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Ramie Fiber-Reinforced Polylactic-Acid Prepreg: Fabrication and Characterization of Unidirectional and Bidirectional Laminates

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Abstract. The fabrication of a natural prepreg with poly-lactic acid (PLA) matrix and ramie fiber reinforcement was engineered on a laboratory scale by impregnating the unidirectional and bidirectional ramie fiber with PLA matrix solvent on a glass die. The obtained composite prepreg has been stored at a very low temperature to maximize its shelf life. Tensile and biodegradability tests of the composite laminates prepared by the hot-pressing method have also been conducted. Tensile test results show that the freezer-stored bidirectional 0/90° prepreg laminate specimen has the highest tensile strength of 71.44 MPa with a modulus of 2.70 GPa on average. Meanwhile, the unstored bidirectional 0/90° prepreg laminate specimen has the highest level of elasticity, with a modulus of 1.29 GPa on average. The biodegradability test shows the decomposition process of the composite laminate under actual composting conditions. Microscopic observation of the damaged specimen results shows good adhesion between the PLA matrix and ramie fiber and the decomposition of the biodegradability test samples.

Keywords: Composite; Natural prepreg; Polylactic Acid (PLA); Ramie fiber

1. Introduction

Natural fiber-reinforced polymer (NFRP) has been a trend in composite materials research and engineering (John *et al.*, 2008). It offers the biodegradable advantages of a composite (Lotfi *et al.*, 2019) and the comparable mechanical strength (Holbery and Houston, 2006) to the conventional synthetic ones (Faruk *et al.*, 2012). Polylactic acid (PLA) is a biodegradable polymer that has become the most pledging biodegradable material that has been used as a matrix in a composite material due to its vulnerability to bacteria (Siakeng *et al.*, 2019). It is frequently used to replace synthetic polymers to address the disposal (environmental) problem (Alsaeed *et al.*, 2013) we have faced in the recent decade. PLA is environmentally friendly and can be decomposed naturally. PLA also had good physical and mechanical properties (Bhardwaj and Mohanty, 2007). Furthermore, the usage of PLA as a matrix to natural fiber-reinforced composite will bring out the maximum potential of fully biodegradable composites.

On the other hand, ramie fiber has been used as reinforced in PLA composites due to its strength superiority among the stem fibers. Consequently, PLA and ramie fiber have

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been considered the most common natural constituents in biodegradable composite (Lololau *et al.*, 2021) as a ramie fiber-reinforced polylactic-acid (RFRPLA).

Unfortunately, its composites still have to be fabricated in conventional procedures. It can be improved by using a prepreg or pre-impregnation during its preparation. Prepregs or pre-impregnated composites are semi-finished composite products made by impregnating a textile/fabric architecture of a fiber reinforcement with a thermoplastic or thermoset matrix (resins). Therefore, a prepreg can be defined as a preform braided structure of the reinforcement used as a composite (Potluri and Nawaz, 2011). Composite prepregs reduce the risk of poor impregnation quality by ensuring that the amount of each constituent is correct and interacts well (Duhovic and Bhattacharyya, 2011). It will also reduce the risk of possible composite processing defects, such as applying complex geometries like curvature indentations (Wang *et al.*, 2020). Prepreg is generally used as a material for manufacturing components in the aircraft industry because of its advantages: having a high track and drape, which is useful for components with complex shapes (Seferis *et al.*, 2011).

The reinforcing fiber in the prepreg will still be aligned as it was before during the manufacturing process. Consequently, it is considered suitable and capable of making parts with lower fiber defects with excellent characteristics (Cairns *et al.*, 2001). Prepreg has a very good performance compared to other forms of composite materials. This material is suitable for manufacturing composite parts that are very light but can bear significant loads (Wolff-Fabris *et al.*, 2016). Prepregs require good storage, i.e., away from direct sunlight, heat, and strong chemicals. To extend its shelf life, prepregs need to be stored at temperatures below 0°C (Bhatnagar *et al.*, 2006). The method of prepreg preparation on composites (especially thermoplastics) with natural fiber reinforcement can be carried out by spinning the reinforcing yarn with matrix filaments (Baghaei and Skrifvars, 2016; Baghaei *et al.*, 2015; Baghaei *et al.*, 2013). Another study also conducted the preparation by hot-rolling a matrix sheet with reinforcing fabrics (McGregor *et al.*, 2017). Preparation of prepregs can also be carried out by dissolving the matrix granules into a solvent compound, which is then used to impregnate the reinforcing fabrics (He *et al.*, 2019).

