SUSTAINABLE HIGH-QUALITY BIO-COMPOSITE OF NOVICE NATURAL FIBER-REINFORCED POLYLACTIC ACID (PLA)

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ABSTRACT

Current biodegradable plastics have inferior properties to non-biodegradable plastics. To overcome these challenges, this research is proposing to fabricate a biocomposite using a biodegradable polymer polylactic acid (PLA) as a matrix and natural fiber obtained from Giant Ragweed (*Ambrosia trifeda)*. The addition of natural fibers into PLA is a challenge because of opposing hydrophilic and hydrophobic interfacial bonding between fiber and matrix. Experiments were conducted to reduce hydrophilic tendencies of the natural fibers by surface treatment with 2% maleic anhydride (MA) in xylene. To find the optimum fiber/matrix ratio, fibers were studied at 10 wt.% and 15 wt.%. The scope of this paper focuses on 10 wt.% due to its consistent fiber diameter compared to 15 wt.%. Single screw extrusion was employed for the filament manufacturing process, and samples were fabricated using fused filament fabrication additive manufacturing. SEM imaging was conducted to check the dispersion of fibers in the PLA matrix before and after surface treatment. Tensile and flexural properties were evaluated using ASTM standards D638 for the tensile test to study ultimate tensile strength and tensile modulus. Similarly, ASTM D790 for flexure test to study flexural strength and flexural modulus. The research has established detailed process parameters and analysis of maleic anhydride treated Ragweed reinforced PLA Biocomposites at 10 wt.% loading level.

Keywords: Bio-Composites, Natural fiber, Thermoplastics, Surface treatment, Additive manufacturing

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1. INTRODUCTION

1.1 Motivation

Industrial uses of natural fibers are growing rapidly to meet the needs of the transportation, low-cost building, and other construction industries (Sanjay, 2016) as well as medical instruments, geotextiles, molded products, filters, etc. German auto-manufacturers are the leaders in the use of natural fiber-reinforced composites (NFRC) in interior and exterior applications; their first commercial example being the inner door panel of the 1999 S-Class Mercedes Benz made of 35% Baypreg F semi-rigid (PUR) elastomer from Bayer and 65% blend of flax, hemp, and sisal (Koronis, 2013). Regarding luxury automobiles, emphasis is placed on the decision to transfer to composite materials not because of cost, but because of the desire to compete environmentally. As of 2015, glass fibers are the dominant reinforcing fiber (~95%) in the polymer matrices. Previous studies have shown that the substitution of 30% glass fibers with 65% of hemp fibers results in a net energy saving of 50,000MJ (3 tons of emission). The projected global natural fiber-reinforced composites market size is expected to increase from $4.46 billion in 2016 to $10.89 billion in 2024, according to The Natural Fiber Composites Market forecast report.

Natural fibers can be found in plants and animals. The plants, which produce cellulose fibers can be classified into bast(jute, flax, ramie, hemp, kenaf), seed (cotton, coir, and kapok), leaf (sisal, pineapple, and abaca), grass and reed (rice, corn, and wheat), and core (hemp, kenaf, and jute) as well as all other kinds (wood and roots). Qualities of natural fibers are strongly influenced by the growing environment, age of plant, species, temperature, humidity, and quality of the soil. Natural fiber advantages include lower density, biodegradability, abundant availability, good damping properties, less abrasive damage to equipment, and reduced skin irritation compared to glass fibers. Advantages of natural fiber composites include high specific strength, durability, low thermal conductivity, low density, corrosion resistance, high-impact strength, dimensional stability, and radar transparency (Puglia, 2004). Kenaf and hemp fiber bundles increased tensile strength and Young's modulus of composites; they lower the impact strength of neat PLA; thus, should be used for applications requiring high tensile strength and stiffness but low impact strength. Technical barriers to producing high-performance NFRC include heterogeneous characteristics of natural fiber leading to variation in fiber quality, lower mechanical properties than traditional fiber reinforcements, hydrophilicity leading to incompatibility and aggregation within hydrophobic polymer matrix, low thermal stability, and difficulty processing into yarns and fabrics. Improving performance of NFRCs, using fiber treatment and modification in addition to process and product innovation, will continue to push forward development, in response to consumer demand for sustainability.

1.1 Ragweed Microfibers and Fiber Retting Process

Ambrosia trifeda, or Giant Ragweed, is a native plant to North America and is known as a resilient weed crop. Its physical characteristics are like other Bast fibers such as hemp, exhibiting potential for use a natural reinforcement for sustainable composites.

