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Biodegradability of PLA and Tea Waste Composites Based on “CHAMU” and the “Tea Waste Recycling System”

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Biodegradability of PLA and Tea Waste Composites Based on “CHAMU” and the “Tea Waste Recycling System”

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Abstract. This paper proposes and explains the concept of “Chinese Pinyin of tea and wood (CHAMU)” and the establishment of the “Tea Waste Recycling System” for mitigating the problem of tea waste disposal in the tea production process. Based on the “reuse of fully biodegradable waste” proposed in the “Tea Waste Recycling System,” this paper investigates existing biodegradation studies related to plant materials and analyzes the factors that contribute to the degradation of plant material. This paper analyzes the composition of tea waste and establishes a theoretical basis for the biodegradation of tea waste. The soil-buried method was used to test the degradation performance of samples prepared from polylactic acid (PLA) and tea waste composites made by injection molding. This paper mainly focuses on the experiment and discussion of the biodegradability of “CHAMU” materials and considers a theoretical basis to assist in the establishment of a tea waste recycling system.

1. Introduction

Tea, cocoa, and coffee are the world's three major non-alcoholic beverages ^[1]. Tea production has shown a steady growth trend in the past two decades; according to the latest statistics posted by the International Tea Board, the total tea production in 2017 is 5.686 million tons, a year-on-year growth of 2.6% ^[2]. The Twenty-third Session of the Intergovernmental Group on Tea (FAO-IGG/Tea) predicts that tea production will continue to grow at a rate of 2.2% over the next decade ^[3]. How to effectively treat tea waste in the tea production process has become a major problem for tea producers. Tea waste refers to the tea pruning branch, the tea stem picked out during the processing of the tea, and the tea dregs produced during tea beverage production. The current treatment of these wastes is direct disposal or in situ combustion, and only a small part is used in scientific research. In a survey of a tea beverage manufacturer in Japan, it is found that if the tea dregs produced by a tea beverage factory are not treated in time, the tea dregs will rot and deteriorate after being stacked for two to three days. Therefore, the factory must pay nearly one million yen per month to process the discharged tea dregs ^[4].

Previous studies on the reuse of tea waste are mostly concentrated in the agriculture and chemical industry fields. For example, some studies focus on using tea waste as a raw material for bioorganic fertilizers and as an animal feed additive to reduce fat deposition and improve disease prevention ability. Tea waste itself is a porous structure with good adsorption properties. Researchers use this feature to use tea dregs as a raw material for biosorbents ^[5]. In addition to researchers, after the tea



farmers have processed the tea, they will bundle the selected tea leaves and tea stems into bags for sale. Placing tea waste in newly decorated rooms or new cars can play a role in absorbing peculiar smells.

The author's research is dedicated to making tea waste available for use in people's daily lives through material innovation and design. To improve the utilization efficiency of tea waste the concept of "Chinese Pinyin of tea and wood (CHAMU)" is proposed based on the characteristics of tea waste. "CHAMU" refers to the tea stem and tea residue produced in the process of tea production and processing, which can then be used as a product after mechanical reprocessing as shown in Figure 1.

In past experiments, the authors used the characteristics of tea stems rich in plant fibers in tea waste and prepared an artificial board based on tea stems by using a high-temperature hot pressing process (as shown in Figure 2).



Figure 1. "CHAMU" reprocessed products.



Figure 2. "CHAMU" artificial board.

Through the mechanical testing machine test, as shown in Table 1, the mechanical indicators are in line with national standards ^[6].

As shown above, the study proves that tea waste has processability as a raw material. To prove that tea waste is also a sustainable environmentally friendly material, the authors will analyze the degradation properties of tea waste as a plant material in the following study.

With the development of the plastics industry and the increase in the consumption of plastic products, the total output of plastics in the world has exceeded 120 million tons, and the amount of waste plastics in the world has exceeded 40 million tons. Most of the waste plastics are landfilled and incinerated, and the recycling rate is less than 10%, causing a serious burden on and pollution of the environment ^[7]. After noticing the pollution problem, people began to turn their attention to the research of new biodegradable materials. Shanshan Lv et al. ^[8] summarized the biodegradable plastics that have been developed so far; these include polyhydroxybutyrate (PHB), polyhydroxyvalerate (PHV), polybutylene succinate (PBS), polycaprolactone (PCL), PLA, etc.

Among all biodegradable plastics, PLA has quickly become a research hotspot because its raw materials are natural renewable materials and have good biodegradability and mechanical properties. However, due to the high price and high brittleness of PLA, it has not been widely used in daily life. The use of plant materials as fillers to prepare PLA materials can increase its mechanical properties, shorten the degradation cycle, and reduce the processing cost; it has become a trend in this field in recent years.