Due to the gap from the predecessor studies, this research had brought in the engineered fabrication of fully biodegradable composite materials of ramie fiber as reinforcement and PLA as the matrix on a laboratory scale by using a manual solvent casting impregnation method. Also, this research aimed to determine the characteristic of its biodegradability, interface bonding, and tensile properties.

2. Methods

2.1. Materials

Ramie plain-woven fabric was supplied by Guangzhou Xinzhi Textile Co., Ltd. (China), and Bio-poly 103 PLA granules were chosen as the matrix from Shanghai Huiang Industrial Co., Ltd. (China). Meanwhile, NaOH and dichloromethane were supplied by a local distributor PT. Indogen Intertama (Indonesia).

2.2. Prepreg preparation

Two types of reinforcement were fabricated: unidirectional and bidirectional. The bidirectional will be prepared in 0/90° and ±45° fabrics. The ramie fabric used in this study is a plain weave type. The ramie woven fabric was cut into 250 × 190 mm^2. Then, some of those cut woven yarn is yanked in the perpendicular direction to make a unidirectional fabric reinforcement. Since the ramie fiber has hydrophilic properties and PLA has hydrophobic properties, it is necessary to apply a surface treatment to the ramie fiber to increase the interfacial adhesion between both constituents (He *et al.,* 2019). Ramie fiber was soaked in NaOH solution (5% wt) with a ratio of fiber and solution of 1:10 for 2 hours, then rinsed until the pH reached 7. The fiber then being dried at room temperature for 12 hours.

Figure 1 illustrates the flow of the impregnation process. The PLA granules were dissolved in dichloromethane solvent with a ratio of 1:10 using a magnetic stirrer for 2 hours at room temperature. The ramie fabric then being manually impregnated in the PLA/dichloromethane solution. The reinforcement was impregnated on a glass mold (impregnation bath) of 25.4 × 19.4 cm^2 until the matrix was thoroughly pervaded and the excess evaporated. After that, the resulting prepregs are taken out and covered with parchment paper, as seen in Figure 2. Then the prepregs were rolled up, and half of the 0/90° one was stored in a refrigerator freezer at -18°C for a week as a preserving act.

