Mechanical Characterization of Gelatin-Flax Natural-Fiber Composites for Construction

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ABSTRACT: This article concerns the development and characterization of a protein-based alternative to traditional fiber-reinforced polymer (FRP) composites used in construction. In this work, gelatin-based resins were prepared at various gelatin-to-water (g/w) ratios. The effects of g/w ratio and curing time on resin mechanical properties were investigated. Using gelatin resins with a 30% g/w ratio, (i) gelatin-flax and (ii) gelatin-fiberglass composites were fabricated, and their mechanical properties were characterized and compared to both (iii) epoxy-flax and (iv) epoxy-fiberglass composites. Fracture surface morphologies were investigated using scanning electron microscopy. Results indicate that gelatin-flax composites exhibit similar mechanical properties compared to the epoxy-fiberglass composites and that FRP composites with fully hydrophobic or fully hydrophilic constituents have better tensile strengths than composites with a combination of hydrophobic and hydrophilic constituents. Based on this preliminary mechanical and physical property investigation, gelatin-based resins exhibit a marked potential to be used as biobased materials in the construction industry, especially in temporary structural retrofit and rehabilitation applications.

KEYWORDS: Gelatin, natural fibers, composites, mechanical properties

1 INTRODUCTION

1.1 Fiber-Reinforced Polymer (FRP) Composites in Construction

Plastics and fiber-reinforced polymer (FRP) composites are currently used in a variety of low- and high-performance construction applications, including, piping, formwork, bridge decks, and temporary structures. The demand for FRP materials used in structural retrofit and rehabilitation applications has increased over the last decade due to the deterioration of civil infrastructure in the United States. Wrapping structural elements using FRP composites is considered a state-of-the-art technique that is employed to prolong the service life of aging structures [1–4]. In addition to retrofitting, the use of FRP composites for new construction projects has demonstrated potential to ensure the long-term durability of new infrastructure [3,4].

Traditional FRP composites exhibit good chemical resistance, high strength-to-weight ratios, and ease of constructability. When FRP composites are externally bonded to structural elements, the strength, stiffness, ductility, and confinement is improved, and the technique can be executed quickly with relatively low labor costs [1,2,4]. Since FRP composites exhibit high strength and stiffness, less material is needed to achieve acceptable performance. Minimizing material quantities benefits the environment by limiting resource use and waste production [3].

FRP composites are used in a wide range of fields, including the automotive, marine, consumer goods, appliances, electronics, and construction industries [5]. Currently available FRP composites are made from non-degradable resins (e.g., epoxies, polyurethane) and high-strength synthetic fibers (e.g., graphite, aramid, glass). These materials are typically petroleum-based, which is an environmental disadvantage given that petroleum is currently consumed at a globally unsustainable rate. In addition to their non-renewability, petroleum-based FRPs do not readily degrade in landfills when they reach the end of their usable life [6,7].

1.2 Natural Fiber-Reinforced Polymer Composites

Using natural fibers and bio-derived resins in FRP composites can alleviate the negative environmental impacts of petroleum-based resins and synthetic fibers [6]. Natural fibers have many advantages when

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compared to synthetic fibers, including low cost, high toughness, low density, good specific strength, renewability, recyclability, and biodegradability [8,9]. There exists much potential for the use of natural fibers in FRP composites in structural applications. For example, natural fiber reinforced composites could be used not only in retrofitting existing infrastructure, but also for constructing temporary structures in disaster relief efforts.

Disposal of materials at the end of their usable life has become a growing environmental concern and has subsequently led to interest in developing biobased composites that are both rapidly renewable and fully biodegradable. A fully biobased composite is comprised of a natural reinforcement (e.g., flax, jute, hemp) within a natural polymer matrix. Recent research suggests that a variety of natural polymers (e.g., polyhydroxyalkanoates, polylactic acid, gelatin) exhibit a potential for use in the development of biobased FRPs for construction [7,9].