Table1. Test report on properties of wood-based panels.

Sample name	Tea Waste Particleboard		Sample specification	320X340X13mm
			Inspection basis	GB18580-2001 GB/T4897.3-2003
Inspection item	Unit	Standard value	Test result	Result judgment
Density		0.4-0.9	0.9	Meet the standard requirements
MOR	Mpa	$\bar{x} \geq 13$	$\bar{x}: 13.2$	Meet the standard requirements
		$x_{\min} \geq 10.4$	$x_{\min}: 10.8$	
MOE	Mpa	≥ 1600 (Thickness 13-20mm)	1632	Meet the standard requirements

Yang Zhang et al. ^[9] find that because the plant fiber itself is highly hydrophilic, it is easy to promote the hydrolysis of the PLA material itself. No matter what degradation method is used under the same degradation conditions, PLA composites with added plant fibers have better degradation properties than pure PLA. Xia Zheng et al. prepared bamboo fiber composite plastics by mixing bamboo fiber with PLA at a ratio of 50 to 50. Through the 12-month soil-buried experiment, it was found that the weight loss rate of the composite material reached 8.87%, and impact strength and tensile strength of the composites were reduced by 44.0% and 43.8%, respectively ^[10]. Wenjing Guo ^[11] proved via a 6-month soil-buried experiment that the Pinus kesiya fiber PLA composite with 70% addition has a higher degradation performance than the general PLA material and also proved that under the same material ratio, the addition of a compatibilizer can control the degradation rate of the composite to suit the needs of different environments.

Plant fiber material as a filler can increase the degradation rate of plastics due to the addition of the plant fibers expanding the contact surface of the plastic with the air, thus increasing the permeability of the air. There are many easily degradable organic substances, such as proteins, lipids, and cellulose in plant fibers. Yan Wang ^[12] used ramie fiber to make tableware instead of disposable plastic tableware. The degradation rate of ramie fiber tableware reached 60.59% during the 28-day degradation period by the activated-sludge method. Fudong Xu and Xiulun Wang ^[13] used sago tree fiber to make a biodegradable artificial board; it has been proved by mechanical tests that it conforms to the national (China) strength standards for MDF and particleboard. Jin Zhang ^[14] made a fully degradable biomass board by using rice straw.

According to Tuan Su's research ^[15], the chemical composition of tea is generally composed of 93–96.5% organic matter and 3.5–7.0% inorganic matter. It mainly contains proteins, amino acids, tea polyphenols, alkaloids, and other substances. According to the research on the tea dregs processed by the tea beverage factory, such as the study conducted by Ayako Takeshita ^[16], the nutrients remaining in the tea dregs were found such as carbohydrates, proteins, lipids, food fibers, and a variety of vitamins. Based on this feature, Fumio Okada et al. ^[17] accelerated the composting rate and improved the quality of the fertilizer by mixing the tea residue with the chicken manure.

Tea dregs and tea stems have different composition components; tea dregs have more water-soluble substances, and tea stems have more plant fiber content. The main components of tea stems are cellulose, hemicellulose, and lignin, as well as some tea polyphenols, theaflavins, and caffeine. The “CHAMU” mentioned in the foregoing is a composite material, and the main source of the molding strength is the plant fiber present in the tea waste. This study divides tea waste into two categories: tea dregs and tea stems. The tea dregs are mainly composed of tea leaves, and the tea stem inclusions are not more than 5%.

As a kind of plant material, tea waste also contains substances such as cellulose which are easily decomposed. The mixing process with PLA plastics should also reduce production costs and improve biodegradability.

Through investigations into existing research, research on plant materials is more concentrated on crop straw, and research on tea waste is very limited. This study will focus on the preparation and biodegradability of tea waste and PLA plastics.

The tea waste has been mixed with polylactic acid (PLA) plastics in fiber and powder form, and the preparation of the tea composite plastics is achieved via a single screw extruder in subsequent experiments. Experiments show that tea waste can be used as a filler to reduce the production cost of PLA plastics. This experiment will be described in detail later.

2. Experimental method

Experiment 1: Preparation of PLA and tea waste composite materials.

Experimental method: Experimental samples are prepared by hot-melt extrusion-injection molding and are tested for mechanics of materials.

Experimental materials: PLA granules, tea waste powder, tea fiber.

Experimental equipment: SJ-20X25 plastic extruder, pulverizer, injection molding machine, mechanical testing machine.

