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Research Article

Development of Starch-Based Bioplastic from Jackfruit Seed

Trieu Khoa Nguyen, Nguyen Thy Ton That, Nien Thi Nguyen, and Ha Thi Nguyen

Faculty of Mechanical Engineering, Industrial University of Ho Chi Minh City, Ho Chi Minh City 70000, Vietnam

Correspondence should be addressed to Trieu Khoa Nguyen; nguyenkhoatrieu@iuh.edu.vn

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In this article, jackfruit seed starch plasticized with common plasticizers was developed and characterized. At the first step, the research papers that dealt with the fabrication and characterization of starch-based bioplastics were synthesized and analyzed. Next, jackfruit seeds were selected as a source for starch because of their large availability, low price or even free, and high starch capacity. Afterward, a starch-based bioplastic fabrication procedure was proposed. From preliminary tests, plasticizers were sufficiently selected, including water, glycerol, natri bicarbonate, and acid citric. Using different combinations of these plasticizers, four types of bioplastics were then fabricated to study the effect of the plasticizers as well as to characterize the properties of the corresponding bioplastics. A cutting tool for ASTM D412 type A standard tensile testing specimen was then designed and fabricated. Using these dog-bone specimens, tensile results showed that the hardness of the fabricated bioplastic was positively proportional to the ratio of the starch. Furthermore, from SEM characterization, the bioplastic specimens were fully plasticized. Although the fabricated bioplastic has lower mechanical properties than petroleum-based plastics, its environmental friendliness and high potential added value promise to be a material of the future.

1. Introduction

There has been an increasing interest in the production of bioplastics as the environmental pollution from conventional plastics is increasingly evident [1–3]. The bioplastic industry is being considered in the world as the 4.0 revolution of green technology [1, 4]. Environmental pollution, especially pollution caused by plastic waste, is globally becoming more and more serious [5, 6]. Petroleum-derived plastics pollute the soil, water, and air. They could also destroy the habitats of many species, reduce biodiversity, cause many direct impacts on humans, and hence, is likely to result in the unsustainability of global development. Bioplastics, new environmentally friendly materials, are promised to be a great solution to replace traditional plastics, thereby contributing to reducing plastic waste pollution and protecting the living environment on earth.

Bioplastics can also be biobased (originated from renewable materials) and biodegradable (that can be back to nature). In terms of renewability and production, some of the most widely known biobased plastics nowadays are polylactic acid (PLA), starch-based plastics, protein-based plastics, cellulose esters, polyhydroxyalkanoates (PHAs) [1], and plant fibres [7, 8]. Bioplastics, manufactured from biomass, with or without modifications, such as starch, protein, and cellulose, are biodegradable and therefore gaining more and more attention. Among these, starch has shown potential for the fabrication of biodegradable materials as it is cheap, readily available, and renewable [2]. It should also be noted that, in addition to being used to produce bioplastics, biomass can also be used to produce biogas, biooil, biocoal [9], and bioethanol [10].

Bioplastics produce remarkably less greenhouse gas (carbon dioxide) emissions than traditional oil-based plastics over their lifetime [11]. Since the plants that biobased plastics are made from absorbed that same amount of greenhouse gas as they grew, there is no net increment in this gas. With such advantages, bioplastics have been studied a lot in the world, especially the production of bioplastics from starch. The authors in [12, 13] used rice starch and some plasticizers such as glycerol, sorbitol, and formamide to produce bioplastics. These authors chose rice starch because it contains a high amount of amylose, about 30%, which is suitable for coating materials. Furthermore, Taghizadeh and Favis [14] used wheat starch to make plastic. Meanwhile, Thunwall et al. from Sweden [15], Talja et al. from Finland [16], and Ozdamar and Ates from Turkey [17] utilized potato starch to produce bioplastic because of its popularity. In addition, Niu et al. [18] utilized response surface methodology (RSM) combined with the multi-index comprehensive evaluation method to prepare and characterize biodegradable composited films using potato starch. Another starch, corn starch, in addition to fermentation to make PLA, was utilized by many authors to make bioplastics directly [19–21], mainly for packaging. Similarly, there have been many other authors using other starch sources such as sweet potato [22, 23], tapioca [24, 25], avocado seeds [26–28], jackfruit seed [2, 29, 30], and durian seed [31, 32]. In addition, many authors also utilized many types of starch blends to achieve bioplastics with better properties. For example, Lopattananon et al. from Thailand [33] used a mixture of tapioca and rice starch to make bioplastics, while Yin et al. [34] utilized oxidized wood pulp fiber to incorporate into thermoplastic starch (TPS) to prepare new composites. And Marichelvam et al. from India [35] mixed corn and rice starch to result in shorter biodegradation time bioplastics. The common feature of these studies was that the fabrication of bioplastics is based exclusively on starch which requires the supplement of plasticizers. Because these bioplastics had low machinability, they were difficult to adapt to the conventional processes. In addition, whatever the botanical origin, starch-based bioplastics showed a strong hydrophilic character, which made them unsatisfactory for several applications [2].