1.3 Gelatin-Based Materials

Gelatin, a protein found in the connective tissue of animals, is a partially degraded form of collagen (e.g., bovine, porcine) from skin and bones [9–11]. Commercial gelatin is derived from thermochemical degradation of collagen, which results in the disassembly of triple-helices and formation of random coils, which are stabilized by hydrogen bonds and covalent crosslinking. When dissolved gelatin is cooled below 40°C, a three-dimensional network is formed as the gelatin molecules partially reorder into a triple-helical structure [11–13]. The regeneration of the triple helix structure, which depends on gelatin-water interaction, contributes to mechanical strength and other material properties of gelatin films [9–11,13,14].

Currently, gelatin-based materials are extensively used in the food, packaging, pharmaceutical, photography, and biomedical industries [11,15,16]. Having mechanical properties comparable to commercial plastics, gelatin is an excellent candidate for biobased resins in natural fiber composite applications. While gelatin has a high elastic modulus and tensile strength compared to other biopolymers (e.g., starch, acacia gum) [17], these properties are highly dependent upon the gelatin source. For example, bovine and porcine gelatins have better mechanical properties than other gelatins [18]. Gelatin has other distinct advantages, including rapid renewability, biodegradability, global abundance, and nontoxicity [10,11,19,20].

1.4 Scope of Work

In this study, gelatin and epoxy films were prepared and their tensile mechanical properties were characterized before evaluating their performance as composite resins in natural fiber composites. This study investigated the tensile mechanical properties and fracture morphology of four formulations of fully biobased (e.g., gelatin-flax), fully synthetic (e.g., epoxy-fiberglass), or partially biobased (e.g., gelatin-fiberglass, epoxy-flax) FRP composites. The viability of using gelatin-based resins in construction applications is disussed herein.

2 MATERIALS AND METHODS

2.1 Materials

Gelatin was commercially obtained from Knox (Kraft Foods, Inc.) in granular form. Two-part epoxy was obtained from Loctite with a specified strength of 24 MPa (3500 psi). For the natural fiber reinforcement, woven flax linen in a 7.5-ounce fabric was supplied by Fabric Empire. For the synthetic fiber reinforcement, woven 8-ounce fiberglass fabric was supplied by Plasticare.

2.2 Methods

2.2.1 Gelatin Film Preparation

A 300 mL beaker of water was heated to 45°C on a Corning PC-420D hotplate. Powdered gelatin, measured in percent gelatin to weight of water (g/w ratio), was added to the water and allowed to dissolve for 10 minutes under continuous stirring by a magnetic stir bar. The mixture was then poured into a 45 cm × 30 cm rectangular form for gelation. Upon gelation, approximately 20 to 60 minutes depending on g/w ratio, the material was removed from the form and placed between two grated plates. The plates were tightly secured with zip ties to prevent warping. The gelatin films were cured in ambient conditions at a temperature of 21±2°C. Four classes of films with varying g/w ratios (10%, 20%, 30%, and 40%) were produced.

2.2.2 Epoxy Film Preparation

A 45 cm × 30 cm rectangular form was coated with Blaster Dry Lube, a polytetrofluoroethylene (PTFE) lubricant spray. The powder-based Teflon lubricant was applied as a mold release for the hardened epoxy resins. Two equal parts of the epoxy resin and hardener measuring 118 mL each were combined until a uniform

mixture was obtained. The solution was poured into the rectangular form and allowed to cure according to the conditions specified in Section 2.2.1.

2.2.3 *Gelatin and Epoxy Fiber-Reinforced Composite Preparation*

For this comparative study, one-ply composite specimens of gelatin-flax (G-Fl), gelatin-fiberglass (G-Fi), epoxy-flax (E-Fl), and epoxy-fiberglass (E-Fi) were fabricated. Gelatin (30% g/w ratio) and epoxy resins were prepared as described in Section 2.2.1 and Section 2.2.2, respectively. Woven fabrics of flax and fiberglass were measured and cut to fit inside of a 45 cm × 30 cm rectangular form. Once the resins were poured into the rectangular forms, one sheet of woven fabric was placed in the resin and allowed to saturate. The fiber was then flipped over so that each side would be completely coated by the resin. Upon initial gelation (approximately 20 minutes), the gelatin-based composite material was removed from the form and placed between two grated plates, while the epoxy-based composite material was allowed to cure in the form. The plates were tightly secured with zip ties to prevent warping. Both epoxy and gelatin composites were allowed to cure according to the conditions specified in Section 2.2.1.