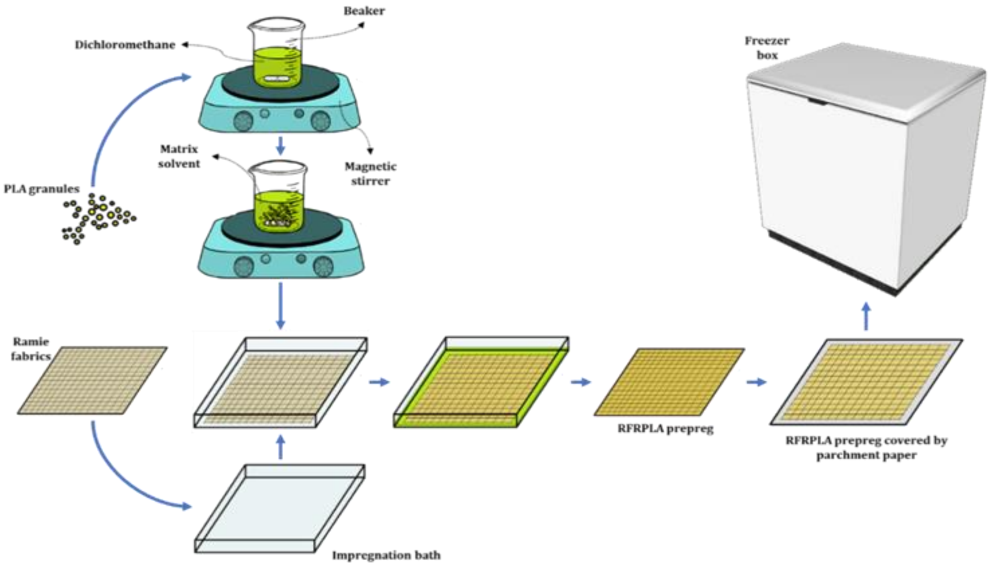


Figure 1 Impregnation process flow

Figure 2 Prepared prepregs

2.3. Specimen preparation

Figure 3 shows the flow of RFRPLA prepreg specimen fabrication. Before undergoing a tensile test, the fabricated prepregs were prepared into a plate specimen through the hot-press polymerization method. The prepreg sheets made previously were removed from the parchment paper covering, then stacked in a 25.4 × 19.4 cm^2 AA 6061-T6 mold, and then hot pressed at 120°C with 132 bar pressure for approximately 90 minutes. The composite laminate plate is then cut using laser cutting according to the geometry of the American Society for Testing and Materials (ASTM) D3039 standard (ASTM, 2017). Also, some residual-cut specimens will be decomposed as biodegradability test samples.

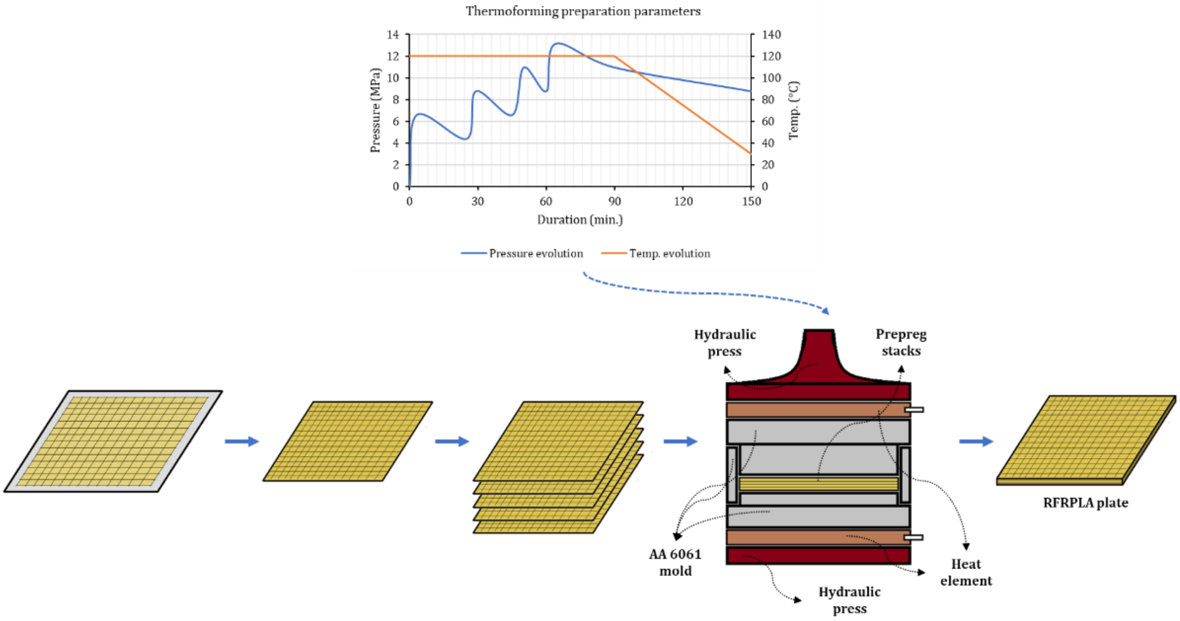


Figure 3 Specimen preparation flow

2.4. Characterization

2.4.1. Tensile test

Tensile tests are carried out according to the ASTM D3039/D3039M standard. The RFRPLA specimens were tested on the Tinius Olsen universal uniaxial testing machine at the Metallurgy and Materials Research Center (P2MM) LIPI. The machines were equipped with a 30 kN load cell with a 2 mm/min displacement rate. The test was performed on four samples, consisting of a unidirectional (UD) sample, unstored bidirectional (BD) 0/90° sample, freezer-stored BD 0/90° sample, and BD ±45° sample. The test was also performed on 6 (six) duplicated specimens of each sample.