Retting techniques include microbiological, enzymatic (bio scouring), mechanical, physical, and chemical. Dew retting and water retting are common practices for removing non-cellulose material from bast fiber and separating the inner core from the outer core (Konczewicz, 2018). Dew retting is widely used in industry due to its economic viability in locations with heavy night dew and warm days. The most common retting method is water retting, where stems are submerged in water for 14-28 days. This method, however, generates low-quality fibers and is less attractive due to its long-term processing and potential for water contamination. Mechanical extraction methods produce high-quality fibers but are more expensive to produce. Thus, to obtain high-quality fibers, chemical treatments used in combination with water retting is the optimal processing method. Such chemical processing is used to compliment the hydrophilic nature of natural fibers and the hydrophobic nature of polymers. There are various known treatments to reduce the hydrophilic characteristics of natural fiber, but this research will focus on the Sodium hydroxide treatment, which is the best known for reducing hydrophilic features.

The performances of the materials increase with increasing cellulose presence on the fiber surface and fiber aspect ratio (length/diameter) When the retting degree is low, impurities (pectin, lignin, hemicelluloses, etc.) between the fibers can cause stress concentrations in composites and lead to an early fracture. The low linear density of fibers ensures more uniform fiber distribution in a composite allowing for a higher aspect ratio.

1.2.1 Surface Treatment of Natural Fibers

Natural fibers have hydrophilic properties, so to enhance bonding with hydrophobic materials like PLA, surface treatment of fiber is done using Maleic Anhydride. Previous research by Hong showed surface treatment using maleic anhydride enhanced the mechanical properties of jute fiber/ Polypropylene matrix (Hong, 2008). Similarly, other articles written by S Mishra have also mentioned that maleic anhydride is a suitable surface treatment for natural fiber (Mishra, 2005). So, based on works of literature reviews surface treatment of Ragweed fiber is done using 5wt% of MA with 1Wt% of o-xylene and refluxed on a heating mantle maintaining a temperature of 50 degrees Celsius for 3 hours, then fibers were cleaned by washing with xylene to remove unreacted maleic anhydride and dried in an oven at 80 degrees Celsius for 12 hours.

1.2.2 Extraction and Grinding of Fibers

Fiber extraction was done manually after surface treatments were completed. Care was taken to obtain only outer Bast fibers. Fibers were then be dried for 72 hours and cut to approximately between 3mm to 5mm in length.

1.2 Suitable Matrix material for Biobased Material System

There are various thermoplastics available as a matrix material for fiber-reinforced composites e.g., PLA, PP, HDPE, Nylon, LDPE, PET, thermoplastic starch (TPS) etc. (Mochane, 2019). Among all matrix materials, PLA was selected as a suitable material because it has a better bonding tendency with Ragweed fiber and maleic anhydride.

1.3.1 PLA Characteristics

PLA is a bio-based thermoplastic made from the starch of corn and other plants source. The following table shows the general processing parameters of neat PLA.

Table 1. Processing parameters of PLA

Melting temperature:	210-degree Celsius
Feed Temperature:	190-degree Celsius
Die Temperature:	201-degree Celsius
Screw Speed:	20-150rpm
3D printing Temperature:	190-230-degree-Celcius
Annealing Temperature:	110-120-degree-Celcius

1.3.2 PLA/Maleic Anhydride Adhesion

Surface treatment is the most essential process for fiber/matrix adhesion (Singh, 2017). Since fibers are hydrophilic and matrix(polymer) are hydrophobic so, to have strong fiber/matrix interfacial bond surface treatment is necessary. For this research maleic anhydride is suitable as a surface treatment for both plant fiber and polymer matrix. The polymer matrix PLA, having a –COOH as a functional group, will have a better bonding tendency with maleic anhydride (Detyothin, 2013).

1.4 Manufacturing Process of Natural Fiber Reinforced Composite

Multiple different manufacturing methods were considered to produce samples. These methods include compression molding, twin-screw, and single-screw filament extrusion followed by fused deposition modeling (FDM) additive manufacturing, and injection molding. Compression and injection molding are both processes where the stock material would need to already be completely mixed with the reinforcing fibers before being liquified and injected or heated and compressed. These processes also require a die set which forms a negative cavity of the desired part when placed together, which can be designed or sourced. Another concern was if the ragweed fibers would cause binding issues with the injection molding process when filling the mold cavity.

The screw extrusion processes involve feeding metered parts of matrix and reinforcing fibers into a hopper, then one or two screws, depending on the machine forces these materials down a heated tube where they are melted and mixed before being forced out of a die and into a cooling bath. This filament can be recycled through this process to better homogenize the components of the composite. The filaments produced by these processes are not the final product but can be used as an extrusion stock for certain manufacturing processes such as filament 3D printing if they have the appropriate thermomechanical properties.