2.2.4 *Tensile Mechanical Properties*

After the curing of films and composite plates, tensile test specimens were laser cut according to the dimensions outlined in ASTM D638 using an Epilog Legend 36EXT laser system. The tensile properties of the gelatin films and fiber composites were determined according to ASTM D638 standard test methods. Using a displacement-controlled rate of 5 mm/min, the tension tests were conducted using an Instron 5869 Universal Testing Machine and an Epsilon Technology Corp axial extensometer model 3542 with a 25.4 mm gauge length. For each parameter of the study, at least five replicate specimens were tested.

2.2.5 *Scanning Electron Microscopy*

The fracture surfaces of the tensile-tested composites were investigated using scanning electron microscopy. The samples were sputter-coated with gold before examination in a JEOL JSM 6480LV scanning electron microscope (SEM). The specimens were observed in a nitrogen atmosphere under vacuum with a voltage of 10 kV.

3 RESULTS AND DISCUSSION

3.1 Tensile Mechanical Properties of Gelatin and Epoxy Resins

3.1.1 *Gelatin Films*

3.1.1.1 Stress-Strain Response

The stress-strain response of representative gelatin film samples with varying gelatin-to-water (g/w) contents by weight is shown in Figure 1. The results indicate that, while all samples exhibited similar stress-strain relationships, the elongation-to-break increased with g/w content from 10% to 40%, as shown in Table 2. While the 10% g/w samples remained linear elastic before rupture, the 20%, 30%, and 40% g/w samples exhibited an increasingly plastic response. This response may be attributed to increasingly high concentrations of triple-helix structures in the gelatin films, since the development of helix structures has been linked to increases in gelatin concentration. A number of authors have noted that helix content will cause increases in mechanical properties [9–11] and that high g/w ratios increase the probability of triple-helix structure nucleation and helix content in gelatin films [22].

The tensile strengths and elastic moduli of the gelatin resins are depicted in Figure 2. According to the data, the tensile strength and elastic modulus reached average maximum values of 66.3 MPa and 4728 MPa at 30% and 20% g/w content, respectively. An analysis of the statistical significance of the differences in the mechanical property results between the 10%, 20%, 30%, and 40% g/w films was completed using an analysis of variance (ANOVA). The results are shown in Table 1. Using a p-value of less than 0.05, the mean

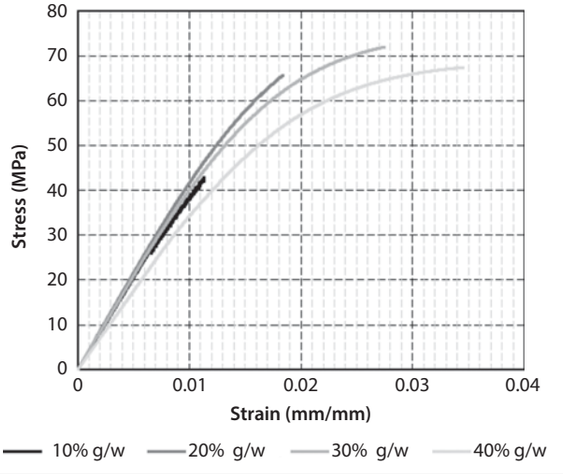

Figure 1 Representative stress-strain curves for films with different gelatin-to-water ratios.