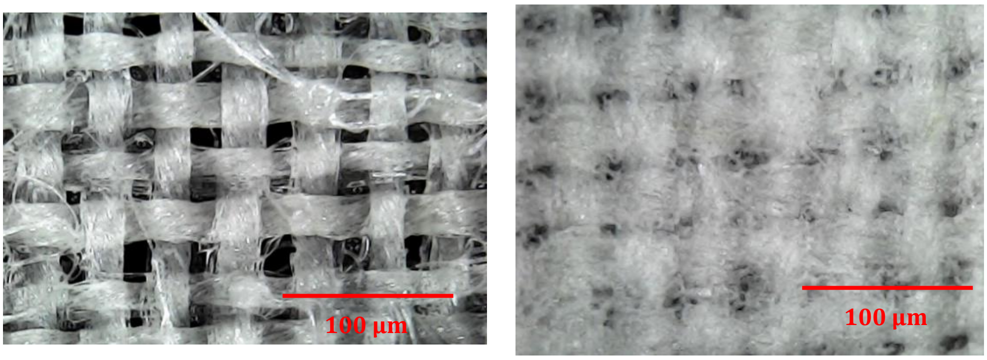
2.4.2. Biodegradability test

The biodegradability test was carried out to see the decomposition process in the RFRPLA composite. This test is carried out by placing a small sample on the soil with actual composting conditions. The sample used was the unused cut of unstored 0/90° RFRPLA prepregs laminate plate. Those samples were used as the control sample depicts any other unstored samples. The test sample consists of two sizes, Sample A (5cm x 5cm) and Sample B (2.5 cm x 5 cm), with four duplications, respectively. The composting condition consisted of cow dung, wood shavings, and animal feed waste placed in a wooden box with a width of 0.5 m, length of 0.6 m, and height of 0.3 m. The decomposition process of the composite was measured by mass change and observed for 120 days to see changes in the shape and color of the sample.

3. Results and Discussion

3.1. Prepreg fabrication

Prepregs stored in the refrigerator with those not had been compared. Both prepregs were left for a week; then, it was found that the unstored prepregs experienced faster resin hardening. Figure 4 shows the microscopic condition of the prepregs, where there is a very significant difference. The freezer-stored prepregs have a higher matrix density and penetration than the unstored ones.



(a) (b)

Figure 4 0/90° Prepreg: (a) Unstored; and (b) Freezer-stored

3.2. Tensile properties

A couple of tensile tests were conducted to determine the mechanical characteristics of the RFRPLA composite. Table 1 shows the Ultimate Tensile Strength, 0.2% Offset Stress, Strain, and Young Modulus data from the tested specimens, while Figure 5 shows the stress-strain trajectory of the tested prepreg laminates.

