener for the Green Turtle epoxy system in the polymeric matrix negatively affected the mechanical properties of the resulting laminates (F-GT-R), which showed the lowest flexural strength and modulus values (i.e., 42.7 MPa and 2.3 GPa, respectively). These findings are quite predictable, being in full agreement with those observed for neat polymeric matrices (i.e., unreinforced), as detailed in the previous paragraph.

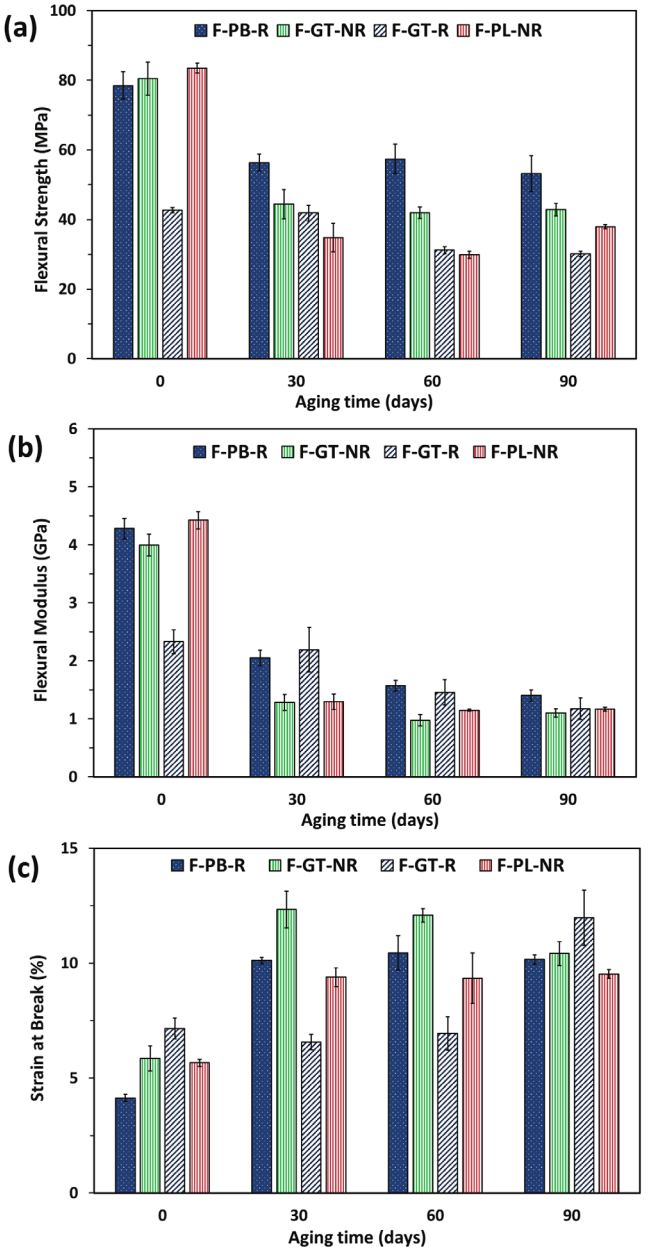

FIGURE 4 | Three-point bending test results for all bio-based composites subjected to 3 months of salt-fog exposure: (a) flexural strength, (b) modulus, and (c) strain at break.

Concerning the aging resistance, F-PB-R, F-GT-NR, and F-PL-NR laminates, while exhibiting high flexural properties in their unaged condition, demonstrated a significant decrease in mechanical stability after only 1 month of salt-fog exposure. In more detail, during the first month of aging, F-PB-R composites exhibited reductions in flexural strength and modulus of 28.3% and 52.1%, respectively. Furthermore, the flexural strength of F-GT-NR and F-PL-NR samples was reduced by

44.8% and 58.3%, respectively, and their flexural modulus by 67.9% and 70.7%. Accordingly, these bio-based composites exhibited improved deformability after the first month of aging. Specifically, the strain at break values for F-PB-R, F-GT-NR, and F-PL-NR laminates increased from 4.1% to 10.1%, 5.8% to 12.3%, and 5.7% to 9.4%, respectively. These results clearly demonstrated that plasticization of the matrix, leading to reduced material stiffness and increased flexibility, was the main degradative mechanism due to salt-fog for F-PB-R, F-GT-NR, and F-PL-NR laminates, observed already within the first 1 month of exposure. Indeed, the exposure of thermoset-based composites to humid environments often results in a physical and reversible degradation termed plasticization [37]. This phenomenon is attributed to the enhanced macromolecular chain mobility resulting from water absorption within the epoxy matrix. Specifically, this process is predominantly observed in amine-based epoxy materials due to their significant water absorption tendency [38].

Conversely, the F-GT-R laminates surprisingly exhibited the best stability against salt fog, demonstrating, after 30 days of exposure, only slight decrements in flexural strength and modulus compared to their initial values (i.e., 1.9% and 6.1%, respectively). Regarding flexural deformability, the F-GT-R laminate was able to retain its strain at break during the first month of aging, unlike other bio-based composites that experienced noticeable plasticization of the matrix.

Prolonging the salt-fog exposure time to 2 and 3 months (60 and 90 days, respectively) led to reductions in mechanical performance, such as flexural strength and modulus, for all bio-based composites. Nevertheless, it is worth noting that after the initial noticeable drop in these flexural properties observed during the first month of aging, F-PB-R, F-GT-NR, and F-PL-NR laminates showed no further significant performance degradation over the remaining 60 days of salt-fog exposure. Specifically, at the end of the aging campaign (90 days), the average flexural strength values of the F-PB-R, F-GT-NR, and F-PL-NR laminates were reduced by 5.6%, 5.3%, and 10.2%, respectively, in comparison to 30-day aged samples. Furthermore, these laminates experienced modulus reductions of 31.6%, 16%, and 9.9% in the same time frame (i.e., between 30 and 60 days of salt-fog exposure). Overall, 90-day aging led to global reductions in flexural strength for F-PB-R, F-GT-NR, and F-PL-NR laminates of −32.3%, −46.8%, and −54.5%, respectively, when compared to unaged samples. Correspondingly, their flexural moduli decreased by −67.2%, −72.3%, and −73.6%. The slight reductions in the flexural strength and modulus exhibited by F-PB-R, F-GT-NR, and F-PL-NR during the latter part of the aging campaign (i.e., between 1 and 3 months of salt-fog exposure) were coupled with a relatively constant trend in the strain at break for all these bio-based composites. These results can be considered primarily governed by the main degradative phenomenon (i.e., matrix plasticization), which appears to play a significant role in mitigating some internal damage due to a deterioration in interfacial adhesion, as will be shown by SEM analysis.

More surprisingly, the Green Turtle-based composites with recyclable hardener (F-GT-R) not only retained their mechanical performance after the first month of aging but also exhibited only gradual declines in properties even after 60 and 90 days of salt-fog exposure. After 90 days, for example, flexural strength and modulus showed reductions of only 29.5% and 49.6%, respectively, in comparison to unaged samples.

In terms of deformability, this laminate showed a dramatic increase in strain at break during the last 1 month of salt-fog exposure: from 6.9% for 60-days aged samples to 12.2% for 90-days aged samples, evidencing the occurrence of a more intense and time-delayed plasticization compared to other investigated bio-based composites.

Overall, all these findings clearly highlighted that, despite their lower initial mechanical properties, the Green Turtle-based composites (containing over 40% bio-carbon content) with the recyclable hardener demonstrated the best aging resistance against salt-fog among the investigated bio-based composites.