1.4.1 Single Screw Extrusion of Natural Fibers

After going through many literature topics and weighing options single screw extrusion was found to be better for the filament extrusion process. This decision was made due to potential issues with ragweed fiber reinforcements in the injection and compression molding machines, a lack of appropriate dies for either of these processes and the desire to determine the 3D-printability of composite in a fused deposition modeling 3D printer. Moreover, the requirement for a printable filament left the option of single or twin-screw extrusion for producing a printable filament. The fiber length not being appropriate on the twin-screw extruder led to believe the single screw extruder would be the most appropriate option in manufacturing bio composite filament.

Research by Bhattacharjee and Dilpreet (2018) suggests that extruding the same hybrid fiber reinforced sample multiple times degrades the fibers' mechanical properties. The paper suggested that 20wt% fiber-loaded samples would see a dramatic reduction in tensile strength and the modulus of elasticity after only two extrusions and five extrusions was the limit for a 50wt% sample. They instead suggested a twin-screw high shear mixer be used as it results in better fiber disbursement in one pass than the low shear single screw mixer. This was concerning as we believed we may not be able to use a twin-screw extruder with our fiber and would be forced to produce inadequately prepared samples.

Kraiem et al. (2013) had different results in their experiments with reed fiber reinforced HDPE. Their data seem to concur with Bhattacharjee and Dilpreet's experiments regarding single screw extrusion disbursement issues when compared to twin-screw extruders as well as fiber degradation above 200°C. However, Kraiem suggests that multiple extrusions of a sample with natural long fiber reinforcement do not harm mechanical properties after multiple extrusions and improves thermomechanical properties across the board as the number of extrusions is increased. They suggest that this is due to the single screw extruder's lack of even fiber disbursement being corrected in successive passes, and if the fiber is not pushed above its degradation temperature, they are better suited to bear the load of the composite while not suffering a loss to mechanical properties themselves.

2. EXPERIMENTATION

2.1 Ragweed Fiber Harvest /Retting Process

Ragweed natural fiber harvest took place near the Blanco River in Kyle, Texas sometime in late August. The fibers were immediately cut to the main stem of the stalk, put in a horizontal storage container, and steeped in tap water. The fibers were in the water retting process for 14 days, washed in tap water, then left in a 30% hydrogen peroxide retting process for 10 days. After the retting process, the fibers were rinsed with DI water then left to air dry on the open table.

*2.*2 Extraction and Grinding of Microfibers

After drying the ragweed stalk remains, the fiber was manually decorticated, and as much leftover pulp as possible was removed. The fibers still required more extraction from the core, so a jet sheet metal shear/brake/roll was used to press the fibers in bulk as a second attempt to extract fibers. Fibers were manually chosen at random concerning fineness. Single fibers were then set on an industrial paper cutter to be cut to 3 - 5 mm length fibers.

2.3 Maleic Anhydride (MA) Surface Treatments

Dried fibers were weighed out to 5-g and placed in a beaker. The fiber/solvent ratio of 1:20 wt./vol is estimated for 5-g fiber to 100-ml total solvent. For 100 ml total solvent, 98-ml of Xylene and 2-ml of Maleic Anhydride (MA) is prepared and maintained to account for 2% MA. Then fibers and solvent are combined into a beaker that is placed on a heating mantle for 3-hrs at

65°C. After the heating process, the fibers are filtered out and dried in the oven at 60°C until a constant weight of the fiber is achieved. Then, ragweed fibers were separated based on MA treated and untreated for the single screw extrusion process.

2.4 Single Screw Extrusion

Extrusion grade PLA pellets manufactured by NatureWorks USA and sold by Filabot were used to produce a printable filament in the single screw extruder located in the Advanced Composites Lab at Texas State. As the material was processed, there were challenges in maintaining a consistent filament diameter due to irregular spooling rates from the spooling machine, air pockets escaping from the main extruder nozzle, and trapped bits of burned material working out of the extruder tube and into the filament. The 3rd attempt yielded the most consistent filament diameter. The filament was extruded at 195°C at 30 RPM and the spooling machine performed consistently at the speed it was set, which is an imprecise dial indicator. This winding speed was the last adjustment, where increasing the speed stretched the filament more at the point of extrusion, decreasing the diameter, and vice versa. The resulting diameter, measured with a Mitutoyo digital indicator, was 1.65 $\pm$ 0.5 mm.