Experimental steps: First, the tea waste is mixed as a filling material with PLA particles in three different proportions (as shown in Table 2).

Pour the material when the extruder temperature is heated to 165 degrees. After high temperature extrusion molding, the semi-finished product shown in Figure 3 is obtained. The three kinds of semi-finished product obtained by the experiment were further subjected to pulverization, drying, and injection molding to obtain the final experimental sample shown in Figure 4.

Experiment 2: Degradation performance analysis of PLA and tea waste composites.

Experimental method: Soil burial method.

Experimental materials: Three kinds of PLA and tea waste composites.

Experimental equipment: Outdoor garden soil.

The soil contains a variety of microorganisms that can degrade plant material. In Hongmei Zhang's research^[18], a corn field in Jilin Province is used as the experimental object. The study found 54 kinds of cellulose decomposing fungal strains in the soil depth of 40 cm. Allgaier et al.^[19] found in research on improving the efficiency of plant composting that during the composting process the physical and chemical properties of the materials, especially the temperature and pH, undergo drastic changes. Therefore, it can be proved that a series of actinomycetes with strong adaptability, such as heat resistance and alkali resistance, must exist in the composting process. Hui Deng's study^[20] found that actinomycetes in the composting process secrete a cellulolytic enzyme that has different degrees of degradation on cellulose, hemicellulose, and lignin in plant fibers.

For the degradation test of PLA and tea waste composites, the method of outdoor soil burial is adopted. The use of microorganisms to decompose materials is mostly in line with the requirements of materials for the tea waste recycling systems. After use, the PLA and tea waste composite can be completely decomposed under natural conditions. Using outdoor garden soil as experimental soil, three different experimental samples were buried in the soil. The test period is 4 months. Every month, the experimental sample is taken out, washed, and dried, and the sample is weighed to measure the weight loss rate.

Table 2. Samples and proportions.

Sample	1	2	3
Proportion	50%tea drags50%PLA	50%tea fiber50%PLA	10%tea drags90%PLA



Figure 3. PLA and tea waste composite.



Figure 4. PLA and tea waste composite samples.

3. Experimental result

The text of your paper should be formatted as follows: Through the forming experiments of three kinds of composite materials, it can be found that all materials can be smoothly passed through a screw extruder and be injection molded at a high temperature. In terms of mechanical strength testing, the shear strength of the three samples is determined by the Universal Testing Machine (UTM). The results are shown in Table 3.

As can be seen from Table 3, the more tea waste powder is added, the more the mechanical strength of the composite material decreases. This indicates that the tea stems or tea dregs in the powder state cannot form strength. The composite material with added tea fiber is slightly better than the tea waste powder in terms of mechanical strength. This is because the tea fibers are intertwined during processing to provide a certain strength. However, due to the influence of the shape and processing method of the tea fiber, the improvement of mechanical strength is not obvious. Therefore, more attention should be paid to the preparation of high-strength composite materials in the future.

The Soil burial method experimental results are shown in Table 4. As shown in Table 4, the first is the total weight loss rate for four months. All three samples are degraded to varying degrees. In the previous experiments using the soil burial method for PLA materials, PLA plastic without any filler is hardly degraded in the first month of soil burial, and the total weight loss rate within four months does not exceed 10%.

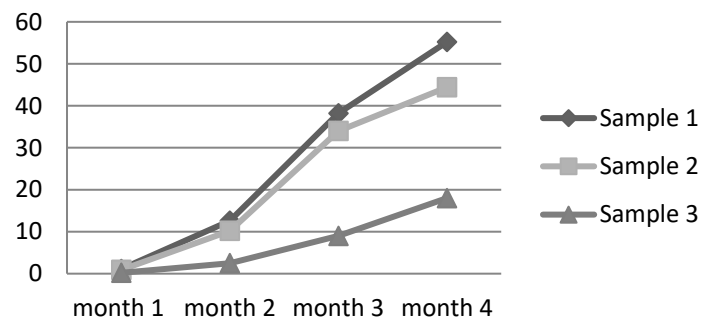
Table 3. Mechanical test results of PLA and tea waste composites.

	Sample 1	Sample 2	Sample 3
Position (mm)	1.5	1.57	3.36
Maximum force (N)	1202.09	1268.65	2249.49
Internal bond strength (Mpa)	26.68	29.01	51.44

Table 4. Sample weight loss rate.

	Sample 1	Sample 2	Sample 3
Weight loss rate	55.2%	44.4%	18%

Table 5. Four months Sample weight loss rate.