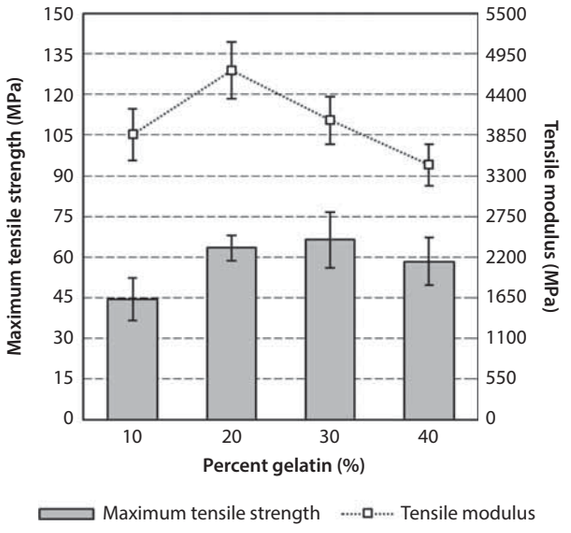

Figure 2 Tensile strength and elastic modulus of gelatin films.

mechanical property values that are statistically different can be seen in Table 1. For maximum tensile strength, 20%, 30%, and 40% g/w content have comparable mean values according to the ANOVA, but all are statistically greater than the 10% g/w film samples. A notable peak is observed for the 30% g/w samples. For tensile modulus, the 20% g/w samples have the greatest mean value.

The data suggest that an optimal g/w ratio (approximately 30%) exists that maximizes mechanical strength and stiffness while considering elongation-to-break. ACI 440.8-14 specifies the minimum tensile property requirements for the saturating resins of FRPs [23]. The ACI 440.8-14 minimum ultimate tensile strength is 6000 psi (41.4 MPa), minimum tensile modulus is 250,000 psi (1724 MPa), and minimum elongation-to-break is 0.03. Given the experimental mechanical property results and the ACI 440.8-14 minimum requirements, the 30% g/w resin was selected herein for continued analysis.

3.1.1.2 Effect of Curing Time on Tensile Mechanical Properties

Figure 3 shows the changes in mechanical properties of the 30% g/w resin that occurred over a curing period of 21 days. Both tensile strength and elastic modulus increased over time with an average increase of approximately 96% and 21% from 3 to 21 days, respectively. The data is plotted on a logarithmic scale to show the relationship between time and mechanical property development. The changes in both strength and stiffness properties exhibited approximate linear relationships on a logarithmic scale as denoted by the coefficient of determination, R^2. These data suggest that the mechanical properties of the gelatin films may continue to gain in mechanical strength and stiffness beyond 21 days.

Table 1 ANOVA results for tensile strength and modulus of gelatin films.

Mechanical Property	Gelatin Concentration		p-value
	Selected	Compared to	
Maximum Tensile Strength	10%	20%	0.0011
		30%	0.0019
		40%	0.0204
	20%	30%	0.569
		40%	0.297
	30%	40%	0.205
Tensile Modulus	10%	20%	0.0034
		30%	0.343
		40%	0.0640
	20%	30%	0.0110
		40%	0.00031
	30%	40%	0.0095

3.1.2 *Epoxy Films*

3.1.2.1 Tensile Mechanical Properties

The mechanical properties, namely ultimate tensile strength, tensile modulus, and elongation to break, of the epoxy resin are shown in Table 2, along with the 7-day mechanical properties of the gelatin films. The epoxy exhibits both a lower tensile strength and a lower tensile modulus compared to the gelatin resins, while the elongation-to-break of the epoxy is comparable to the gelatin films. Specifically, the maximum tensile strength and tensile modulus achievable by the gelatin films were 133% and 107% higher than the epoxy resin, respectively.