Figure 1. 10 wt.% Ragweed/PLA Biocomposites filament

2.5 Fabrication of Test Specimen Using Additive Manufacturing

Test specimens were manufactured on a CraftUnique Craftbot XL 3D printer. Specimen dimensions were as per ASTM standards D638 (Tensile Test) and D790 (Flexure Test) and printed with a uniform internal structure with all lines except the 3-line border along the far edges printed in the X direction, as seen in Figure 2. When testing, this resulted in all relevant internal lines aligned with the force of the test. Specimens were printed with a 0.8 mm nozzle at 215 degrees Celsius at 40 mm/s with a 0.2 mm layer height. The aluminum bed with 1 mm polyimide adhesive-backed film (trade name: Kapton Tape) was heated to 70 degrees Celsius. First layer modifications included lowering the speed to 20 mm/second. All modifications are displayed in Figure 3 below. The surface texture of the filament was also improved over the last attempt. There were no clogging issues or any print failures of any kind after switching from the default 0.4 mm nozzle to 0.8 mm. Specimens were printed separately, but in alternating pairs to keep results consistent as the concentration of ragweed might vary across the spool of filament.

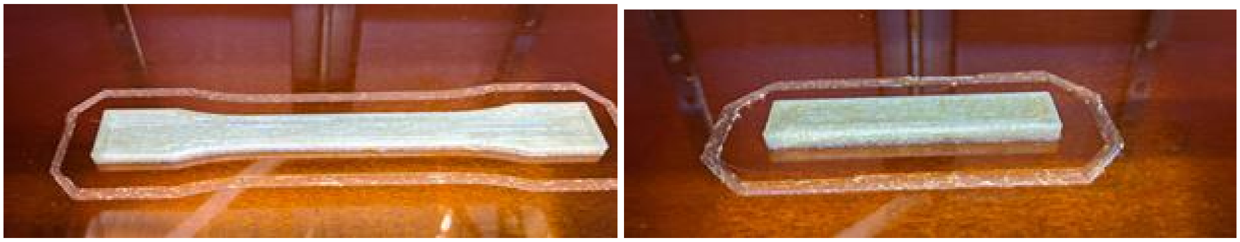

Figure 2. ASTM D638 (left) and ASTM D790 (right) test samples.

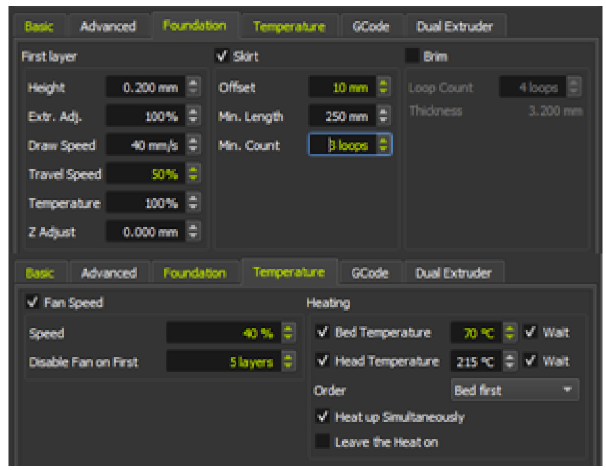

Figure 3. Printer Settings.
Yellow denotes changes from defaults

2.6 Mechanical and Morphology Test

The neat PLA samples produced with additive manufacturing had dimensions specified by the ASTM D638 tensile test and D790 flexural test. The tests were conducted per the standards as defined by ASTM. A neat, non-annealed test sample yielded an ultimate flexural strength of ≈22 MPa. Morphology of fibers is studied using JEOL SEM equipment at the ARSC at Texas State.

3. RESULTS AND DISCUSSIONS

3.1 SEM Analysis

3.1.1 SEM of Ragweed after Retting

The analysis of morphology and dispersion is still under review as better SEM is required to show the maleic anhydride effects on fiber/matrix adhesion. Sample preparation procedures, such as Imaging Sputter Coating multiple passes to achieve a high layer of carbon on the samples throughout SEM analysis. The images in the figure below were obtained using previous SEM analysis and not characteristic features of ragweed fiber. The fibers obtained in this semester's experiment were approximately 500 µm, like fibers obtained in previous experiments. With ragweed fibers, it may be inferred that the fibers will need extensive surface treatment studies due to their ridged and rough surface texture/appearance.

