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Formulation of Composite Materials Using the Biomass from a Newly Developed Pineapple Leaf Fiber Extraction Process

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Abstract

The harvesting of pineapples results in a large amount of discarded pineapple leaves. However, pineapple leaf fibers (PALF) have low density, high tensile strength, and good thermal insulation properties. In this study, pineapple leaves were first pretreated, and then fibers were extracted using a newly developed hydraulic fiber extraction and degumming process machine. The goal was to use a more environmentally friendly method for simultaneous fiber extraction and degumming. The resulting biomass can be further combined with bioplastics to produce biodegradable composite materials for applications. In this study, the novel hydraulic fiber extraction and degumming process machine was tested for improving and the bioplastic samples were formulated by adjusting the ratios of different bioplastics with the pineapple leaf residue biomass powders. The tensile strength and the elongation at break of the pineapple leaf residue composites were optimized for the composition using the response surface methodology (RSM). The optimal tensile strength of 15.48 MPa and elongation at break of 12.71% were achieved with a composition of 60% polylactide (PLA), 15% polybutylene succinate (PBS), 10% polybutylene adipate terephthalate (PBAT), and 15% pineapple leaf residue biomass in the composites.

Keywords

Pineapple Leaf Fiber, Composite, Biodegradable, Bioplastic

1. Introduction

Generally speaking, most farmers will burn or leave pineapple leaves to decompose naturally. However, improper burning will not only cause air pollution, but also have a negative impact on the environment. Another treatment method is to bury pineapple leaves in the field and use it as fertilizer. However, this method is not only time-consuming and labor-intensive, but also difficult to decompose quickly due to the hard skin layer of pineapple leaf, so it is not a good fertilizer compared with other microbial green manures.

Pineapple leaf fiber is mainly composed of cellulose, hemicellulose, lignin and pectin [1]. Common fiber extraction methods include manual fiber extraction [2], mechanical fiber extraction [3], and retting fiber extraction [2], enzyme fiber extraction [4], chemical fiber extraction [5]. The hydraulic fiber extraction method was developed for this study. In order to solve the problem of pineapple leaves and promote sustainable development of the environment, we can process these agricultural by-products through the following steps to increase the economic value of the entire pineapple industry. First, the pineapple leaves are extracted through the proposed fiber extraction machine, and the fiber can be further processed into products such as textiles, paper or composite materials. The residue after fiber extraction can also be used as the biomass materials. This can not only solve the problem of agricultural waste treatment, but also increase the value of the pineapple industry and achieve the purpose of reuse.

A large amount of waste has obviously become a serious problem, especially agricultural wastes and plastics, which have caused a great influence on the environment and human health. The plastic wastes have caused damage to the natural environment and habitats of animals and plants. By the way, plastic particles can be ingested by humans through the food chain in different ways, causing various complications or neurological problems [6]. In this study the agricultural wastes are also tried to be mixed with biodegradable plastics to reduce the amount of wastes and the cost of biodegradable plastics.

Pineapple spread rapidly from South America to all parts of the world and Southeast Asia is also one of the major pineapple growing area. Pineapple leaf fiber (PALF) is a natural columnar fiber with excellent antibacterial, deodorizing and mechanical properties. Moreover, it is extracted from agricultural waste and other characteristics, making it one of the materials that people are actively researching. The main components of PALF are composed of cellulose, hemicellulose and lignin [1]. Therefore, after being extracted, PALF can not only replace chemical fiber as a spinning raw material to make fabrics, but can also be recycled. It can be processed into paper and environmentally friendly packaging materials, or combined with other materials to form composite materials. Since it can increase the tensile strength and bending strength of composite materials, it is very suitable for use in automobile interiors [7]. At present, PALF has not been widely used. As people's demand for environmentally friendly materials increases, PALF has gradually become a hot emerging material. The pineapple leaf residue from the PALF manufacturing process also needs to find applications.