3.2.2 | Short Beam Shear

Figure 5 compares the average values (and the related standard deviations) of ILSS for biocomposites aged under salt-fog spray conditions for up to 3 months. Given that the SBS test is specifically designed to minimize flexural stresses while maximizing induced shear stresses in composite materials, thus enabling the characterization of their interlaminar failure resistance, ILSS is considered a key parameter that provides crucial information about fiber-matrix interface strength [39]. Therefore, data reported in Figure 5 highlight the effect of short-term aging on the quality of fiber-matrix adhesion in the biocomposites.

First, it can be stated that the ILSS values of unaged biocomposites generally reflected their flexural properties. More specifically, prior to salt-fog exposure, F-GT-NR laminates displayed the highest ILSS values (11.9 MPa), followed by F-PL-NR (9.22 MPa), F-PB-R (9.10 MPa), and F-GT-R, which showed the lowest (6.86 MPa).

Similar to their flexural behavior, the investigated biocomposites exposed to salt-fog showed a noticeable difference in the impact of exposure on their ILSS between the initial 30 days of aging and that observed during the subsequent 60 days. Indeed, during the first month of aging, the reduction percentage in

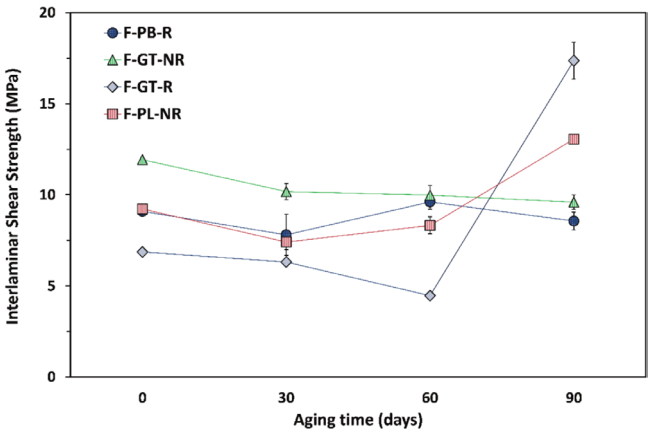

FIGURE 5 | Interlaminar shear strength (ILSS) values of all bio-based composites over 3 months of salt-fog exposure.

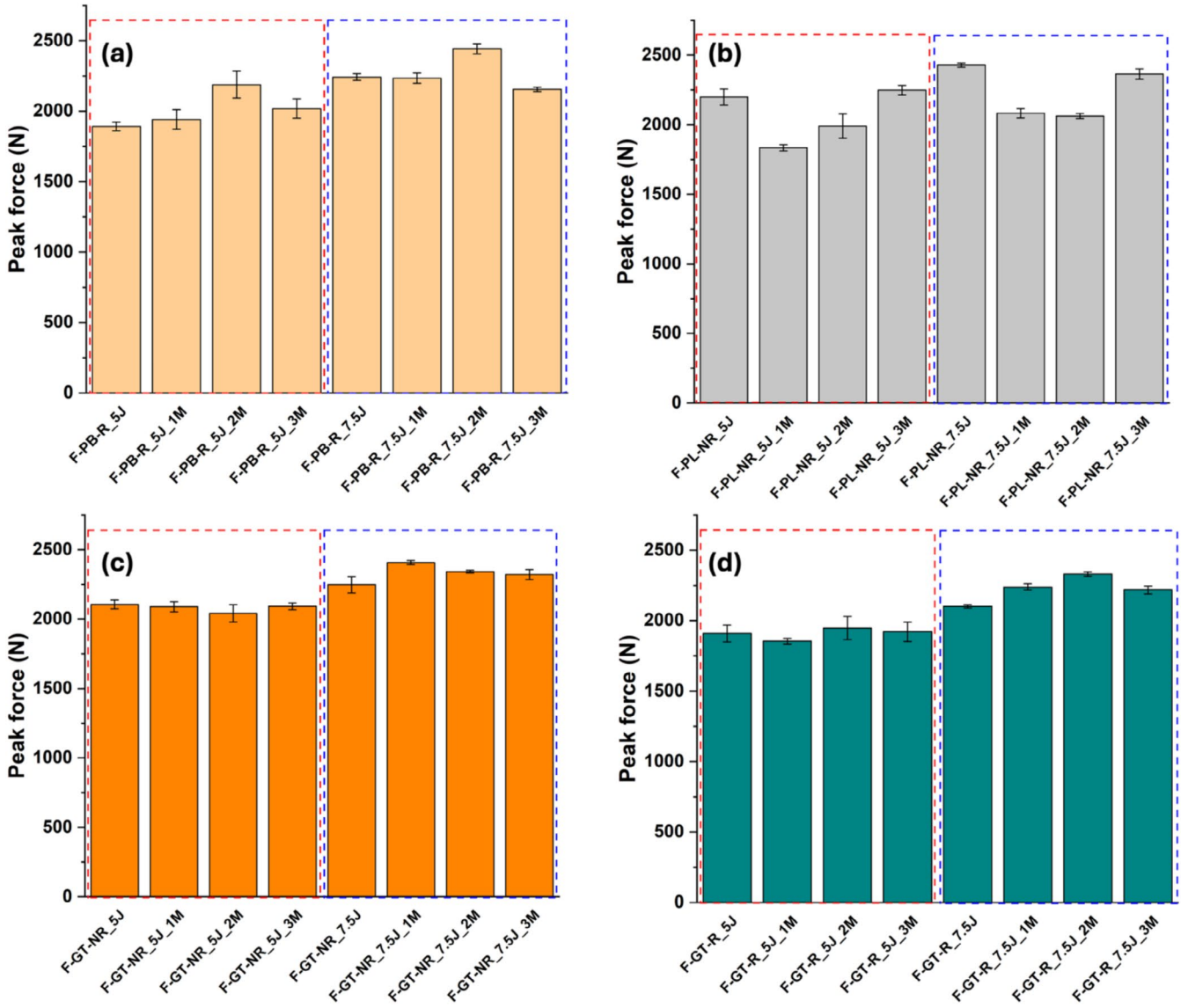

FIGURE 6 | Evolution of peak force as a function of aging time and impact energy for: (a) F-PB-R, (b) F-PL-NR, (c) F-GT-NR, (d) F-GT-R laminates.

terms of ILSS observed for F-PL-NR, F-GT-NR, F-PB-R, and F-GT-R laminates, in comparison to their unaged samples, was equal to 19.7%, 14.7%, 14.2%, and 8.7%, respectively. Therefore, it was evident that, although the unaged F-GT-R composite exhibited the lowest interlaminar properties among the biocomposites, its interlayer interface resistance was less affected by salt-fog exposure, at least during the first month. These variations in the ILSS experienced by the biocomposites during the early stage of the aging campaign mirrored the flexural results, thereby justifying the reductions in flexural properties recorded after 1 month of salt-fog exposure.

However, the extended aging beyond 30 days highlighted different and quite unpredictable ILSS trends for the investigated bio-based composites. Specifically, the ILSS of all biocomposites increased with salt-fog exposure time. F-PB-R and F-PL-NR laminates exhibited this surprising behavior as early as 60 aging days, showing increases of 23.1% and 12.3%, respectively, compared to 30-day aged samples. It is worth noting that for F-GT-R samples, this effect was more pronounced and temporally delayed, with ILSS values increasing only after 3 months (90 days) of salt-fog exposure, showing a dramatic 289% increase compared to 60-day aged samples.