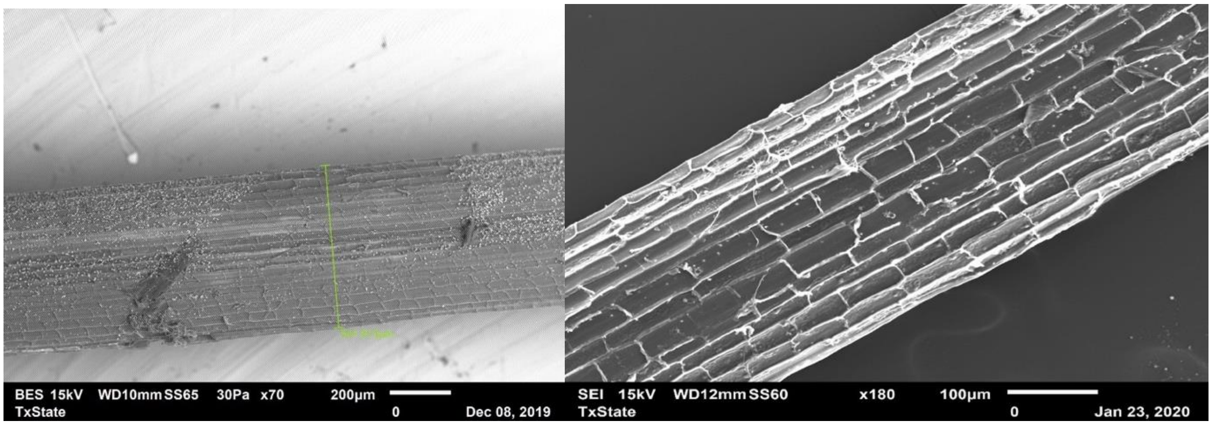

Figure 4. SEM Images

3.1.2 SEM of Neat PLA Vs. PLA/Ragweed

Figure 5. below shows a sample of PLA/Ragweed at a magnification of 30x with examples of voiding. Voiding could be due to multiple factors in the single screw extrusion process, specifically dealing with drying the reinforcement/matrix before mixing. Improper drying can lead to water boiling off during extrusion causing steam to release from the composite leaving being porous voids.

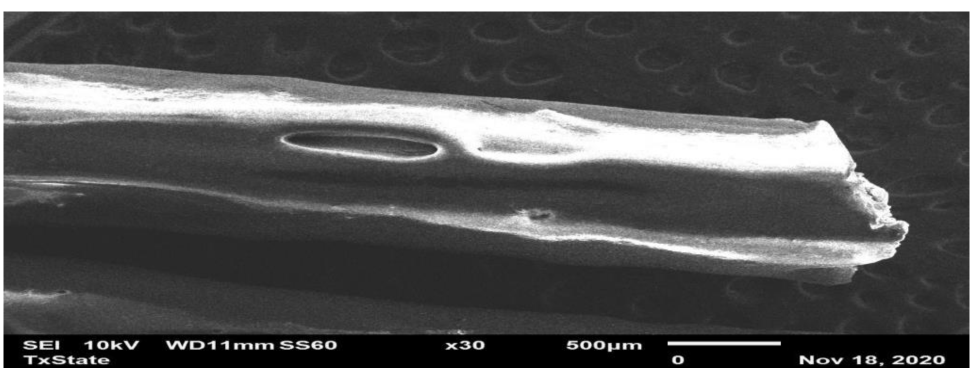

Figure 5. SEM Image

3.2 Mechanical Analysis

Mechanical testing of neat PLA and Maleic Anhydride treated PLA/Ragweed biocomposites was conducted according to ASTM standards D638 for tensile and D790 for flexure. According to ASTM D638 for the tensile test, ultimate tensile strength and tensile modulus are calculated, similarly, flexural strength and flexural modulus are calculated according to ASTM D790.

3.2.1 Tensile Test Results

Tensile Test was conducted for five samples of each neat PLA and five samples of Maleic Anhydride (MA)_10wt%_Ragweed/PLA biocomposites (Figure 7). Test was conducted as ASTM-638, on MTS servo hydraulic 810 (Figure 6). The table below (Table 2) shows the detailed test parameters, and the graph shows the average result of tensile strength and tensile modulus.