Biodegradable plastic refers to a type of plastic that can naturally decompose in the environment. The plastic is decomposed into natural elements such as water, carbon dioxide, and biomass through the action of microorganisms such as bacteria, fungi, or algae. The raw materials of biodegradable plastics come from biomass (e.g. pineapple leaves) and natural polymers, such as corn, cassava and sucrose. Finally, other biodegradable bio-based plastics are added to adjust their mechanical properties for application in different places, we use renewable resources to produce plastics and find alternatives to traditional plastics. However, the current usage rate of biodegradable plastics is low, mainly for the following two reasons. The first is that the production cost is about twice as high as that of traditional plastics, and the mechanical properties may be worse than traditional plastics, resulting in low market acceptance. The second is that although it is advertised as "biodegradable", the fact is that biodegradable plastic requires "specific temperatures and environments" for a period of time to decompose, and there is no good collection and classification system for recycling. As a result, it is mixed with other plastic waste and increases recycling costs [8].

The objectives of this study were to propose a newly developed process machine of PALF extraction and formulate the biodegradable bioplastic composites which were made by adjusting the ratios of pineapple leaf residue biomass powders with bioplastics. The tensile strength and the elongation at break of the pineapple leaf residue composites were optimized the composition using the response surface methodology (RSM).

2. Experiment

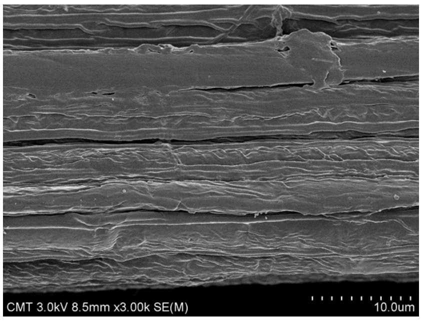
Most of the pineapple leaf fibers on the market are obtained by scraping, but the scraped fibers cannot completely remove the pectin in between the fibers. Therefore, the scraped fibers will be reused with chemicals for further degumming, cleaning and bleaching steps. Although unnecessary impurities on the fibers can be removed, the fibers will be damaged to a certain extent during the process, making the fiber quality degraded or easier to break. If by-products such as industrial wastewater are not properly treated, it may lead to more serious environmental problems. Therefore, in order to extract higher quality PALF and reduce the emission of process waste, this experiment developed a new hydraulic fiber extraction and degumming process machine as shown in **Figure 1**. After the pineapple leaves are sprayed with high-pressure water, the pineapple leaf fibers can be extracted and the gum in between fibers can be partially removed. The parameters of the hydraulic fiber extraction machine can be adjusted to achieve the optimal fiber extraction process under the conditions of conveyor belt conveying speed of 100 rpm, water jet swing frequency of 65 times/min and water column pressure of 40 kg_f/cm^2. The water conservancy system in the experiment will filter the water after the fiber is sprayed. It is then reused in a water cycle to achieve the purpose of saving water resources.

In this study, a chemical solvent method was used to prepare biodegradable plastic samples mixed with pineapple leaf residues. Because this method has relatively easy to operate in the laboratory, this method was selected to prepare the samples. Put different compositions of PLA, PBS, PBAT, and pineapple leaf residue into the sample bottles, and then put them in a vacuum oven at 100˚C for 24 hours to dry the materials to avoid containing moisture, which would affect the accuracy of the experiments. Pour the materials from the sample bottle into the beaker, use a micro-balance to weigh the required contents of the four materials respectively, then pour the weighed materials into a 100ml beaker, and then add 22.5 ml chloroform to the beaker to prepare a 10%wt solution. After placing the magnet into the beaker, seal it with aluminum foil to prevent the solution from evaporating. Finally, place the beaker on an electromagnetic heating stirrer and set the temperature to 30˚C and 300 rpm for 1 hour of stirring. After it is completely mixed, pour it into the sample plate, and after it is completely dry, the required plastic sample can be prepared. The prepared sample is then mechanically stretched using a mechanical strength testing machine, and the tensile strength and elongation at break are measured to understand the properties of the composites.