The inverting trend observed can be attributed to the improved deformability of bio-based composites after salt-fog exposure, as ILSS is also sensitive to bulk matrix properties [40]. Specifically, as corroborated by the strain at break values exhibited by bio-based composites under flexural loads, the main degradative effect of salt-fog exposure is the plasticization of the matrix, rather than the weakening of the interlayer interface due to the occurrence of some internal damage, regardless of the polymer system used.

3.2.3 | Low-Velocity Impact

Figure S2 and Figures 6–8 include both the representative force-displacement curves and the main parameters related (peak load, maximum displacement, absorbed energy and damage degree) to the low-velocity impact (LVI) tests as a function of salt-fog exposure time, respectively. Damage degree is evaluated

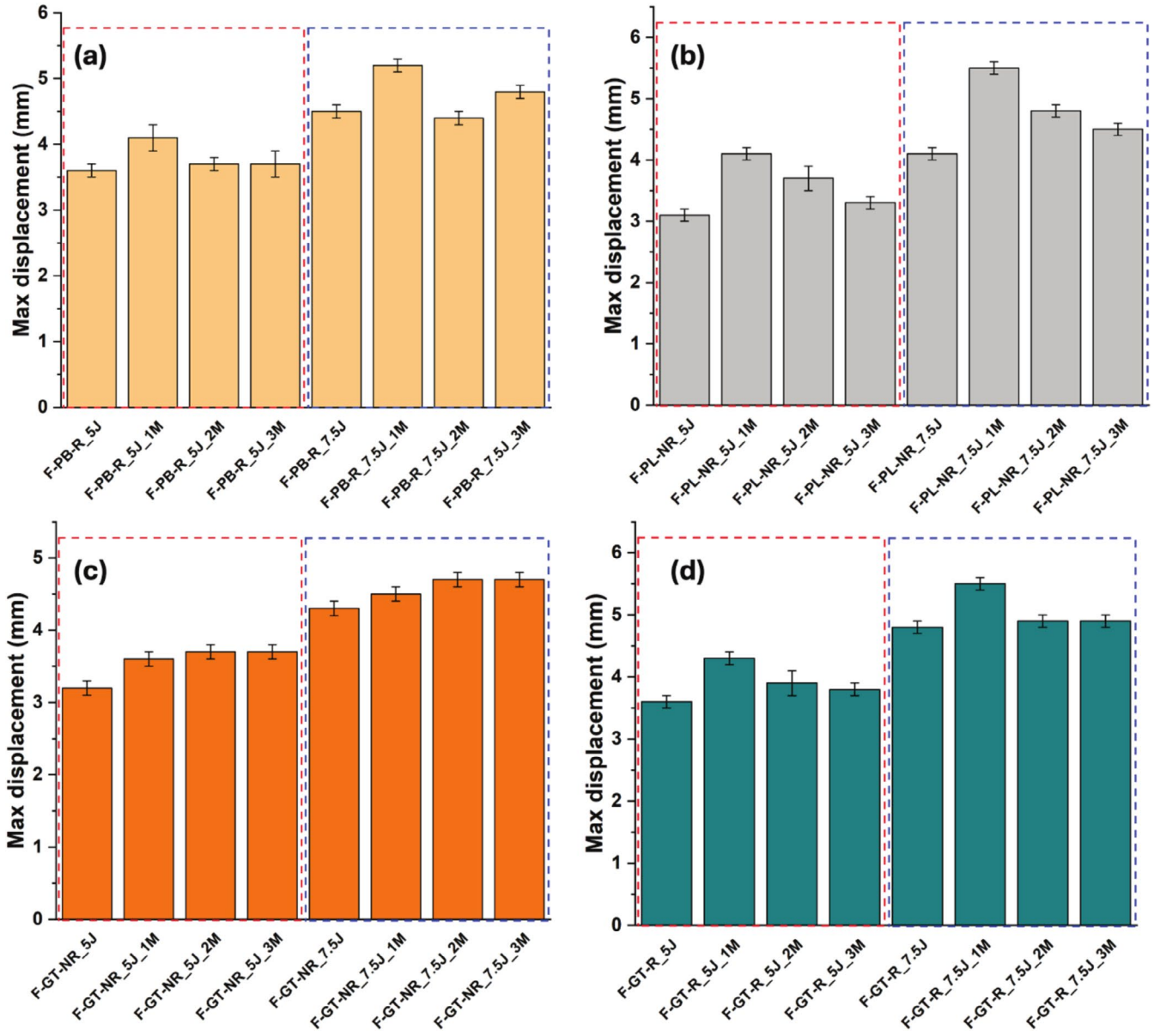

FIGURE 7 | Evolution of maximum displacement as a function of aging time and impact energy for: (a) F-PB-R, (b) F-PL-NR, (c) F-GT-NR, (d) F-GT-R laminates.

as the ratio between absorbed energy and impact energy. The graphs were deliberately given an offset on the x-axis to facilitate their visualization.

From the results, it is evident that in no case was penetration of the laminates achieved, as the curves are closed. Overall, the performance of the various bio-based composites in relation to impact energy is very similar. The Green Turtle-based composites with recyclable hardener (F-GT-R_5J and F-GT-R_7.5J) demonstrated the worst mechanical properties in terms of maximum force and damage degree compared to the other configurations in the unaged condition, although such performance appears to be minimally affected by 1 month of aging, in line with the results from quasi-static mechanical tests. However, exposure to salt-fog for 1 month proved particularly severe for the Plankton-based composites (i.e., F-PL-NR_5J_1M and F-PL-NR_7.5J_1M), which showed the greatest increases in damage degree.

For all the configurations analyzed, consistent with the data from quasi-static mechanical tests, aging resulted in an increase in the compliance of the laminates, as indicated by a significant increase in the maximum displacement of the impactor and a decrease in linear stiffness.

This explains the greater absorbed energy after exposure, which can be attributed to a plasticization of the matrix, a phenomenon evident from the observation of the sample surfaces after the impact tests, as shown in Figures S3–S10.

It is worth noting that in all cases, after aging, there was a significant reduction in the extent of the indentation on the impacted face and the severity of cracks on the face opposite to the impact, confirming greater plasticity of the composite and the occurrence of some internal damage due to a deterioration in interfacial adhesion, which was confirmed by SEM analysis. Indeed, Figures 9–16, by way of example, display the micrographs of all