Table 2. Tensile Test Parameters- ASTM-D638

Test Name	Total Sample	Average Sample Size (mm)	Gauge Length(mm)	Test Rate (mm/min)
Tensile Test	5	Width- 13.00 Thickness- 4.00 Overall Length- 165	33.40	5

Figure 6. Illustration of tensile testing

Figure 7. 10 wt.% Ragweed/PLA-Tensile test failed Sample

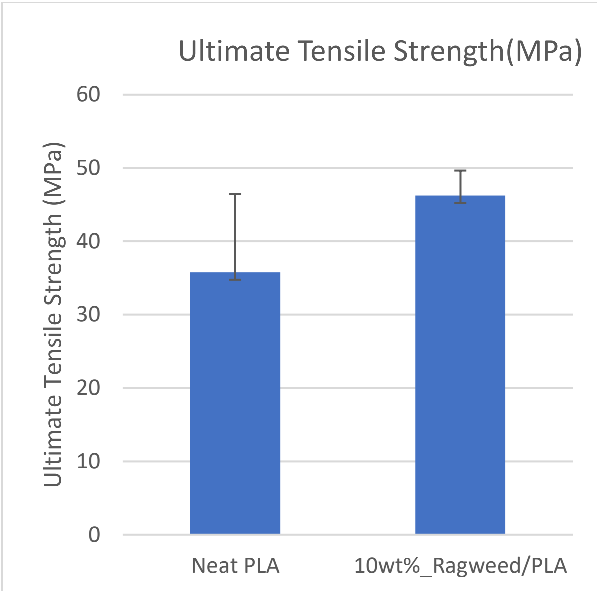

Figure 8. Tensile Strength (MPa)

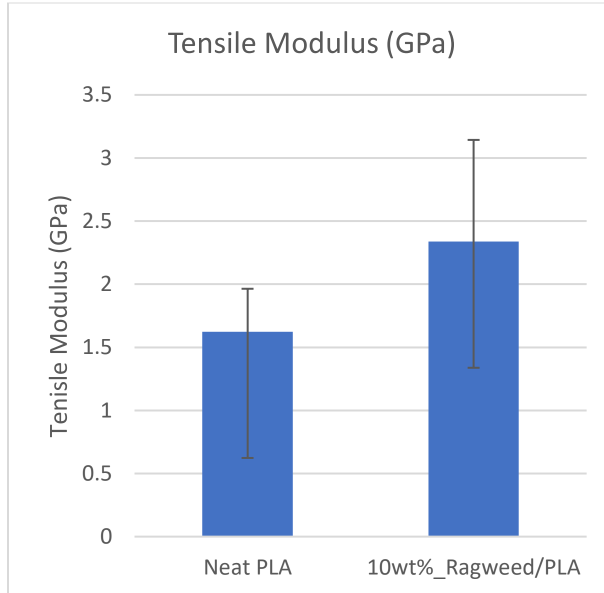

Figure 9. Tensile Modulus (GPa)

The results show the improvement of mechanical properties of 10wt% (MA treated) Ragweed reinforced PLA. On average compared to neat PLA tensile strength has increased from 38.97 MPa to 47.40 MPa. Similarly, tensile modulus has increased from 1.68 GPa to 2.34 GPa. The betterment of properties was due to Ragweed fiber reinforcement. The properties can be enhanced if double pass extrusion is performed. Overall, the properties has been increased due to the presence of Ragweed Due to better bonding adhesion of fiber/matrix is stronger hence, both the tensile strength and tensile modulus increased slightly compared to neat PLA. But, due to the effect of fiber hydrophilicity swollen filaments introduced voids and pores in 3D printed samples which reduces the strength in composites.

3.2.2 Flexural Test Results

Flexural Test was conducted for five samples of each neat PLA and five samples of 10 wt.% (MA treated) Ragweed reinforced PLA as shown in Figure 10. The test was conducted in MTS servo hydraulic 810, with (Wyoming test fixture) 3-point bend fixture as shown in figure 12 and the test failed sample shown in figure13. Details about testing parameters are listed below in the table.3

Table 3. Flexure Test Parameters- ASTM-D790

Test Name	Total Sample	Average Sample size (mm)	Span Length(mm)	Test Rate(mm/min)
Flexure Test	5	Width- 13.00 Thickness- 4.00 Length- 65.00	50.80	1.00