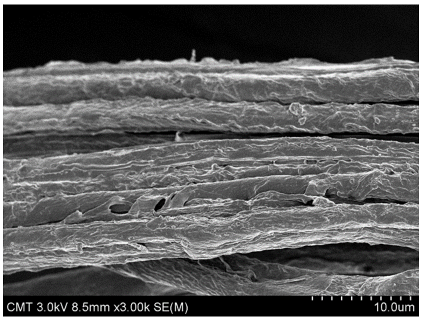
Figure 1. The developed hydraulic fiber extraction and degumming machine.

3. Result and Discussion

In order to confirm that the hydraulic extraction technology for pineapple leaves in this experiment can extract finer PALF, an electron microscope (SEM) was used to observe the surface characteristics. It can be observed from **Figure 2(a)** that the fibers taken out by the mechanical scraping method are basically in the form of bundles, with an average fiber diameter of 12.3 μm, and there are many other substances adhering to the fiber surface. **Figure 2(b)** is an SEM image of the PALF extracted by the hydraulic extraction machine developed in this study. The fiber in **Figure 2(b)** is obviously more dispersed and the average fiber diameter is about 4.7 μm, indicating that this method can more effectively remove pectin and lignin outside the fiber, and obtain more dispersed and finer pineapple leaf fibers.



(a)



(b)

Figure 2. SEM images of PALF from (a) mechanical and (b) hydraulic extractions.

In this study, we tried to use the pineapple leaf residue biomass to produce biodegradable composites. The experiment planning and discussion were conducted by using response surface methodology (RSM) to analyze the proportion of PLA and PBS, PBAT ratio. The composition ratios of the above bioplastics were selected to be the variables for the optimization test to find the best tensile strength and elongation at break of the composites. Finally, the content of pineapple leaf residue was increased to observe whether good mechanical properties can be maintained.

Three mass fraction factors of PLA content (X_1), PBS content (X_2), and PBAT content (X_3) were selected and the tensile strength (Y_1) and elongation at break (Y_2) of the composites were set as the target of this experiment. The operating range of the parameter factor was normalized to represent 3 different level values and the 15 sets of experiments designed using the Box-Behnken model of the response surface methodology (RSM) were shown in **Table 1**.

The tensile strength (Y1) data of the 15 runs of experiments designed by the Box-Behnken model are shown in **Table 1**. In the 15th run of experiments (PLA ratio is 0.6, PBS ratio is 0.15, and PBAT ratio is 0.10), the highest tensile strength reaches 15.48 MPa. From the polymer characteristics, it can be seen that PBS is an aliphatic compound and has a higher crystallinity between molecules than PBAT. Therefore, the better the tensile strength can be obtained when the PBS ratio is higher. In the 13th set of experiments (PLA ratio 0.6, PBS ratio 0.10, and PBAT ratio 0.15), the highest elongation at break reaches 12.71%. From the polymer characteristics, it can be seen that the presence of aromatic rings in PBAT will hinder the crystallization between molecules. The decrease in crystallinity makes the sample more flexible and ductile. Therefore, a better elongation at break can be obtained when the PBAT ratio is higher.

It can be observed from the results of **Table 2** that the P-values of PLA ratio (X_1) and PBS ratio (X_2) are less than 0.05, which means that both parameters have a significant impact on tensile strength. However, due to the P-value value of PLA ratio (X_1) is smaller than the P-value value of PBS ratio (X_2), which means that X_1 is a more significant factor. As for the interactive effect, the quadratic effect of the PLA ratio (X_1) and the quadratic effect of the PBAT ratio (X_3) both have a slight influence, while other factors have less obvious influence.

Table 1. Experimental results of this study.