Although bioplastics are commonly considered to be more environmentally friendly than petroleum-based plastics, that is not necessarily the case when considering the life cycle of the materials [11]. Bioplastic fabrication processes result in a large number of pollutants, because of the fertilizers and pesticides utilized to grow the plants and the chemical processing needed to change organic materials to plastics [11]. While the biodegradability of bioplastics is regarded as an advantage, most bioplastics require high-temperature industrial facilities to be broken down. Very few countries have the necessary infrastructure to deal with this composting procedure. Hence, bioplastics usually end up in landfills where, due to being deprived of oxygen, they may release methane, a greenhouse gas, 23 times more harmful than carbon dioxide. Bioplastics are also relatively costly. Due to the complex processes utilized to turn corn starch or sugarcane into building blocks, PLA plastic can be 20 to 50 percent more high-priced than comparable plastic materials [11].

Therefore, the research on manufacturing, improving the useful properties, and using bioplastics in the world in general and in Vietnam, in particular, is very necessary. In this study, in order to exploit agricultural wastes, the bioplastic was made from the starch of jackfruit seeds. This is so because jackfruit seeds are common in Vietnam and easy to gather in large quantities, especially from those factories producing dried jackfruit. In addition, these seeds are also normally sold at a negligible price or even free of charge. After a starch extracting process, water, glycerol, natri bicarbonate, and acid citric were applied as plasticizers. The environmental friendliness and high-potential added value of the fabricated biopolymer promise to be a material of the future.

2. Materials and Methods

2.1. Materials

2.1.1. Jackfruit Seed. Jackfruit seeds were obtained from a very popular variety, called Changai—originating from Thailand, in the South of Vietnam. Analysis of the main components of jackfruit seeds detected the presence of $30.08\% \pm 0.1$ amylose [2] or $26.4\% \pm 0.7$ amylose in [36]; $7.16\% \pm 0.04$ moisture; $1.17\% \pm 0.03$ total protein; $0.3\% \pm 0.01$ lipids; $0.21\% \pm 0.03$ ash; and $0.15\% \pm 0.02$ fibers [2]. In the present study, it was important to consider the amount of ash, amylose, and lipid found as these components influenced the formation of bioplastics. A low percentage of ash was indicated by a low concentration of minerals in the jackfruit seed. Due to possible interactions between minerals with amylose, amylopectin, and plasticizers, the high concentration of these compounds interfered with the formation of bioplastics [2]. The found lipid percentage in jackfruit seed starch was low ($0.3\% \pm 0.01$). High lipid percentage in starch might affect the color of the bioplastics. Furthermore, lipids could negatively affect the water absorption stage of starch granules, change the plasticizing temperature, and limit the amylose retrogradation, and therefore, the bioplastic becomes brittle. In previous studies, 30.08% amylose was found in jackfruit seed starch, which could be considered as high amylose content [2]. The amylose content is an essential property for the formation of bioplastics because it is responsible for gelatinization and retrogradation process. In other words, amylose is responsible for hydrogen bonding between the hydroxyl groups of biopolymers that form intermolecular junctions and lead to film production. The physical, chemical, and functional properties of the fabricated bioplastics depend on the ratio between amylose and amylopectin [2].