Table 1 Tensile test results

Prepregs condition	Specimen code	Ultimate tensile stress	0.2% offset stress	Ultimate tensile strain	Modulus
		MPa	MPa		GPa
Unstored	UD-A	53.58	27.00	3.67%	1.95
	UD-B	40.87	35.40	2.57%	2.01
	UD-C	42.10	32.60	2.82%	1.87
	UD-D	39.07	29.80	2.62%	1.69
	UD-E	46.97	42.50	2.94%	2.49
	UD-F	54.15	34.20	3.27%	1.60
	Average	46.12	33.58	2.98%	1.93
	St. dev.	6.55	5.33	0.42%	0.31
Unstored	BD090-1-A	45.87	19.00	5.37%	1.77
	BD090-1-B	42.84	18.60	5.49%	2.08
	BD090-1-C	47.58	20.30	5.29%	0.74
	BD090-1-D	47.71	19.10	5.93%	0.91
	BD090-1-E	45.58	18.50	5.61%	0.96
	BD090-1-F	49.30	26.10	5.41%	1.28
	Average	46.48	20.27	5.52%	1.29
	St. dev.	2.24	2.93	0.23%	0.53
Freezer-stored	BD090-2-A	74.10	34.30	4.25%	1.65
	BD090-2-B	78.12	32.10	4.44%	1.83
	BD090-2-C	65.43	32.10	3.32%	3.44
	BD090-2-D	68.36	30.50	3.95%	2.56
	BD090-2-E	74.10	33.30	3.83%	4.52
	BD090-2-F	68.55	27.40	4.13%	2.18
	Average	71.44	31.62	3.99%	2.70
	St. dev.	4.75	2.43	0.39%	1.10
Unstored	BD45-A	48.42	27.80	6.28%	2.82
	BD45-B	45.44	22.60	8.17%	0.62
	BD45-C	51.95	21.70	10.74%	1.73
	BD45-D	38.39	24.30	5.61%	1.35
	BD45-E	46.42	28.70	5.85%	2.22
	BD45-F	40.02	24.90	5.04%	1.80
	Average	45.11	25.00	6.95%	1.76
	St. dev.	5.11	2.78	2.14%	0.75

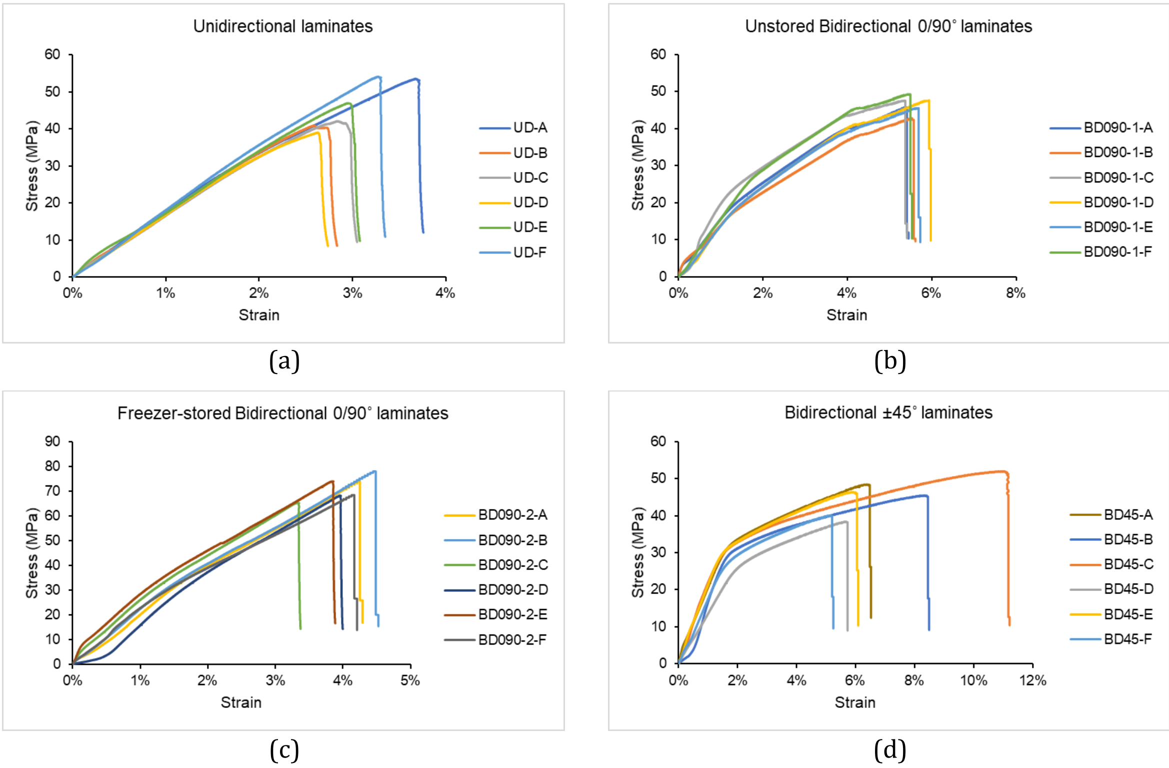


Figure 5 Stress-strain curves of 4 tested samples according to the preparation condition

The tensile test results obtained showed various results. It is probably caused by the manufacturing imperfection, which during the preparation stage, the PLA matrix solution was not evenly distributed, resulting in the difference in each specimen's tensile strength. Whereas in unidirectional fiber, there are differences in fiber density due to misalignment of the fibers, so the ability of the fiber to accept the load on each specimen is different. The manuality of the process causes the misalignment in the unidirectional composite laminate, so the fiber alignment is quite challenging.