Run	Pattern	X_1	X_2	X_3	Y_1(MPa)	Y_2(%)
1	+-0	0.6	0.05	0.10	13.26	11.69
2	000	0.5	0.10	0.10	11.20	9.14
3	000	0.5	0.10	0.10	11.05	9.14
4	+0-	0.6	0.10	0.05	13.20	10.00
5	-0-	0.4	0.10	0.05	7.12	8.11
6	000	0.5	0.10	0.10	10.12	9.60
7	0--	0.5	0.05	0.05	7.09	7.16
8	0+-	0.5	0.15	0.05	11.03	10.04
9	-0+	0.4	0.15	0.10	9.65	10.45
10	0++	0.5	0.15	0.15	10.40	10.40
11	0-+	0.5	0.05	0.15	8.08	10.50
12	-0+	0.4	0.10	0.15	7.25	9.34
13	+0+	0.6	0.10	0.15	13.48	12.71
14	--0	0.4	0.05	0.10	6.70	8.60
15	++0	0.6	0.15	0.10	15.48	12.20

Table 2. The significance of each factor on Y_1.

Term	Estimate	Standard Error	t Ratio	P-value
Intercept	10.79	0.2358	45.75	<.0001*
X_1	3.0875	0.1444	21.38	<.0001*
X_2	1.4288	0.1444	9.89	0.0002*
X_3	0.0963	0.1444	0.67	0.5346
X_1X_2	−0.1825	0.2042	−089	0.4125
X_1X_3	0.0375	0.2042	0.18	0.8615
X_2X_3	−0.405	0.2042	−1.98	0.1042
X_1X_1	0.7975	0.2126	3.75	0.0133*
X_2X_2	−0.315	0.2126	−1.48	0.1985
X_3X_3	−1.325	0.2126	−6.23	0.0016*

Figure 3 shows the main effect of each factor on tensile strength (Y_1), mainly exploring the correlation of a single factor with tensile strength. It can be observed from the trend in the figure. As the factor of PLA ratio (X_1) increases, the tensile strength increases. It can be seen that the PLA ratio (X_1) has a significant impact on it; as the factor of PBS ratio (X_2) increases, the tensile strength also increases slightly. it can be seen that the influence of the PBS ratio (X_2) is also significant; while the PBAT ratio (X_3) has no significant change in the tensile strength, which

can be verified from **Table 2** also. The PLA ratio (X_1) and PBS ratio (X_2) are all significant factors, while the influence of PBAT ratio (X_3) is smaller in comparison as shown in **Table 2**.

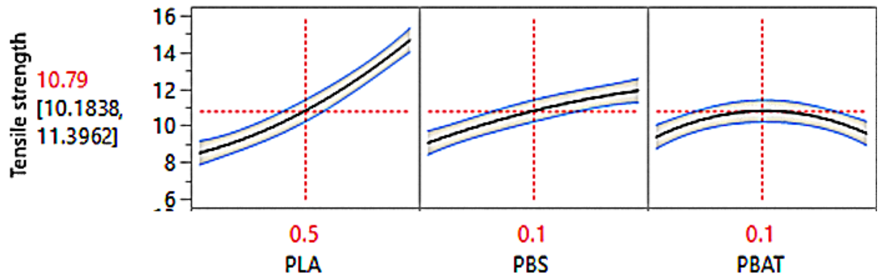


Figure 3. Main effects of each factor on tensile strength.

Table 3. The significance of each factor on Y_2.

Term	Estimate	Standard Error	t Ratio	P-value
Intercept	9.2933	0.1132	82.06	<0.0001*
X_1	1.2625	0.0693	18.21	<0.0001*
X_2	0.6425	0.0693	9.26	0.0002*
X_3	0.955	0.0693	13.77	<0.0001*
X_1X_2	−0.335	0.0981	−3.42	0.0189*
X_1X_3	0.37	0.0981	3.77	0.0130*
X_2X_3	−0.745	0.0981	−7.6	0.0006*
X_1X_1	0.9783	0.1021	9.58	0.0002*
X_2X_2	0.4633	0.1021	4.54	0.0062*
X_3X_3	−0.2317	0.1021	−2.27	0.0725

From the results in **Table 3**, the P-values of PLA ratio (X_1), PBS ratio (X_2), and PBAT ratio (X_3) are all less than 0.05, which means that these three parameters have an impact on the elongation at break. The P-values of both PLA proportion (X_1) and PBAT proportion (X_3) are less than 0.0001, indicating that these two parameters are more significant factors, while the influence of PBS ratio (X_2) is smaller in comparison. As for the interaction effect, except for the secondary effect of the PBAT ratio (X_3), the rest have significant effects.