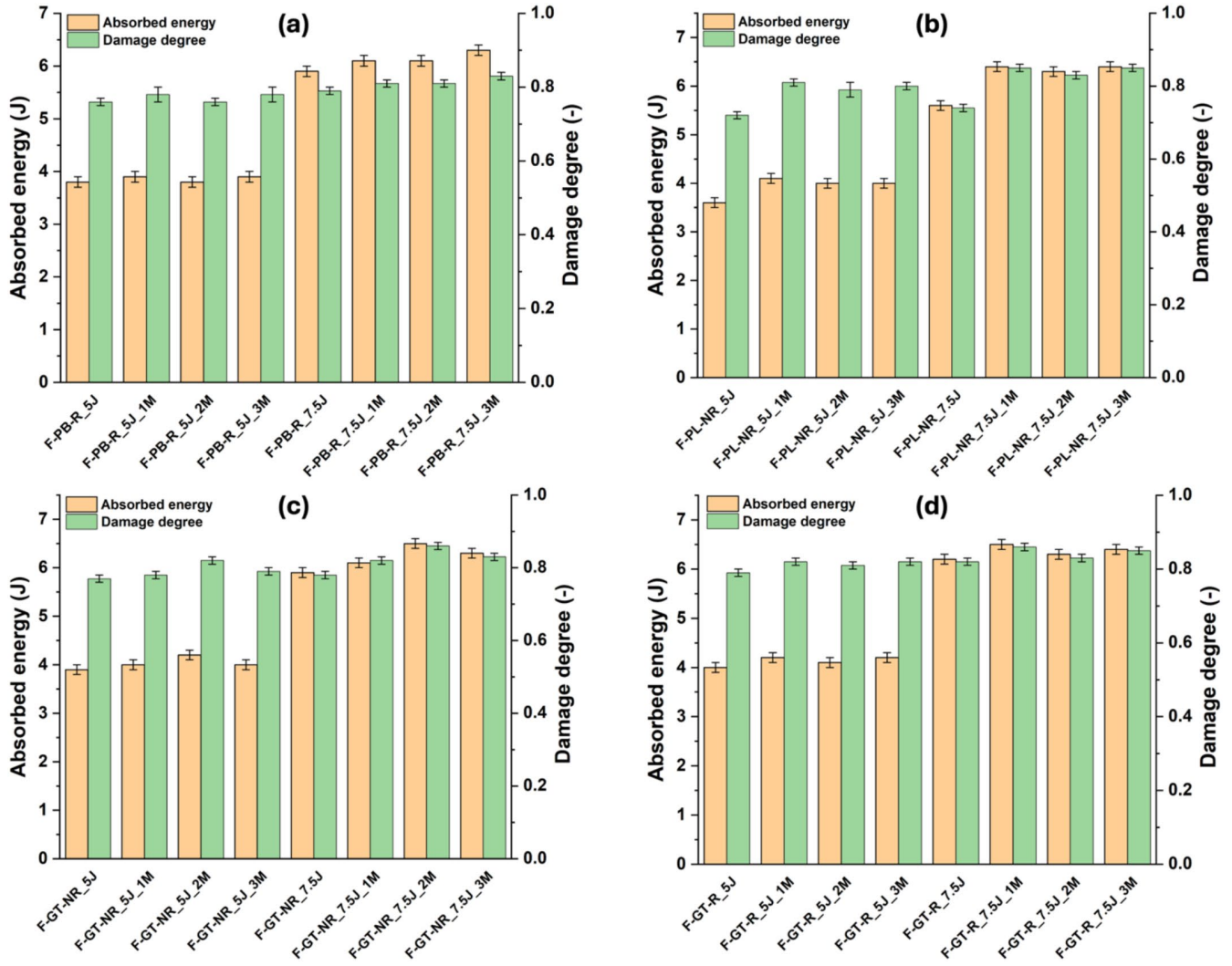

FIGURE 8 | Evolution of absorbed energy and damage degree as a function of aging time and impact energy for: (a) F-PB-R, (b) F-PL-NR, (c) F-GT-NR, (d) F-GT-R laminates.

formulations under both the reference condition and after 1-month exposure to salt-fog. All bio-based composites exhibited the presence of pull-out and debonding, although the most severe damage in this regard was observed in the Plankton-based composites (i.e., F-PL-NR), while signs of fiber damage were detected in the Polar Bear-based composites (i.e., F-PB-R). It is worth noting that the Green Turtle-based composites with recyclable hardener (i.e., F-GT-R) appear to be minimally affected by 1 month of aging, showing fibers still covered by the matrix after pull-out, consistent with the good residual properties observed in both quasi-static and dynamic tests. Despite their lower environmental impact, natural fiber composites demonstrate inferior long-term performance compared to synthetic fiber composites, primarily due to their hydrophilic nature [41]. The long-term durability of flax fiber composites has been extensively examined using accelerated aging protocols under a range of environmental conditions, including hygrothermal exposure, salt-fog spray, UV/weathering, and soil burial [29, 42–44].

These studies consistently report reductions in mechanical performance post-exposure, with the severity of degradation largely influenced by aging parameters such as UV irradiance intensity, temperature, and duration. Given the lignocellulosic nature of flax fibers, they exhibit high sensitivity to moisture and are particularly susceptible to degradation resulting from the synergistic effects of UV radiation, heat, and humidity [45, 46]. Specifically, flax fibers are highly susceptible to moisture during weathering cycles, resulting in swelling-related stress and deterioration at the fiber/matrix interface. The alternating exposure to wet and dry conditions might lead to continuous moisture absorption and release [41]. As the aging time progressed, no penetration was observed for any laminate at an impact energy of up to 7.5J. Overall, the bio-based composites demonstrated comparable impact energy absorption characteristics. In the case of F-PB-R and F-GT-NR laminates, the peak load remained essentially stable up to 3 months of aging. At both energy levels (5 and 7.5 J), the laminates exhibited good compliance, with deformation behavior was primarily governed by matrix plasticization. This mechanism appears to play a significant role in mitigating internal damage. Nonetheless, an increase in the damage degree was recorded, particularly at the higher energy level of 7.5J. The F-PL-NR laminates exhibited a distinct behavior, characterized by a progressive reduction in overall deformability from 2 to 3 months of aging. This trend is likely attributable to a dynamic interplay between matrix plasticization and post-curing

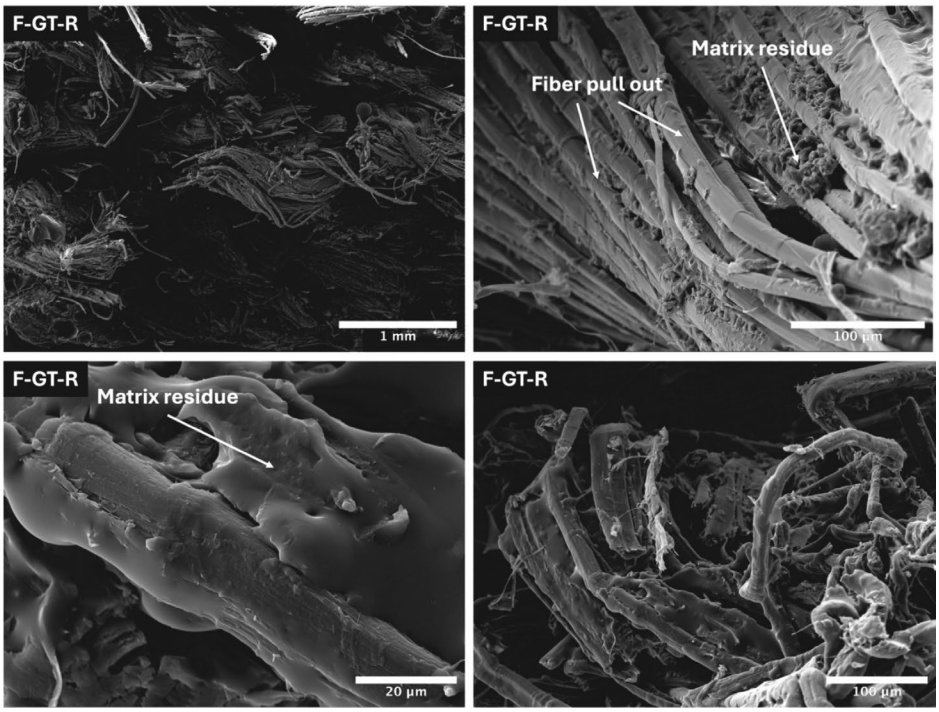

FIGURE 9 | SEM micrographs detailing the fracture surface of F-GT-R laminates.