3.2 Tensile Mechanical Properties of Natural FRP Composites

3.2.1 *Gelatin-Flax (G-Fl), Gelatin-Fiberglass (G-Fi), Epoxy-Flax (E-Fl), and Epoxy-Fiberglass (E-Fi)*

The fiber mass and volume fractions for E-Fi, E-Fl, G-Fi, and G-Fl were determined according to a modified matrix dissolution method adapted from Srubar *et al.*

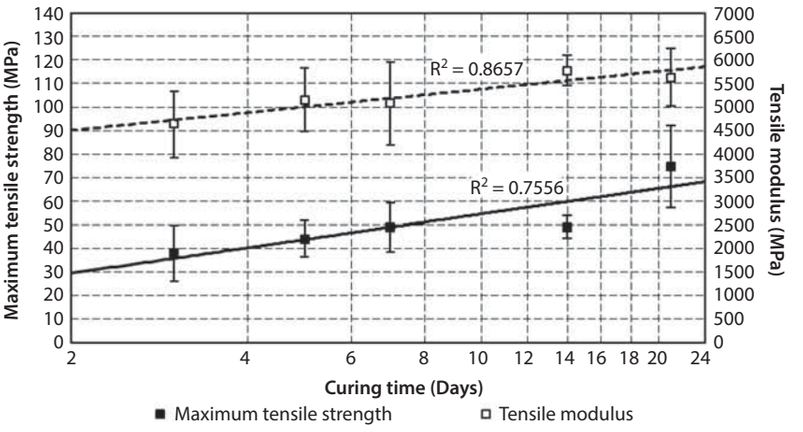

Figure 3 Time-dependent development of tensile strength and elastic modulus of 30% g/w films.

Table 2 Mechanical properties of resins.

	Maximum Tensile Strength (MPa)			Tensile Modulus (MPa)			Elongation-to-Break (mm/mm)		
Resin	Mean	Standard Deviation	Coefficient of Variation	Mean	Standard Deviation	Coefficient of Variation	Mean	Standard Deviation	Coefficient of Variation
10% g/w	44.2	7.9	17.9%	3858	352	9.1%	0.0128	0.0022	16.8%
20% g/w	63.3	4.8	7.6%	4728	381	8.1%	0.0161	0.0033	20.8%
30% g/w	66.3	10.2	15.4%	4052	322	8.0%	0.0278	0.0137	49.1%
40% g/w	58.3	8.8	15.0%	3445	283	8.2%	0.0274	0.0113	41.2%
Epoxy	28.4	5.6	19.6%	1961	142	7.3%	0.0199	0.0051	25.4%

[24], in which the matrix of a composite sample of known mass and volume was dissolved in a solvent and the fiber mass was recovered after full dissolution of the matrix. The fiber mass fractions for the E-Fi, E-Fl, G-Fi, and G-Fl samples were approximately 11%, 15%, 16%, and 16%, respectively, which corresponded to volume fractions that were approximately 5%, 15%, 10%, and 5%, respectively.

The tensile strength and modulus of the four FRP composites are shown in Figure 4a and 4b, respectively, and an analysis of the statistical significance of the results is shown in Table 3. As shown in Figure 4a, the fully synthetic (E-Fi) and fully biobased (G-Fl) composites exhibited higher tensile strengths than the partially biobased composites (E-Fl, G-Fi). For example, the tensile strength of the fully synthetic E-Fi composites was 123% higher than the partially biobased E-Fl composites. Similarly, the tensile strength of the fully biobased G-Fl composite was 52% higher than the partially biobased G-Fi composites. The fully biobased G-Fl composites exhibit a tensile strength only 12% less than the fully synthetic E-Fi composite. The improved tensile strength for the E-Fi and G-Fl composites can be attributed to an improved interfacial bond between the compatible fully synthetic and the fully biobased constituents.