Figure 4 shows the main effect of each factor on elongation at break (Y_2), mainly exploring the correlation of a single factor with elongation at break. It can be observed from the trend in the figure that as the factor of PLA ratio (X_1), PBS ratio (X_2), and PBAT ratio (X_3) increase, the elongation at break increases. From **Table 3**, we know that PLA ratio, PBS ratio, and PBAT ratio are all significant factors, and the influence of PLA ratio and PBAT ratio is more significant.

From **Table 1**, it is found that the sum of the best mixed ratios of PBS and PBAT is 0.25. The PBS ratio is tested in this research and it can be seen that PBS

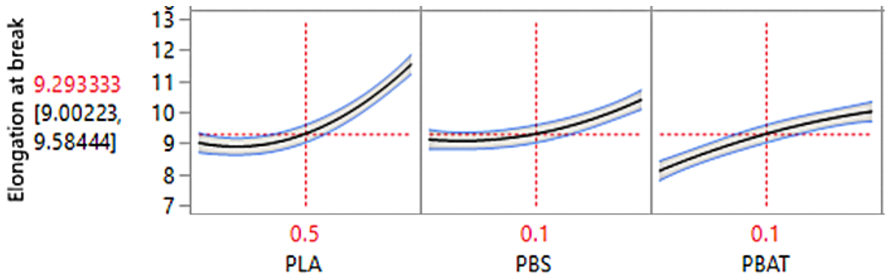


Figure 4. Main effects of various factors on elongation at break.

ratio is the significant factor on the responses of tensile strength and elongation at break, while the PBAT ratio is only a significant factor on the response of elongation at break. Therefore, the PBS ratio was set at 0.15 and the PBAT ratio was set at 0.1 in this experiment when the ratio of pineapple leaf residue was increased to observe the changes of the tensile strength and the elongation at break. It can be observed that as the ratio of pineapple leaf residue increases, the tensile strength and elongation at break have a downward trend. Due to the lower content of bioplastic results in a material that is brittle and difficult to shape and appears powdery, making it impossible to detect its mechanical properties when biomass ratio is above 65%. Although the tensile strength and elongation at break both have a downward trend, the elongation at break decreases more slowly. Since the particle size of the pineapple leaf residue currently used is below 150 mesh, if the blending ratio of the biomass is above 50%, the mechanical properties will be reduced significantly. The tensile strength is reduced to 2.46 MPa and the elongation at break is reduced to 7.46% when the biomass ratio is 55% in the composites. However, the highest tensile strength is 15.48 MPa and the highest elongation at break is 12.71% in this study. Therefore, unless more cement can be added or the biomass can be ground to make it finer, there is no way to increase the proportion of pineapple leaf residue biomass mixed in while still maintaining a certain degree of mechanical strength.

4. Conclusion

In terms of making biodegradable composites using PALF or residue biomass powders from the hydraulic fiber extraction machine successfully developed in this study, by adjusting the different ratios of PLA, PBS, and PBAT, and using the response surface methodology to discuss. It can be found that the PLA ratio is a significant factor in the responses of tensile strength and elongation at break. The PBS ratio has a smaller P-value in the discussion of tensile strength and it can be seen that it is the key factor to tensile strength. In the elongation at break, the PBAT ratio has a smaller P-value and its influence is more significant. It has been verified by mechanical strength testing and it shows the feasibility of using pineapple leaf biomass powders in biodegradable plastics.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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