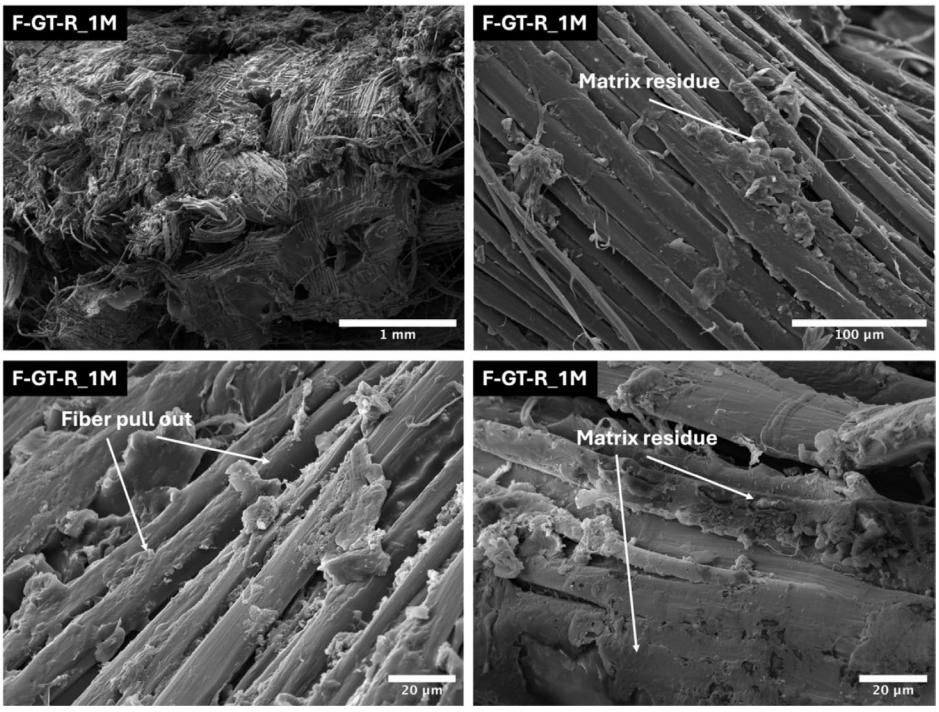

FIGURE 10 | SEM micrographs detailing the fracture surface of F-GT-R laminates after aging.

phenomena as the aging time increases. Nevertheless, at both 5 and 7.5 J impact energies, the damage degree is higher than that of the virgin (unaged) samples, although it remains approximately constant between 1 and 3 months of exposure. It is important to highlight that, in all instances, aging resulted in a marked reduction in the indentation depth on the impacted surface and a decrease in crack severity on the opposing face. This observation confirms an enhancement in the composite's plasticity, alongside the development of internal damage ascribed to a progressive degradation of interfacial adhesion. The same general behavior was observed for the F-GT-R laminates, although, particularly at 5 J, the damage degree is notably higher compared to all other configurations. This can be attributed to increased plasticization, which results in a more compliant composite structure. No significant differences were observed in this regard when compared to the other laminate configurations. It is worth noting, however,

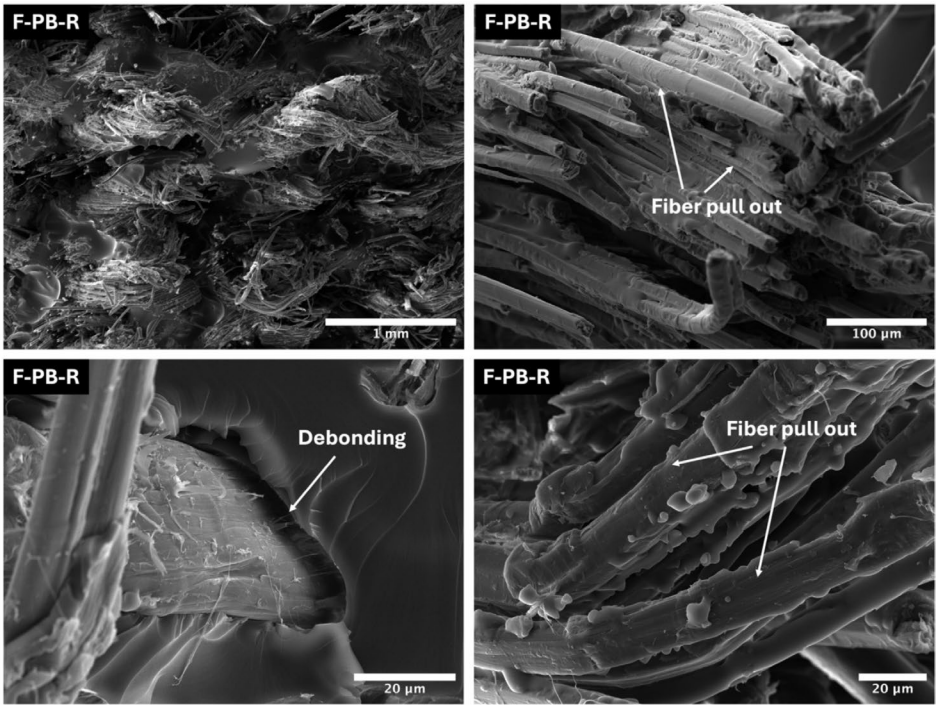

FIGURE 11 | SEM micrographs detailing the fracture surface of F-PB-R laminates.

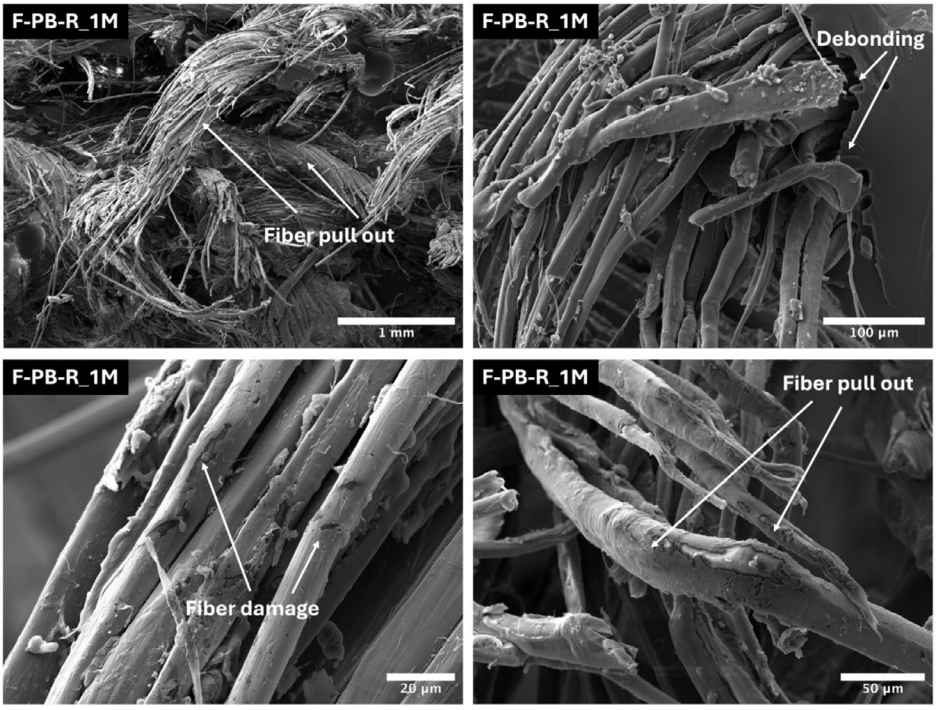

FIGURE 12 | SEM micrographs detailing the fracture surface of F-PB-R laminates after aging.

that the F-GT-R laminates also show less severe damage on the opposite side of impact, unlike what happens in the case of the F-PL-NR laminates, a comparison that, for the sake of completeness, is shown in Figure 17. As observed in the quasi-static testing, all laminates exhibited a stable retention of properties following the first month of aging, with no evidence of progressive or continuous degradation over time.