Table 1 and Figure 5 shows that the freezer-stored 0/90° prepregs composite specimen (BD-0/90-2) had the highest ultimate tensile strength, with an overall average of 71.44 MPa, which also has the highest average Young's modulus of 2.70 GPa. The unstored 0/90° prepregs composite specimen has the lowest average Young's modulus of 1.29 GPa, which can be declared the most elastic composite.

Generally, the yield point indicates the maximum stress value material can accept before undergoing plastic deformation. However, there are two failure modes in composites: matrix and fiber failure modes. Graphically, the yield point cannot be seen clearly on the composite tensile test result curve. Therefore, 0.2% offset stress was used to determine the yield strength of the composite.

The matrix density mentioned in section 3.1 also influences the composite laminate's tensile strength, as seen in Table 1. From Table 1, the freezer-stored prepreg composite has the highest tensile strength. This phenomenon can be studied further in future research.

Microscopic observations were made on the fracture cross-section of the RFRPLA specimens after the tensile test, as shown in Figure 6. The four images show that the most common failures are matrix cracking and fiber breakage. The four figures also show that the ramie fiber and matrix are well bonded. In Figure 6d, it can be seen that the PLA matrix is bonded to the fiber surface. It indicates that the PLA matrix was well impregnated into the fiber during prepreg fabrication due to the good adhesion between the fiber and the matrix. The interfacial adhesion of the fiber and matrix itself was optimized by treating the fiber surface by immersing the ramie fiber in NaOH solution.