2.1.2. Glycerol. A classical starch plasticizer is perhaps the thermoplastic starch's most commonly investigated and utilized plasticizer. This is due to its low price, nontoxicity for human food and biomedical applications, and relatively high boiling point (292°C). Furthermore, the hydrolysis or conversion of lipids (triglycerides) to fatty acids for the biodiesel industry produces glycerol as a by-product [1]. This presents an opportunity to improve the economics of both the biodiesel industry and the bioplastics industry.

2.1.3. Acid Citric. The adhesion between glycerol, water, citric acid, and starch in thermoplastic starch is strengthened by the supplementation of citric acid [37, 38]. Citric acid can create stronger hydrogen bonding interactions with starch than glycerol. Hence, citric acid can effectually inhibit starch recrystallization (i.e., retrogradation), due to its strong hydrogen bonding interaction with starch. Rheological investigations demonstrated that citric acid can clearly reduce the shear viscosity and improve the fluidity of thermoplastic starch. Citric acid can also enhance the elongation of glycerol-plasticized thermoplastic starch and improve its water resistance at high relative humidity, but reduce the tensile stress [37]. Citric acid and glycerol are utilized to enhance the flexibility and fluidity of the material

Table 1: The ratio of the raw material composition of plastic specimens.

Ingredients	Starch : glycerol	Citric acid : (starch+glycerol)	Baking soda : (starch+glycerol)	Starch : water
Ratio	3.50 : 1 3.00 : 1 2.75 : 1 2.50 : 1	1 : 100	5 : 100	12.5 : 100

Table 2: The specific weight of each material.

Specimens	Ingredients				
	Starch (g)	Glycerol (g)	Citric acid (g)	Baking soda (g)	Water (g)
Ratio 3.5 : 1	20	5.71	0.25	1.28	160
Ratio 3.0 : 1	20	6.67	0.26	1.33	160
Ratio 2.75 : 1	20	7.27	0.27	1.36	160
Ratio 2.5 : 1	20	8.00	0.28	1.40	160

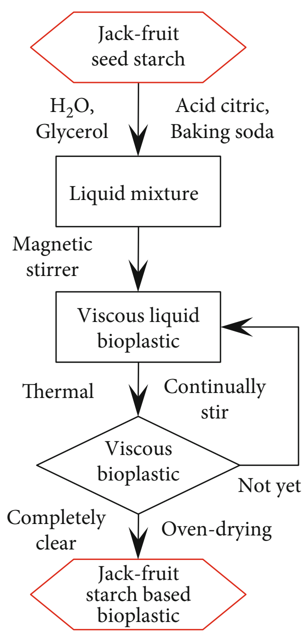


Figure 1: Flow diagram of bioplastic fabrication using jackfruit seed starch.

Figure 2: Flattened viscous bioplastic on the PE film coated tray.

to which they are supplemented. Furthermore, citric acid is a potential cross-linking agent under low-cost, nontoxic conditions.

2.1.4. Baking Soda. Baking soda, also known as sodium bicarbonate ($NaHCO_3$), scientifically, is one of the preservatives and plasticizer additives as the same as sodium metabisulfite ($Na_2S_2O_5$), glycerol, and sorbitol. These additions strengthen bioplastics and make them more durable [37].

2.2. Methods

2.2.1. Starch Extraction. First, starch was extracted to prepare raw materials for making bioplastics. The raw jackfruit seed has a very pungent odor and hard outer shell and then needs to be deseeded for both the hard shell and the brown spermoderm. The deseeded seeds were then washed. The rotten seeds were discarded while the good seeds were overnight soaked for latex removal. After that, the overnight soaked jackfruit seeds were washed using pure water. Clean jackfruit seeds were then ground to obtain a slurry. The slurry was filtered through a filter cloth to obtain a crude starch suspension while the sediment was discarded. The filtrate was precipitated overnight for settlement and latex and minerals removal. The supernatant was removed, and the crude starch was washed with distilled water. This step was reproduced three times [39], and the starch cake was then dried naturally or by an oven. When using the sun-drying method, starch could be discolored and became contaminated, forming a thin light brown layer. Instead, the starch suspension was dried at 45°C for 24 hours in an oven dryer. It should be added that the proposed starch extraction process is simple and easy to implement. It does not include a

(a) (b)

Figure 3: Drying stage. (a) Tray dryer with air circulation at a temperature of 55°C. (b) Dried bioplastic specimens.