The cohesiveness of the interface is well known to influence the tensile strength of natural fiber composites. Improved interfacial adhesion is expected in hydrophilic-hydrophilic and hydrophobic-hydrophobic fiber-matrix systems [25]. The incompatibility of hydrophobic polymer resins with hydrophilic natural fibers is well acknowledged, and surface treatments are commonly used to strengthen interface compatibility [25,26]. The results in this study further suggest that the use of hydrophilic gelatin resins improves interfacial adhesion between natural (hydrophilic) flax fibers, resulting in better tensile mechanical properties without the use of surface modification [26,27]. Thus, given that gelatin and flax are both hydrophilic and epoxy and fiberglass are both hydrophobic, the compatibility of the fiber-matrix interface is expected to be

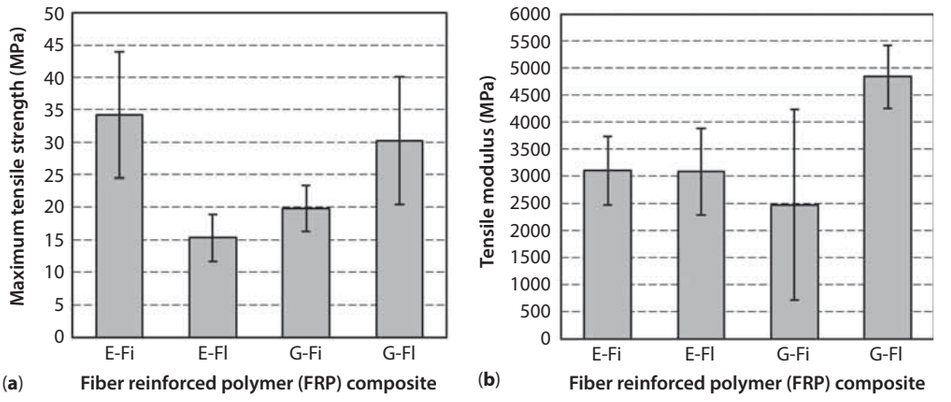

Figure 4 Tensile strength (a) and modulus (b) of FRP composites.

Table 3 ANOVA results for tensile strength and modulus of FRP composites.

Mechanical Property	FRP Composite		P-value
	Selected	Compared to	
Maximum Tensile Strength	E-Fi	E-Fl	0.00042
		G-Fi	0.0018
		G-Fl	0.463
	E-Fl	G-Fi	0.0305
		G-Fl	0.0026
	G-Fi	G-Fl	0.0146
Tensile Modulus	E-Fi	E-Fl	0.961
		G-Fi	0.390
		G-Fl	0.00016
	E-Fl	G-Fi	0.418
		G-Fl	0.00050
	G-Fi	G-Fl	0.0048

superior to the hydrophilic-hydrophobic fiber-matrix systems. This compatibility may not be true for all fully biobased fiber-matrix systems. For example, gelatin is both hydrophilic and biobased, while other biopolymers (e.g., polylactic acid, polyhydroxybutyrate) have inherently hydrophobic chemistries [25–28].

In terms of composite stiffness, the average tensile modulus of the fully biobased G-Fl composites was 56% greater than the fully synthetic E-Fi composites, as shown in Figure 4b. As expected, there is high variability in the data for the gelatin-flax composites. This effect can be attributed to the inherent variability of natural fibers and the variation in gelatin mechanical properties [11,18,21].

While it has been previously noted that fully biobased composites may not have suitable stiffness and strength for high load-bearing applications, the results of these and other studies that have reported on the mechanical properties of flax-reinforced composites demonstrate the potential for fully biobased composite to be competitive with other synthetic and biobased natural-fiber composites, as well as wood and engineered-wood materials [21,26,29]. In addition, the mechanical properties of the G-Fl composites are comparable, if not superior, to mechanical properties of flax-fiber composites previously reported elsewhere. For example, Huang and Netravali [30] found that FRP composites consisting of woven flax fiber within a soy protein concentrate (SPC) matrix exhibited mean tensile strengths of 54.6 MPa and 68.7 MPa and mean tensile moduli of 994 MPa and 1123 MPa for the warp and weft fiber directions, respectively. Kumar *et al.* [31] found that woven flax fiber within a polylactic acid (PLA) resin had a mean tensile strength of 21 MPa and a mean tensile modulus of 137 MPa. The G-Fl composites studied herein achieved tensile strength and stiffness properties of 30 MPa and 4.9 GPa, respectively, suggesting that the tensile strength of gelatin enhanced the mechanical performance of flax-fiber composites in comparison to other biobased resins such as soy or PLA.