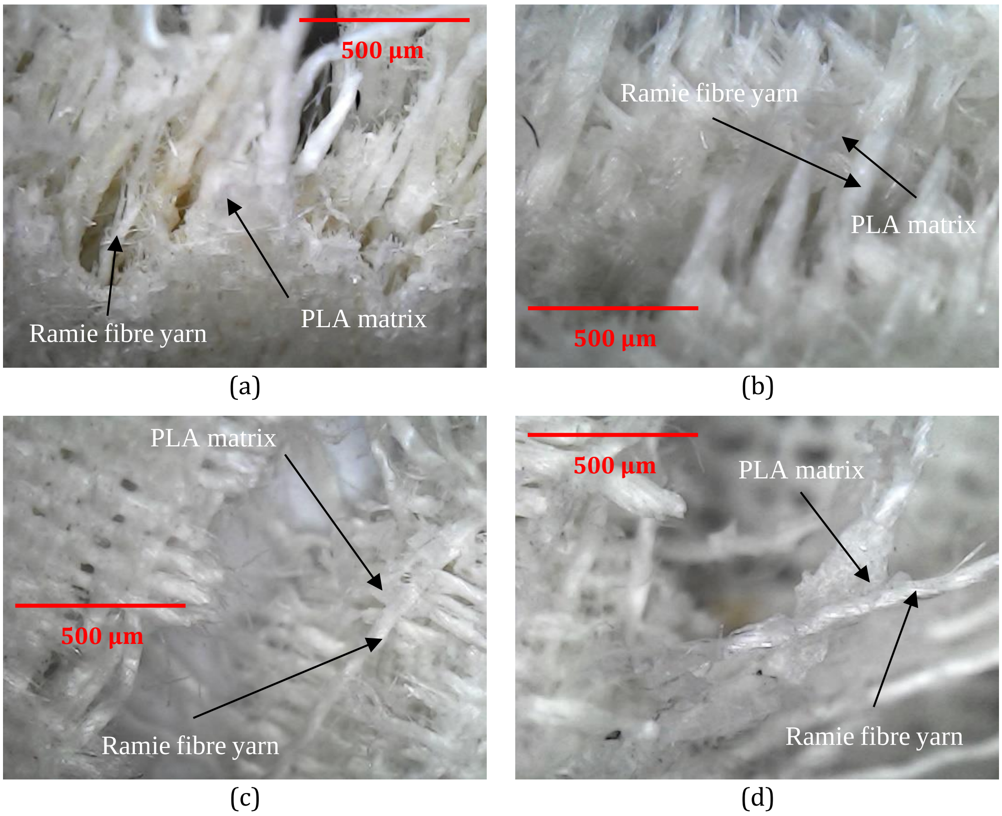


Figure 6 Microscopic view of the damaged specimens: (a) Unidirectional laminates; (b) Unstored 0/90° laminates; (c) Freezer-stored 0/90° laminates; and (d) ±45° laminates

3.3. Biodegradability

The biodegradability test samples were observed visually by observing changes in color and shape of the sample from day 0 to day 120. The biodegradability test sample in this study did not experience a significant change in shape, but changes in the color of the sample could be seen. It is caused by water and soil content absorption into the sample. The absorption of water and soil caused the samples to undergo weathering, which indicated that the samples from the RFRPLA composite in this study were biodegradable. Figure 7 shows the final condition of the decomposition samples, which suffer discoloration and weathering.

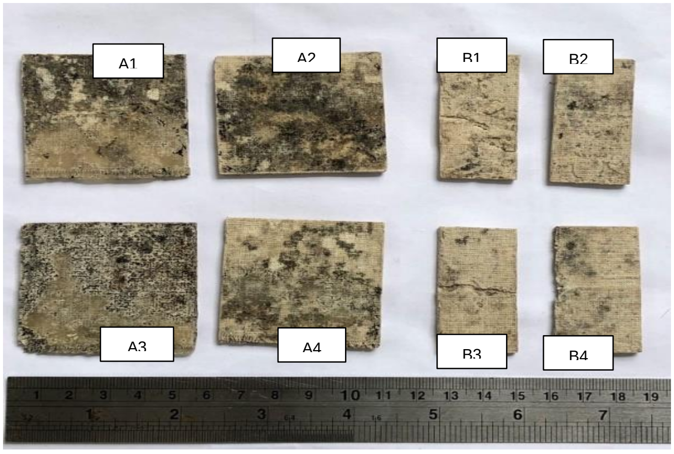


Figure 7 Final (120 days-long) discoloration and weathering of the test samples

Figure 8 shows that the change in mass that occurs in each sample is not very significant due to the absorption of water and compost in the test sample, which affects the mass of the sample. Therefore, the test sample must be dried first and weighed again. Table 2 shows the mass reduction of each test sample before and after the biodegradability test and drying. The reduction in sample mass (averaging 21.15%) indicates that the test sample has been biodegraded.

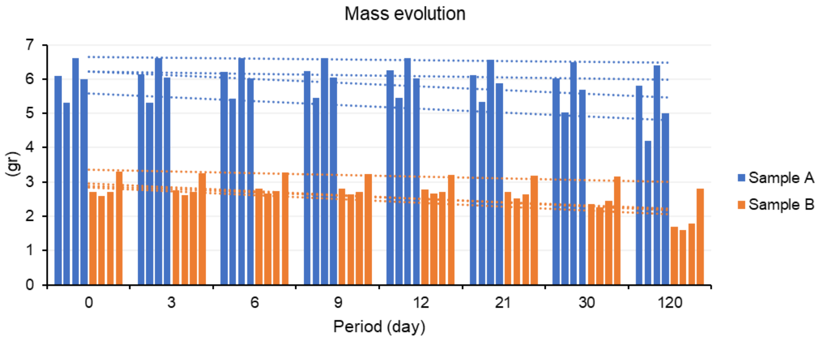
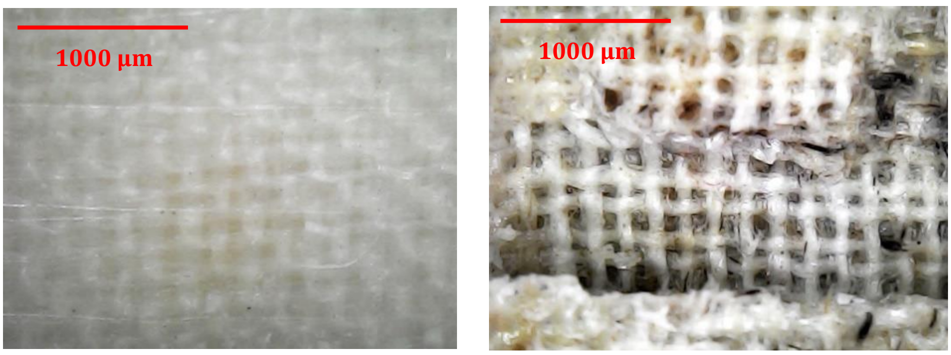