(a) (b)

Figure 4: Characterization for mechanical properties. (a) Stamping process for dog-bone samples. (b) Dog-bone samples for tensile testing.

centrifugation step as in [2], soaking in 2% NaOH and slicing as in [29], or cutting with the size of 1 cm^2 approximately before crushing as in [30].

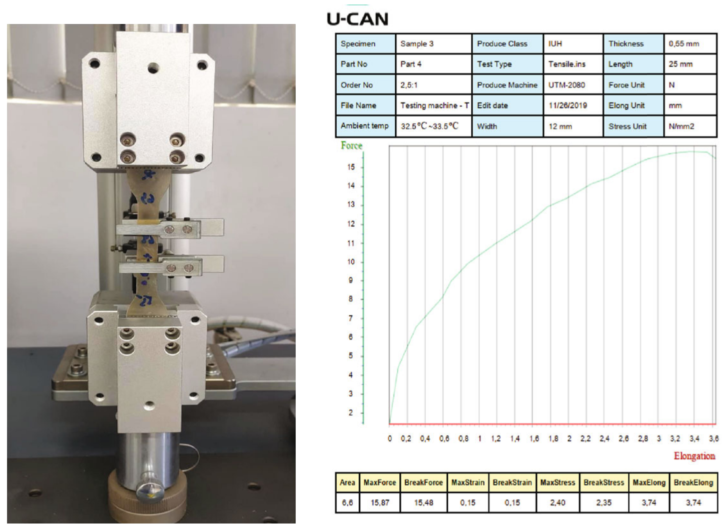
2.2.2. Bioplastic Fabrication. After extracting starch, bioplastic was fabricated. The proportions of ingredients were selected for the production of bioplastics as shown in Table 1. These ratios were selected similarly to [24] for easy comparison of results. From there, the specific weight of each material was calculated as shown in Table 2.

Bioplastics are prepared by mixing starch with glycerol with different starch : glycerol ratios by mass (2.5 : 1; 2.75 : 1; 3.0 : 1; and 3.5 : 1) [2, 23] and by as shown in the flowchart in Figure 1. The plasticization process was carried out by putting the liquid mixture with the ingredients in a given ratio above a heated magnetic stirrer (SCILOGEX model MS-H-S) at a temperature of 95°C. The mixture was stirred with a magnetic stir bar. When the temperature is higher, the process will go faster; however, when the temperature is too high, the mixture can be thermally decomposed before being completely converted into bioplastic. After the mixture turned completely clear and thick, stop stirring. The plastic mixture was poured into trays covered with PE film (Figure 2). Without PE film coating, the bioplastic would stick to the tray and be cracked when drying. After that, the viscous liquid bioplastic was flattened on a tray (Figure 2) and then transferred to an oven at 55°C (Figure 3(a)) and dried for 15 hours until the weight was constant. A natural air circulation MEMMERT UNB400 universal oven was utilized for this step. It is worth noting that the drying time can be shorter or longer depending on the thickness of the viscous bioplastic layer. The dried bioplastic specimens are shown in Figure 3(b).

From the diagram in Figure 1, it can be observed that the proposed procedure is simple and suitable for laboratories at universities. According to the literature review, the shaking step in the ultrasonic bath [2] was eliminated. In addition, since pure starch is used, an additive blending step is also unnecessary both before and after plasticization as in [29, 30], respectively.