3.2.1.1 Fracture Surface Morphology

After mechanical testing, the fracture surfaces of the four FRPs were examined. The SEM micrographs of the fiber-matrix interface of the E-Fl, E-Fi, G-Fi, and G-Fl samples are shown in Figures 5a–d, respectively. It can be seen in Figures 5a and 5c that the interfaces for E-Fl and G-Fi have a relatively weak bond as evident by the separation between the polymer matrix and fiber reinforcement. The fully synthetic (Figure 5b) and fully biobased composite (Figure 5d) exhibited an improved bond as physically evident by the contiguity of the polymer matrix and reinforcing fiber. The improved mechanical properties of the fully synthetic and fully biobased composites can be attributed to the

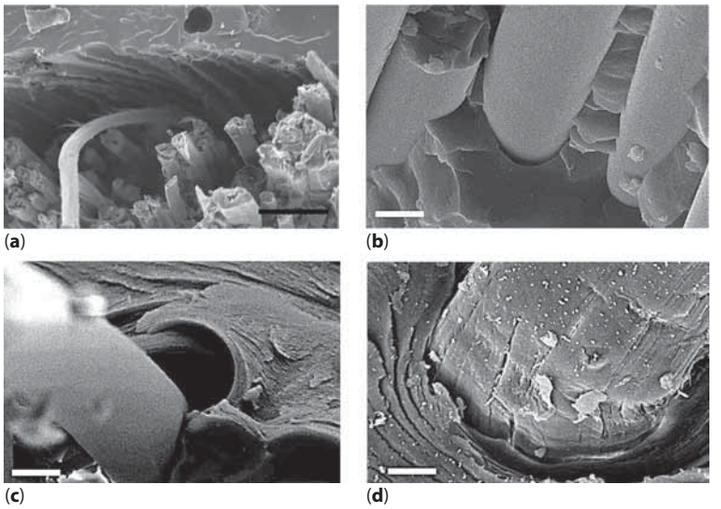

Figure 5 Scanning electron microscopy images of (a) E-Fl (50 μm scale bar), (b) E-Fi (5 μm scale bar), (c) G-Fi (5 μm scale bar), and (d) G-Fl (5 μm scale bar).

enhanced fiber-matrix interface of these composite systems. The tensile mechanical properties and SEM images performed in this study further corroborate the findings within the literature that indicate a marked influence of the interface between the fiber and resin on the mechanical properties of FRPs [3,8,25,26].

Together with the mechanical property data, these images suggest that fully biobased G-Fl FRPs exhibit a marked potential to be an environmentally viable alternative to conventional E-Fi FRPs. However, a number of remaining concerns need to be addressed to advance the science and engineering of these fully biobased FRPs. These concerns, which will be investigated in future work, include further improvements in mechanical properties, such as multi-ply composites and long-term durability, namely moisture and high-temperature resistance.

4 CONCLUSIONS

This study investigated the effect of gelatin-to-water ratio and curing time on the mechanical properties of gelatin-based resins. The mechanical properties of gelatin films and fully biobased G-Fl composites were characterized and compared to the mechanical properties of fully synthetic and partially biobased FRPs. The experimental data indicated the following conclusions:

1. 30% gelatin-to-water (g/w) films have improved tensile properties (i.e., strength, elastic modulus, elongation-to-break) when compared to conventional epoxy resins;
2. The tensile strength and stiffness of 30% g/w films increase 96% and 21%, respectively, from 3 to 21 days, when cured in ambient conditions;
3. Fully biobased gelatin-flax composites exhibited comparable strength and enhanced stiffness compared to fully synthetic epoxy-fiberglass composites, suggesting that composites with fully hydrophobic or fully hydrophilic constituents have improved initial tensile properties when compared to composites with a combination of hydrophilic and hydrophobic constituents; and,
4. The strength of gelatin-flax composites is influenced by the cohesiveness of the interface between the constituents as indicated by SEM micrography.

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