As can be seen from Table 5, the three samples degraded to varying degrees within four months. Among them, the degradation rate of sample 1 and sample 2 is relatively large and the rate of sample 3 is small. The experiment also proved that the higher the proportion of tea waste, the higher the degradation rate.

Specific analysis of the experimental data per month shows that the degradation properties of the material also change with the extension of the experimental time. No significant degradation occurred in the three samples during the first month of the experiment, and the weight loss rates are 1.1%, 0.9%, and 0.2%. Analyze the reason, first of all, in the past experiments the degradation performance of the PLA surface layer is lower than the internal, this is because the PLA material itself has hydrophobic. The hard surface of the composite material caused moisture and the microorganisms could not quickly destroy the surface of the material. Secondly, the outdoor temperature is low during the first month of the experiment, and the low temperature affected the activity of microorganisms in the soil and reduced the hydrolysis performance of the PLA material.

From the second month of the Table 5, it can be clearly seen that the degradation rate of the sample is significantly improved, especially in the third month, the degradation rate of sample 1 and sample 2 is the fastest. This is because the internal tea waste is easily decomposed by microorganisms after the surface layer of the material is destroyed, and the pores generated inside the material after decomposition can further promote the hydrolysis of the PLA material and accelerate the degradation rate of the material.

After 4 months of soil burial experiments, the mechanical strength tests were again performed on the three samples. The test results showed that the mechanical strengths of the three samples showed different degrees of decline, as shown in Table 6.

According to the abovementioned experiment it can be proved that tea waste as the filler can effectively increase the degradation rate of the composite material. As a plant fiber material, tea waste itself is highly hydrophilic and can help promote the hydrolysis of PLA materials. In terms of the physical properties of the material, tea waste as a filler can increase the surface pores of the PLA material and increase the contact area of the microorganisms with the material to promote microbial degradation. In addition, compared to other plant raw materials, the lignin content in tea waste is low and lignin is difficult to be degraded. Jingyu Dai ^[21] analyzed the chemical composition of tea, Chinese milk vetch, and straw and found that the lignin content of straw is higher than that of tea and Chinese milk vetch. In terms of degradation properties, straw is difficult to degrade due to excessive lignin content. In addition, the degradation rates of Samples 1 and 2 are different. The reason for this is because the tea residue contains a considerable amount of protein in addition to plant fibers, which is more easily decomposed by soil microorganisms than plant fibers.

It can be seen from Table 6 that biodegradation is accompanied by a decrease in mechanical strength. Studies have shown that under the action of microorganisms, PLA and tea waste are simultaneously degraded, and the shortening of tea fiber size leads to the deterioration of internal stability of the material and the deterioration of mechanical properties. In addition, the contact of tea waste with moisture causes expansion and creates a space inside the composite material, which on the

one hand increases the area of interaction between the material and the microorganism, and on the other hand destroys the original internal structure and also causes a decrease in mechanical strength.

Table6. Mechanical decline test results of PLA and tea waste composites.

	Sample 1	Sample 2	Sample 3
Internal bond strength (Mpa)	3.74	5.37	30.86
Decline (%)	86	81.5	40

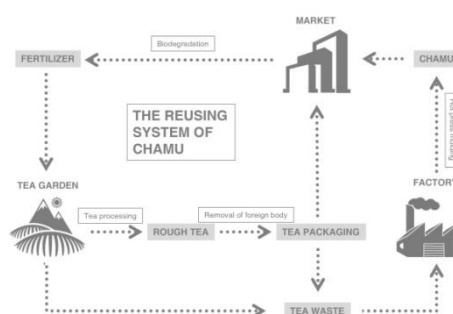


Figure 5. Tea waste recycling system.

4. Conclusion

This study demonstrates the possibility of using tea waste as a filler to prepare PLA and tea waste composites and has verified by soil-buried experiments that PLA and tea waste composites have good degradation properties in natural environments.

As shown in Figure 5, the author established a tea waste recycling system based on previous research results and characteristics of tea waste. Through the cooperation of farms, factories, and shopping malls, the “natural power (the microbes in nature)” will make the “CHAMU” products biodegradable, turning them into fertilizer to return to the land to solve the problem of tea waste disposal and improve the value of waste utilization.

In recent years, Japan has used biodegradable plastics as the fourth new material after metal materials, inorganic materials, and polymer materials. ITO EN, a well-known Japanese tea beverage manufacturer, reprocesses the tea residue produced during the production process and has now developed a variety of tea composite products for sale on the market. As a major tea production and consumption country, China produces a large amount of tea waste every year. By using these wastes, the development of new biodegradable tea waste products based on the “tea waste recycling system” will have broad market prospects.

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