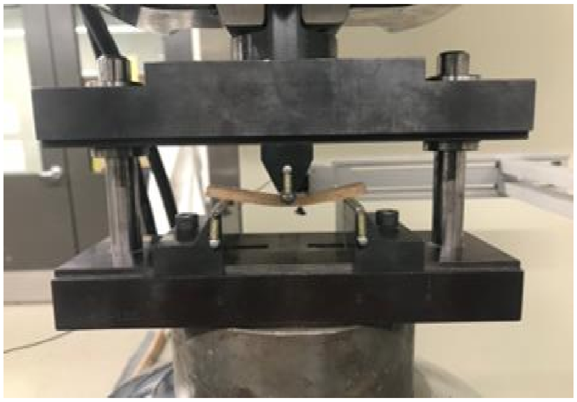

Figure 10. Flexural Test

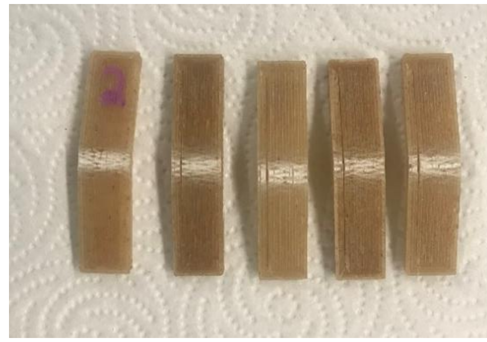

Figure 11. Failed Flexure Sample

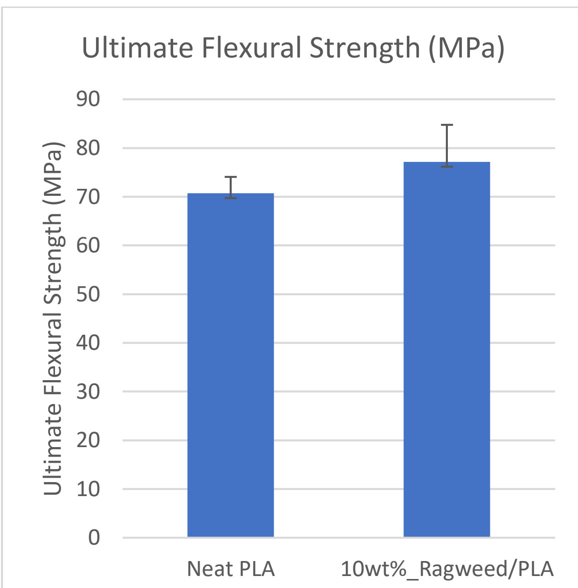

Figure 12. Flexural Strength (MPa)

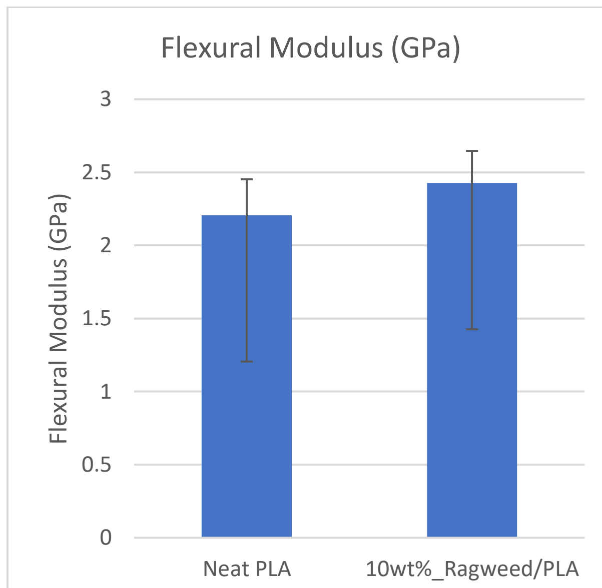

Figure 13. Flexural Modulus (GPa)

Flexural Test results showed improvement in both flexural strength and flexural modulus of Ragweed/PLA biocomposites in compared to neat PLA (Figure 12 and figure 13). In an average of five samples, the flexure strength of neat PLA is 70.72 MPa meanwhile the strength increased to 77.16 MPa on Ragweed reinforced PLA samples. So, the results showed clearly that due to better adhesion of fiber/matrix and Ragweed as a reinforcement property have been improved. Also, flexural modulus has increased from 2.21GPa to 2.43 GPa, which can be the results of ragweed reinforcement and better fiber/matrix bonding.

4. CONCLUSIONS

In general, this research accomplished the main objective exhibiting Ragweed fiber can be used as a reinforcement for biopolymer to manufacture biodegradable sustainable biocomposites. Properties like tensile modulus and flexural modulus of biocomposites were comparable with neat PLA whereas, tensile strength and flexural strength have shown a significant improvement. Improvement in properties was due to Ragweed reinforced and better interfacial bonding of Ragweed fiber/PLA. Overall, this research has shown a promising process of manufacturing bio composites, if the main concerns are given during the retting process of fiber, surface treatment of fiber, and 3D printing parameters. The future work of the research will focus more on the homogenous dispersion of maleic anhydride treated Ragweed fiber in PLA matrix and comparison of surface treated and untreated fiber.

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6. REFERENCES

1 Puglia, D., Biagiotti, J. and Kenny, J.M. (2004) A Review on Natural Fibre-Based Composites—Part II: Application of Natural Reinforcements in Composite Materials for Automotive Industry. Journal of Natural Fibers, 1, No. 3.