3. Results and Discussion

3.1. Mechanical Properties. Bioplastic specimens were stamped into dog-bone samples according to ASTM D412-Type A standard (Figures 4(a) and 4(b)). In this study, to measure the mechanical properties of bioplastic specimens, the U-CAN DYNATEX INC TYPE UT-2080 tensile tester (Figure 5(a))



(a) (b)

Figure 5: Tensile strength testing stage. (a) Tensile strength tester. (b) A typical result of the tensile test of the bioplastic sample.

Table 3: Results of mechanical properties of bioplastics.

Results	Types			
	Ratio 2.5 : 1	Ratio 2.75 : 1	Ratio 3.0 : 1	Ratio 3.5 : 1
Max stress σ (MPa)	3.39	3.90	4.08	5.15
Strain ε	0.29	0.23	0.31	0.25
Young modulus E (MPa)	12.70	16.96	13.53	20.06
Elongation A (%)	29.13	22.60	30.67	25.20
Density (g/cm^3)	1.375	1.385	1.403	1.410

was used. The density of the samples was determined by an electronic densimeter MDS-300. Three dog-bone samples were used for the mechanical characterization of each type of bioplastic. A typical result of the tensile test of the bioplastic sample was shown in Figure 5(b).

The initial observation showed that the higher the glycerol ratio was, the softer and more flexible the bioplastic became, and vice versa. Increasing the percentage of citric acid enhanced the hardness and reduced the plasticity of the bioplastic. At the same time, if the flattened viscous biopolymer layer was made too thick, it would be cracked due to shrinkage.

Based on the measured results (Table 3) and the plot (Figure 6), in general, the mechanical properties of bioplastics increase gradually with the starch : glycerol ratio. However, in the sample with a ratio of 3 : 1, there was a difference. The reason might be that the air bubbles inside the bioplastic were not removed completely. This led to nonuniform mechanical measurement samples in terms of material distribution (more or less air bubbles inside), affecting the measurement results. In addition, another very important cause was the uneven thickness of the specimens due to the manual leveling of the viscous bioplastic in the tray before drying. The tensile fracture position might have a smaller thickness than the clamping position. Bioplastics with a ratio of 3.5 : 1 had the highest strength in this study but were still low compared to traditional petroleum-based plastics. Therefore, bioplastics from jackfruit seeds cannot replace traditional plastics in many fields of application that require high mechanical properties yet, as other common bioplastics [40]. However, compared with other bioplastics that have been studied, the results are encouraging. Also making bioplastics from jackfruit seed starch, the authors in [2] obtained the highest tensile strength of 3.12 MPa with 30% glycerol. As for the authors from Indonesia [23], they made bioplastic from sweet potato starch; a maximum tensile strength of 2.57 MPa with a starch/glycerol ratio of 3.5/1 could be obtained. In addition, the authors in [41] reported that the tensile strength of bioplastics made from potato starch reached 4.87 MPa, from tapioca starch reached 4.5 MPa, and from corn starch 3.59 MPa.

3.2. Scanning Electron Microscope (SEM) Characterization. The SEM characterization was performed using Hitachi scanning electron microscope S-4800. Figure 7(a) showed the morphology of jackfruit seed starch in this investigation. The jackfruit seed starch granules are varied round to bell shapes with smooth surfaces. Previous studies revealed that the granular size lied between 5 and 10 μm [42]. In our case, the average granular size is 8.2 μm as shown in Figure 7(a). Because of its small size, jackfruit seed starch granular can be a great source for encapsulated products [43]. Morphology and structure were firstly analyzed only for bioplastic specimens with a ratio of 2.5 : 1 (Figure 7(b)) and a ratio of 3.5 : 1 (Figure 7(d)). SEM photographs show that the surfaces exposed to air are rough. The micrographs do not illustrate any intact starch granules, which implies that the jackfruit starch was fully gelatinized during the forming of the film specimens. In addition, analyses of the bioplastic surfaces reveal that the specimen with a ratio of 3.5 : 1 has some microcrack on its surface. These microcracks occurred on the tougher specimen with a lower amount of glycerol.