Figure 8 Biodegradability test samples' mass evolution

Table 2 Decomposition mass comparison of biodegradability test samples after 120 days

Specimen number	Mass (g) Before	Mass (g) After	Mass loss percentage
A1	6.1	5.8	4.92%
A2	5.3	4.2	20.75%
A3	6.6	6.4	3.03%
A4	6.0	5.0	16.67%
B1	2.7	1.7	37.04%
B2	2.6	1.7	34.62%
B3	2.7	1.7	37.04%
B4	3.3	2.8	15.15%
Average			21.15%

Figure 9a is a control sample stored for comparison with the biodegradability test sample. A significant difference between the control and tested samples can be seen. In Figure 9b, the test sample undergoes biological weathering, which causes the fiber layer to erode slowly. Meanwhile, Figures 9c and 9d show the presence of voids between the fibers and the surface of the sample, which is caused by the decomposition of the PLA matrix.



(a) (b)

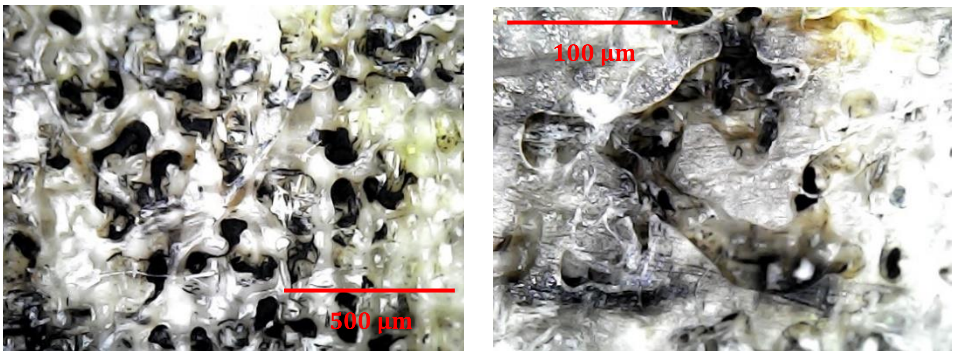


Figure 9 (a) Control sample; and (b)(c)(d) weathering and decomposition of the test sample

4. Conclusions

The fabrication and characterization of pre-impregnated RFRPLA composite were carried out. Freezer-storing (at a temperature of –18°C) a prepreg apparently can preserve and increases its mechanical properties. The tensile test found that the freezer-stored 0/90° prepregs composite had the highest average ultimate tensile strength of 71.44 MPa and had the lowest elasticity level with an average Young's modulus of 2.70 GPa. Meanwhile, the unstored 0/90° prepregs composite had the highest level of elasticity with an average Young's modulus of 1.29 GPa. In the biodegradability test, the test sample underwent weathering after 120 days, marked by a change in color and mass in the sample. The microscopic observations on prepregs showed different structures between the freezer-stored and unstored ones. In the tensile test specimen, it can be seen that there is good adhesion between the matrix and the fiber. Microscopic observations on the biodegradability test samples showed the presence of weathering and decomposition processes.

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