2 Koronis, G., Silva, A. and Fontul, M. (2013) Green Composites: A Review of Adequate Materials for Automotive Applications. Composites: Part B, 44, 120-127. http://dx.doi.org/10.1016/j.compositesb.2012.07.004

3 Singh, J. I. P., Dhawan, V., Singh, S., & Jangid, K. (2017). Study of effect of surface treatment on mechanical properties of natural fiber reinforced composites. *Materials today: proceedings*, *4*(2), 2793-2799.

4 Sanjay, M. R., Arpitha, G. R., Naik, L. L., Gopalakrishna, K., & Yogesha, B. (2016). Applications of natural fibers and its composites: An overview. *Natural Resources*, *7*(3), 108-114.

5 Mochane, M. J., Mokhena, T. C., Mokhothu, T. H., Mtibe, A., Sadiku, E. R., Ray, S. S., ... & Daramola, O. O. (2019). Recent progress on natural fiber hybrid composites for advanced applications: A review.

6 Keya, K. N., Kona, N. A., Koly, F. A., Maraz, K. M., Islam, M. N., & Khan, R. A. (2019). Natural fiber reinforced polymer composites: history, types, advantages and applications. *Materials Engineering Research*, *1*(2), 69-85.

7 Zini, E., & Scandola, M. (2011). Green composites: an overview. *Polymer composites*, *32*(12), 1905-1915.

8 Sisti, L., Totaro, G., Vannini, M., & Celli, A. (2018). Retting process as a pretreatment of natural fibers for the development of polymer composites. In *Lignocellulosic composite materials* (pp. 97-135). Springer, Cham.

9 Sanjay, M. R., Siengchin, S., Parameswaranpillai, J., Jawaid, M., Pruncu, C. I., & Khan, A. (2019). A comprehensive review of techniques for natural fibers as reinforcement in composites: Preparation, processing and characterization. *Carbohydrate polymers*, *207*, 108-121.

10 Konczewicz, W., Zimniewska, M., & Valera, M. A. (2018). The selection of a retting method for the extraction of bast fibers as response to challenges in composite reinforcement. *Textile Research Journal*, *88*(18), 2104-2119.

11 Mishra, S., & Naik*, J. B. (2005). Effect of Treatment of Maleic Anhydride on Mechanical Properties of Natural Fiber: Polystyrene Composites. *Polymer-Plastics Technology and Engineering*, *44*(4), 663–675. https://doi.org/10.1081/pte-200057814

12 Detyothin, S., Selke, S. E. M., Narayan, R., Rubino, M., & Auras, R. (2013). Reactive functionalization of poly(lactic acid), PLA: Effects of the reactive modifier, initiator and processing conditions on the final grafted maleic anhydride content and molecular weight of PLA. *Polymer Degradation and Stability*, *98*(12), 2697–2708. https://doi.org/10.1016/j.polymdegradstab.2013.10.001

13 Balla, V. K., Kate, K. H., Satyavolu, J., Singh, P., & Tadimeti, J. G. D. (2019). Additive manufacturing of natural fiber reinforced polymer composites: Processing and prospects. *Composites: Part B, Engineering*, *174*, N.PAG. https://doi-org.libproxy.txstate.edu/10.1016/j.compositesb.2019.106956

14 Hajiha, H., & Sain, M. (2015). High toughness hybrid biocomposite process optimization. *Composites Science&Technology*, *111*, 44–49. https://doi-org.libproxy.txstate.edu/10.1016/j.compscitech.2015.03.002

15 Kraiem, D., Pimbert, S., Ayadi, A., & Bradai, C. (2013). Effect of low content reed (Phragmite australis) fibers on the mechanical properties of recycled HDPE composites. *Composites: Part B, Engineering*, *44*(1), 368–374. https://doi-org.libproxy.txstate.edu/10.1016/j.compositesb.2012.04.062

16 Bhattacharjee, S., & Bajwa, D. S. (2018). Degradation in the mechanical and thermo-mechanical properties of natural fiber filled polymer composites due to recycling. *Construction & Building Materials*, *172*, 1–9. https://doi-org.libproxy.txstate.edu/10.1016/j.conbuildmat.2018.03.010

17 Hong, C. K., Kim, N., Kang, S. L., Nah, C., Lee, Y. S., Cho, B. H., & Ahn, J. H. (2008). Mechanical properties of maleic anhydride treated jute fibre/polypropylene composites. *Plastics, Rubber and Composites*, *37*(7), 325–330. https://doi.org/10.1179/174328908x314334