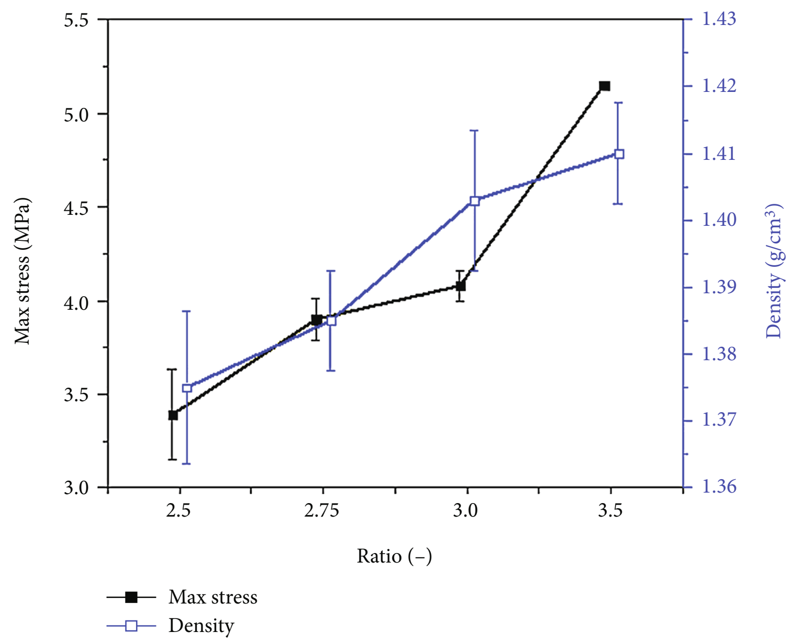


Figure 6: The plot of max stress and density for each type of bioplastic.