3.2.4 | Thermal Stability of Biocomposites by TGA

TGA was used to detect the degradation of composites after accelerated aging. By comparing the TGA curves of aged and unaged samples, it was possible to evaluate their durability and performance over time. Figure S11 shows the typical thermogravimetric curves for all composites up to the third month of aging, while the

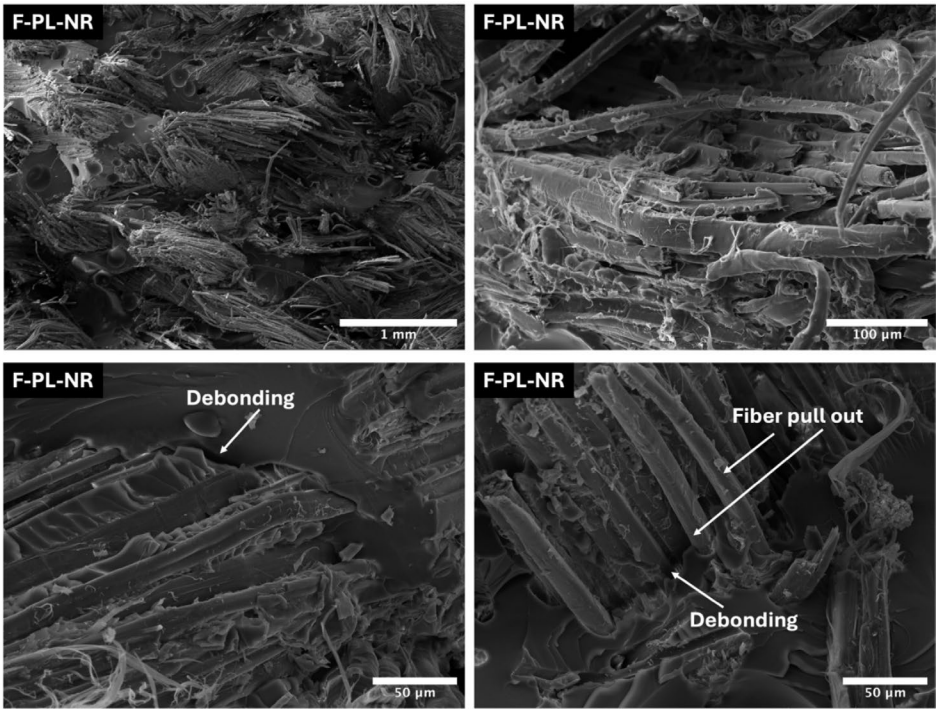

FIGURE 13 | SEM micrographs detailing the fracture surface of F-PL-NR laminates.

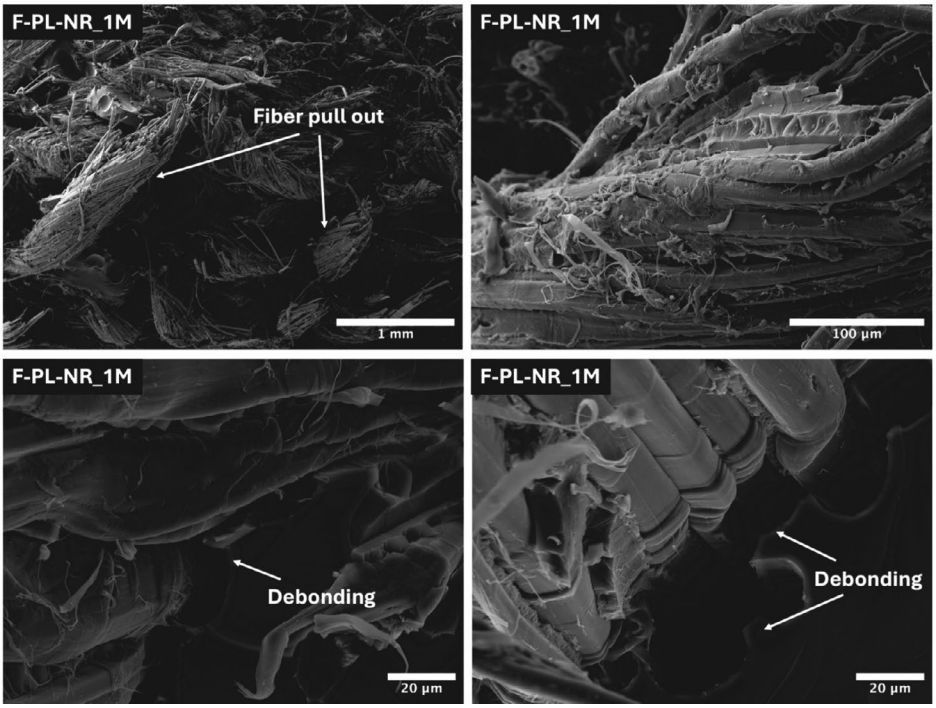

FIGURE 14 | SEM micrographs detailing the fracture surface of F-PL-NR laminates after aging.

main parameters extracted from these curves are summarized in Table 4 (T_{onset} represents the onset temperature of degradation, $T_{5\%}$ and $T_{10\%}$ are the temperatures corresponding to a weight loss of 5% and 10%, respectively. T_{max} represents the temperature of maximum degradation). The thermal degradation behavior of the aged composites closely mirrored that of the unaged counterparts. An initial mass loss was observed near 100°C, primarily attributed to the desorption of physically bound moisture. This was subsequently followed by a series of complex degradation mechanisms, resulting in the evolution of volatile decomposition products [47]. The principal mass loss occurred within the temperature range of 280°C to 370°C, corresponding to the pyrolytic degradation of the bio-based epoxy matrix, hemicellulose, and cellulose components. A secondary degradation event, occurring around 400°C, was associated with the thermal decomposition of lignin and the formation of carbonaceous char [48, 49]. Comparative