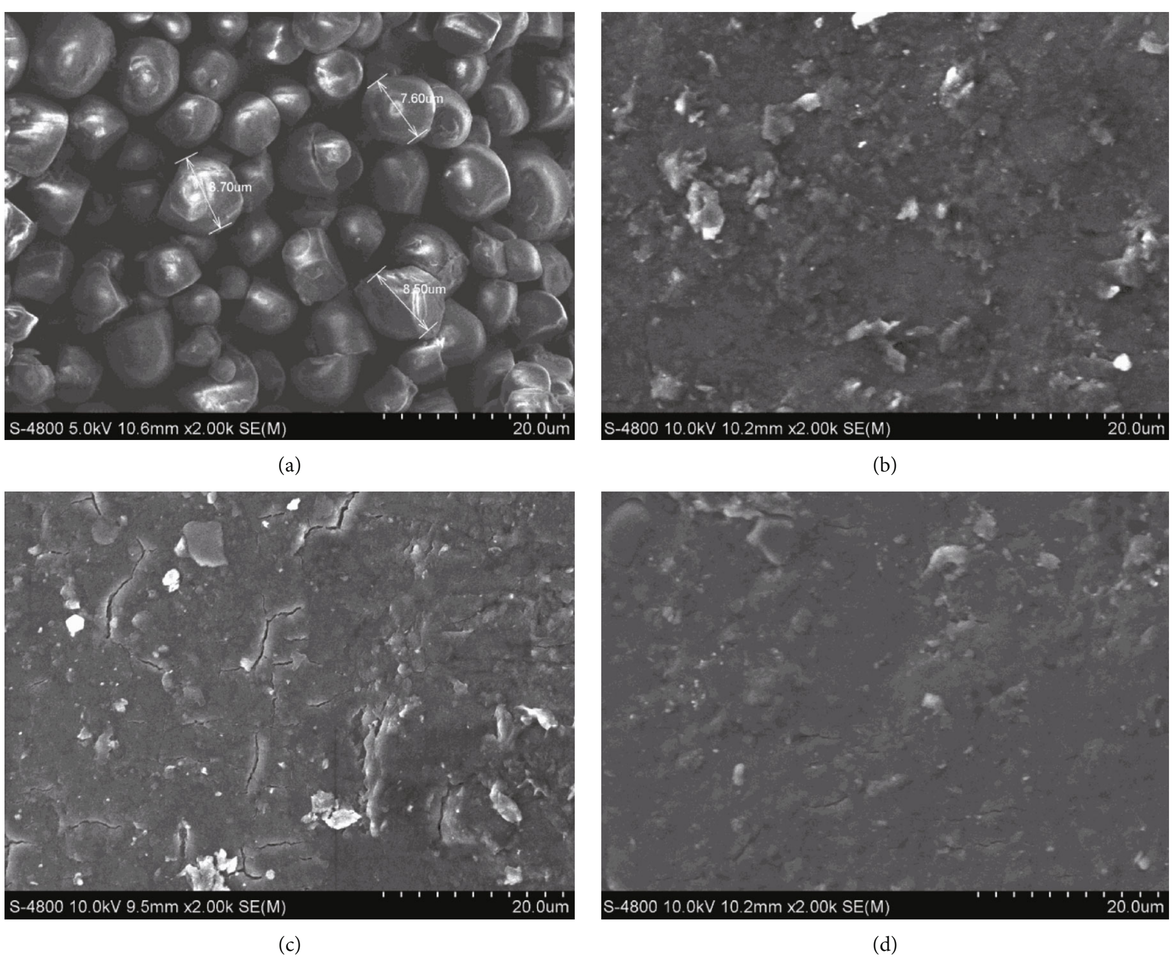


Figure 7: SEM characterization with 2000 times magnification. (a) Starch from jackfruit seed. (b) Bioplastic with ratio 2.5 : 1. (c) Bioplastic with ratio 3 : 1. (d) Bioplastic with ratio 3.5 : 1.

After that, an additional SEM analysis was done for the specimen with a ratio of 3 : 1 due to its special mechanical results. The SEM image in Figure 7(c) is in agreement with others on a full polymerization process. However, there were many microcracks on the surface of the specimen. The occurrence of these microcracks may be due to the presence of microbubbles formed during the gelatinization process [2]. These microbubbles were caused by hydrogen bonding chains of the starch, which began to break down when reaching the gelatinization temperature and the water molecules started to infiltrate into hydroxyl groups in the starch molecule [25]. Unfortunately, in this bioplastic production process, due to the lack of a vacuum step, the number of microbubbles in the specimens was uncontrollable. In addition, the microair bubbles generated by mixing and stirring could not be fully eliminated.

4. Conclusions

A review of the literature revealed that plastic pollution is becoming a burning issue worldwide. To overcome that problem, many measures have been studied. This paper presents a process for making an environmentally friendly bioplastic from jackfruit seed starch. Jackfruit seed has a high starch content (about 30% of fresh seed weight) and is easy to collect in large quantities and extract starch. Therefore, it is suitable as a raw material source for bioplastics and at the same time takes advantage of the source of fruit seeds that can be considered waste products, increasing the value of crops. When making the bioplastic, the strength was generally proportional to the ratio of starch : glycerol. From SEM characterization, bioplastic specimens were fully plasticized. However, an excessive amount of starch would cause incomplete plasticization, while a too low ratio would make bioplastics unable to harden after drying. Bioplastic with the ratio of 3.5 : 1 has the best tensile strength, 5.12 MPa according to our measurements. However, the mechanical properties of the bioplastics could be greater because the specimens still had microair bubbles inside; at the same time, the thickness was not uniform, affecting the measurement results.

From the encouraging results of this study, and the great potential of bioplastics, the next research topic is to build a jackfruit seed peeling machine and a centrifuge to facilitate the starch extraction process. In addition, a vacuum desiccator and vacuum pump will be used to remove microair bubbles before drying, to achieve more accurate measurement results. The time and decomposition of the bioplastic under the normal using conditions and in the natural environment also need to be studied precisely. After that, the plasticizing temperature will be studied to develop a lab-scale injection molding machine for the bioplastics. Products from bioplastics will be then researched and developed. In addition, the simple processes of starch extraction and bioplastic fabrication also promise that this proposed method can be applied to industrial production in the near future.

Data Availability

No data were used to support this study.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

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