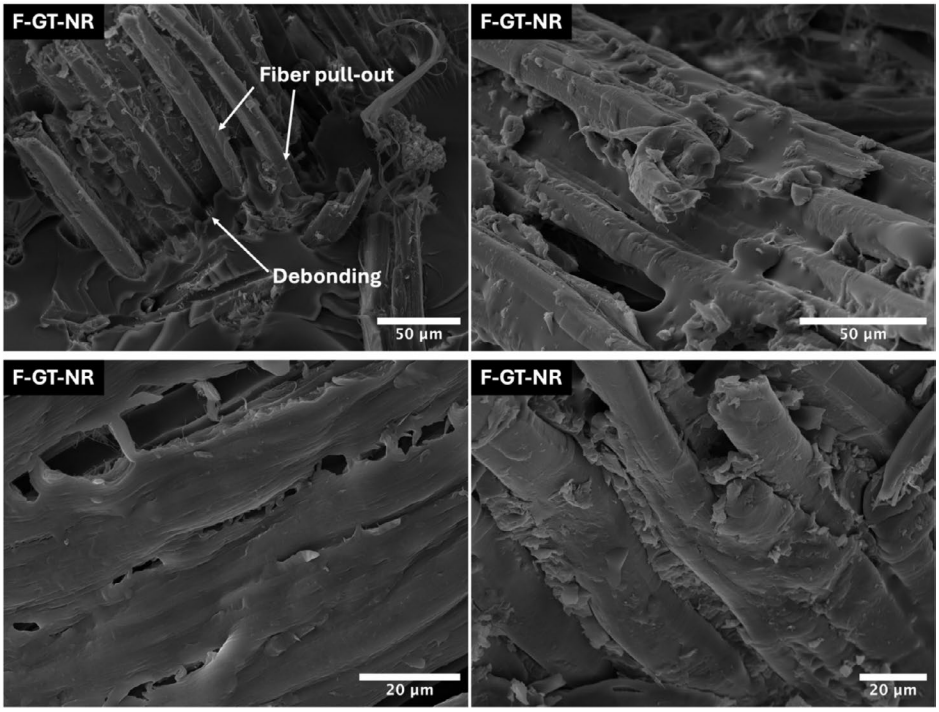

FIGURE 15 | SEM micrographs detailing the fracture surface of F-GT-NR laminates.

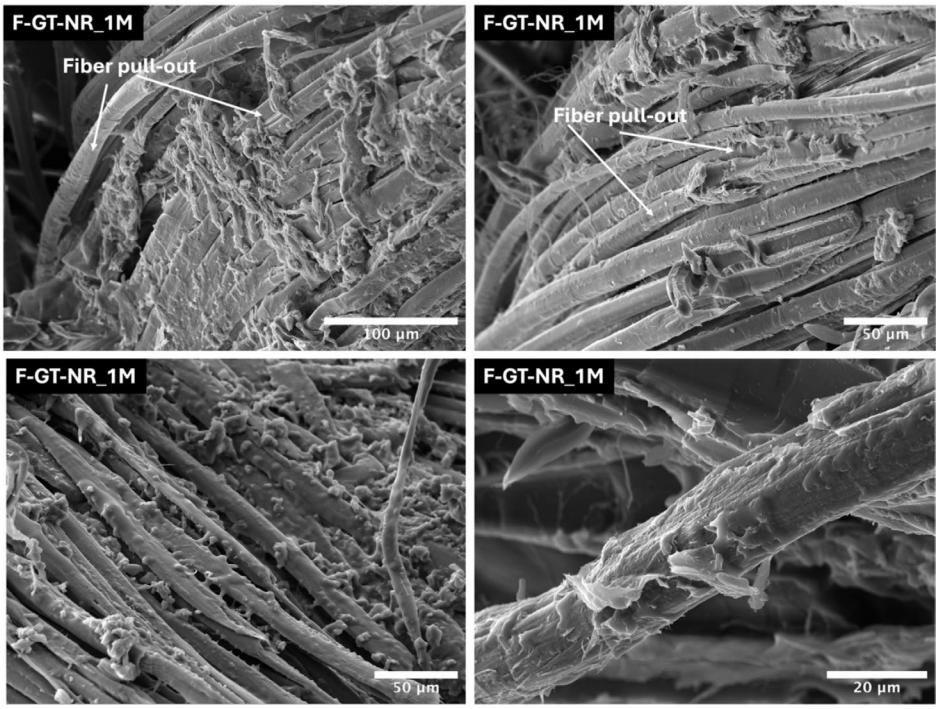

FIGURE 16 | SEM micrographs detailing the fracture surface of F-GT-NR laminates after aging.

analysis of the thermogravimetric data presented in Table 4 indicates minimal deviation in degradation onset and peak temperatures between the aged and unaged (virgin) bio-based composites. This observation is corroborated by the similar levels of residual char observed in both cases, affirming the preservation of thermal stability post-weathering. Notably, the aged bio-based composites did not exhibit a reduction in char residue, further substantiating their resistance to thermal degradation [50].

4 | Conclusions

This study evaluated the mechanical performance, aging behavior, and thermal stability of flax fiber composites reinforced with three bio-based epoxy systems—Polar Bear, Green Turtle, and Plankton—combined with recyclable and non-recyclable hardeners. Polar Bear-based composites demonstrated superior mechanical properties and thermal

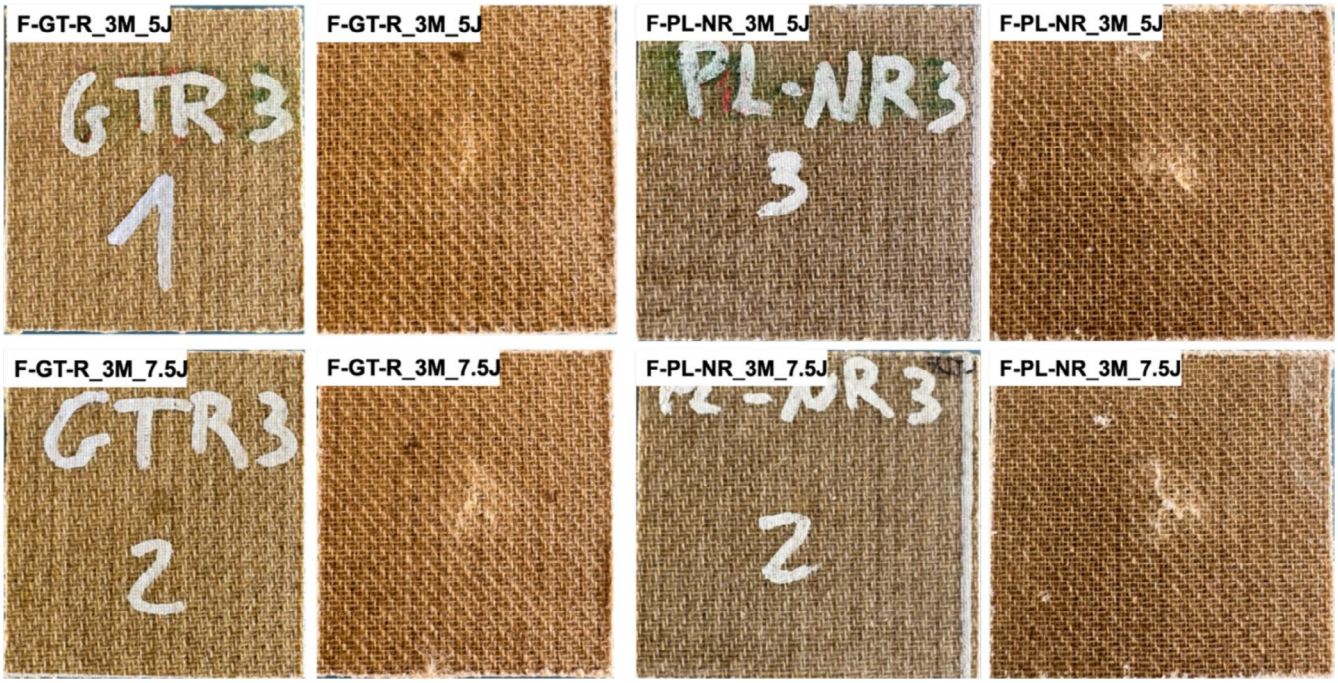

FIGURE 17 | Close-up views of front and back surfaces of F-GT-R and F-PL-NR laminates as a function of impact energy after a 3-month salt-fog exposure.

TABLE 4 | Main parameters obtained from the TGA analysis.

Sample ID	T_{onset} (°C)	$T_{5\%}$ (°C)	$T_{10\%}$ (°C)	T_{max} (°C)	Residue (wt%)
F-PB-R_0M	337.5 ± 1.2	296.2 ± 0.9	326.0 ± 0.8	370.4 ± 0.6	14.2 ± 0.5
F-PB-R_1M	329.3 ± 0.9	281.2 ± 0.6	318.0 ± 0.7	364.1 ± 0.4	14.8 ± 0.4
F-PB-R_3M	324.7 ± 0.7	264.4 ± 0.7	313.1 ± 0.4	362.2 ± 0.6	15.9 ± 0.5
F-PL-NR_0M	270.3 ± 0.4	271.3 ± 0.6	278.4 ± 0.5	296.1 ± 0.6	18.4 ± 0.3
F-PL-NR_1M	275.3 ± 0.3	252.0 ± 0.4	279.5 ± 0.4	293.2 ± 0.5	20.1 ± 0.3
F-PL-NR_3M	276.2 ± 0.4	262.4 ± 0.5	280.2 ± 0.6	295.3 ± 0.6	18.1 ± 0.4
F-GT-NR_0M	323.0 ± 0.5	325.1 ± 0.6	351.4 ± 0.4	359.7 ± 0.4	11.9 ± 0.6
F-GT-NR_1M	316.7 ± 0.6	283.8 ± 0.4	311.4 ± 0.7	357.8 ± 0.6	13.3 ± 0.7
F-GT-NR_3M	315.8 ± 0.5	280.1 ± 0.5	309.8 ± 0.4	357.8 ± 0.6	12.9 ± 0.5
F-GT-R_0M	326.1 ± 0.6	261.8 ± 0.4	309.1 ± 0.5	358.1 ± 0.4	15.3 ± 0.4
F-GT-R_1M	317.1 ± 0.7	266.8 ± 0.5	304.7 ± 0.6	355.7 ± 0.7	15.9 ± 0.5
F-GT-R_3M	319.1 ± 0.5	245.8 ± 0.6	298.7 ± 0.7	357.4 ± 0.8	14.5 ± 0.4

stability, maintaining performance after accelerated aging in salt-fog conditions. Green Turtle-based composites with recyclable hardener showed flexural strength and modulus decreases of up to 59% and 61.2%, respectively, compared to the reference system, but deformation at break increased by up to 91%, featuring minimal degradation after aging, indicating good durability and recyclability. Plankton-based systems had smaller changes, with PL-NR showing only slight reductions in strength and modulus and a modest increase in deformation. Impact strength improved by up to 83% for GT-NR, GT-R, and PL-NR, while the recyclable PL-R system showed a 20% decrease despite higher displacement. Overall, the PL-R system lacks sufficient mechanical properties for use in natural fiber-reinforced biocomposites despite its recyclability. Biocomposites with recyclable matrices containing > 40% bio-carbon (F-GT-R) exhibited lower initial flexural strength (42.7 MPa) and modulus (2.3 GPa) but superior durability under salt-fog aging. After 30 days, reductions in strength and modulus were minimal (1.9% and 6.1%), compared to up to 58.3% and 70.7% for other composites. At 90 days, F-GT-R maintained higher mechanical integrity with strength and modulus losses of 29.5% and 49.6%, respectively. ILSS showed the smallest initial decline and a significant increase (289%) at 90 days. These results confirm the enhanced aging resistance

of F-GT-R composites despite lower initial mechanical properties, mainly due to a plasticization effect.

Accelerated aging led to increased laminate compliance and matrix plasticization across all formulations, causing reduced indentation and crack severity post-impact but promoting internal damage due to interfacial degradation. No penetration was detected up to 7.5 J impact energy for any composite, confirming its impact resistance under the tested conditions. TGA revealed negligible differences in thermal degradation onset and residue between aged and unaged samples, affirming the composites' thermal stability.

Overall, Polar Bear and Green Turtle-based composites offer a promising combination of mechanical performance, durability, and recyclability for sustainable composite applications, while the Plankton system requires further optimization to improve curing and long-term stability. These results highlight the trade-off between initial mechanical performance and long-term durability in bio-epoxy systems with varying bio-carbon content and recyclability. In future work, particular emphasis will be placed on optimizing the parameters of the recycling process to enhance the quality and efficiency of fiber recovery. This will involve fine-tuning key variables such as temperature, duration, and chemical treatment conditions to ensure maximum retention of fiber integrity and mechanical performance. The recovered fibers will then be utilized to manufacture new composites, allowing for a thorough evaluation of their structural and functional viability in second-life applications. Additionally, the influence of salt-fog exposure on the recyclability of the two systems under study—PB-R and GT-R—will be systematically investigated. To the best of our knowledge, this aspect has not yet been explored in the literature, and its examination will provide critical insights into the durability and end-of-life behavior of these materials in aggressive environmental conditions. This novel approach aims to contribute to a more comprehensive understanding of circular strategies for composite materials.

Author Contributions

Vincenzo Fiore: conceptualization, project administration, methodology, funding acquisition, writing – review and editing, supervision, validation, formal analysis, resources. **Riccardo Miranda:** data curation, writing – original draft, investigation, visualization. **Iryna Kovinchuk:** data curation, writing – original draft, investigation. **Marco Luciano:** writing – original draft, data curation, investigation. **Alessia Pantaleoni:** writing – original draft, data curation, investigation. **Irene Bavasso:** writing – original draft, data curation, investigation. **Fabrizio Sarasini:** conceptualization, project administration, methodology, writing – review and editing, funding acquisition, supervision, formal analysis, resources.

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Ethics Statement

The authors disclose that language processing software was employed during the text revision process to enhance the readability and structure of the manuscript. All intellectual and analytical content remains the authors' responsibility.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The raw data supporting the conclusions of this article will be made available by the authors on request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Figure S1:** Evolution of weight as a function of temperature for the different resin formulations as obtained from TGA. **Figure S2:** Representative force versus displacement curves for all bio-based composites obtained during low velocity impact tests after aging for 1, 2 and 3 months. **Figure S3:** Close-up views of front and back surfaces of F-GT-R laminates as a function of impact energy. **Figure S4:** Close-up views of front and back surfaces of F-GT-R laminates as a function of impact energy and 1-month aging. **Figure S5:** Close-up views of front and back surfaces of F-PB-R laminates as a function of impact energy. **Figure S6:** Close-up views of front and back surfaces of F-PB-R laminates as a function of impact energy and 1-month aging. **Figure S7:** Close-up views of front and back surfaces of F-PL-NR laminates as a function of impact energy. **Figure S8:** Close-up views of front and back surfaces of F-PL-NR laminates as a function of impact energy and 1-month aging. **Figure S9:** Close-up views of front and back surfaces of F-GT-NR laminates as a function of impact energy. **Figure S10:** Close-up views of front and back surfaces of F-GT-NR laminates as a function of impact energy and 1-month aging. **Figure S11:** Thermogravimetric curves of all bio-based composites in the unaged condition (0M) and after aging for 1 (1M) and 3 (3M) months.