

On the Mechanical, Thermal and Biodegradation of Jackfruit Seed Starch Bioplastic

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Abstract: *In this work, we have prepared jackfruit seed starch-based bioplastic with glycerol as plasticizer. Since it has been reported that the amount of plasticizer can affect the properties of a bioplastic, we have tried to optimize the jackfruit seed starch: plasticizer ratio for the preparation of bioplastics. The mechanical properties, thermal stability, and the biodegradability of bioplastic in air, soil, water and bacterial medium were investigated. It was observed that the strength of the bioplastic decreases with increasing amounts of plasticizer. We have prepared degradable bioplastic with reasonable shelf life which can find applications in the field of disposable carry bags and garbage bags.*

Keywords: jackfruit seed starch, bioplastics, shelf-life, mechanical properties, degradation studies

I. INTRODUCTION

The extensive use of petroleum-based plastic and dumping of the same in soil and water body greatly affects our environment and biodiversity [1-2]. Petroleum-based polymers are the biggest threat to the environment as they are non-degradable [3]. However, bioplastics can reduce the volume of waste and thus, could be a promising alternative to the petroleum-based non-degradable polymer to address environmental issues [4].

Among natural polymer resources, starch is the most appealing candidate in bioplastic making because of its low cost, pervasive availability and renewability [5-6]. The amylose content in starch is an important characteristic for bioplastics production as it is responsible for gelatinization and retrogradation, which are required during film formation [7-9]. Jackfruit seed, potato, corn, rice, banana and cassava contains high amount of starch and these can be used as raw materials for bioplastic making [5-6].

Artocarpus heterophyllus Lam., which is commonly known as jackfruit is a tropical climacteric fruit, belonging to the family Moraceae, is native to Western Ghats of India. Jackfruit seeds comprise 70% of starch content [10]. The recovery yield of starch extracted from jackfruit seeds is about 81-82 % [11], which implies its possibility of being used as a potent source of starch in food and pharmaceutical industries, as a stabilizer, thickener, and as binding agent. Jackfruit seeds starch consists of two macromolecules: amylose (20–30%) and amylopectin (70–80%), which are associated with each other by hydrogen bonds. The amylose content in it is an essential property for the formation of biofilms. Menzel et al. found that the increased amylose content and structural changes in amylopectin enhances film-forming behavior and improves barrier and tensile properties in starch films [12].

Jackfruit seed starch has high amylose content, high gelatinization temperature (75–85 °C), thermomechanical shear tolerance, low lipid content, acidity resistance, higher temperature and pulp viscosity, and lower pulp breakage than the gels produced by cassava and potato starches [13].

However, 8 to 15% of jackfruit seeds are primarily discarded after eating. Since they are readily available and there is little information in the literature about the synthesis of bioplastic from this source, we employed jackfruit seed starch in this work to create bioplastic films. As a result, it can be applied to future value-added product development.

It has been reported that the amount of plasticizer can affect the properties of the bioplastics [10]. The plasticizers used are glycerol, water, sorbitol [10, 14-15] etc.

The commercial application of such bioplastics is limited. We need bioplastics which has enough strength, more shelf-life i.e., more long-lasting, yet biodegradable so that it causes no harm to the environment. Only then useful products like carry bags, sheets etc can be prepared from bioplastics and can be used as alternatives for conventional plastics. To achieve this, we have tried to optimize the jackfruit seed starch: plasticizer ratio during the preparation. The mechanical properties, thermal stability, and the biodegradability of bioplastic in air, soil, water and bacterial medium were investigated.

II. EXPERIMENTAL

2.1 Materials

Jackfruit seeds were collected locally from different parts of Kochi, mainly Mulanthuruthy, Vyppin and Palluruthy. Commercial vinegar and glycerol were purchased from Nice Chemicals (P) Ltd, Kochi, Kerala.

2.2 Methods

Preparation of jackfruit seed powder:

The raw jackfruit seeds were collected and cleaned. The outer white coating was removed. The cleaned seeds were smashed, dried and ground to fine powders. The starch powder was then sieved with a 100-mesh sieve.

Preparation of bioplastics:

Jackfruit seed powder, glycerol, vinegar and distilled water were taken in calculated amounts as indicated in table 1 and mixed well. The mixture was stirred well until no lumps remained. At this stage, the mixture had a light brown colour and was quite watery. The mixture was heated to boiling on medium to low temperature with continuous stirring. As the temperature of the mixture increased, it became more translucent and began to thicken. The mixture was removed from heat when it became clear and thick. Care was taken to avoid the mixture getting over-heated so that lump formation can be avoided. Then the mixture was spread on a ceramic tile and was left for two days to allow it to cool and dry. The thin bioplastic sheet obtained was peeled off from the tile. Fig. 1 represents various stages of preparation of the bioplastics.

Table 1: The weight ratios of the components of the prepared samples

Sample name	Ratio	Starch powder	Glycerol	Vinegar	Distilled water
Sample 1	1: 0.1	15	1.5	15	150
Sample 2	1: 0.25	15	3.75	15	150
Sample 3	1: 0.5	15	7.5	15	150
Sample 4	1: 1	15	25	15	150

*All weights in grams

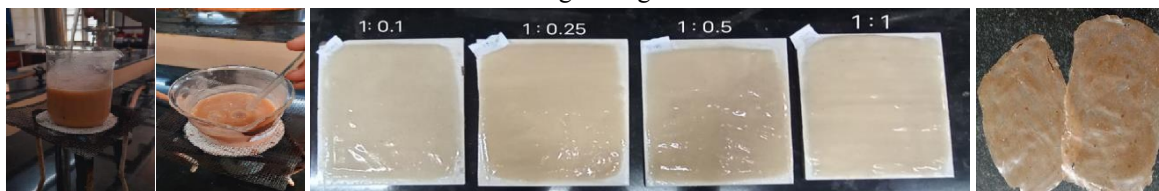


Fig.1 Various stages of preparation of the bioplastic

Mechanical Strength testing:

Four samples were cut in I-shape of dimensions 8 cm length and 2 cm width and tested using Shimadzu Autograph Series Universal Testing Machine (AGS-X5kN). Out of these four samples, one with the highest tensile strength was used for biodegradation studies.

Thermogravimetric analysis:

Thermogravimetric analysis (TGA) of the prepared samples was performed using TGA instrument, Hitachi-STA 7300 at a temperature range of 0-1300 °C.

Biodegradation Studies

To test whether the prepared bioplastics can degrade in the natural environment, we carried out the following biodegradation studies.

- Soil degradation
- Air degradation
- Water degradation
- Bacterial degradation

Soil degradation

Weight loss measurement is a method to study the biodegradation of a polymer. A small piece of bioplastic of length and width 5 cm was cut off from the sample and weighed, then it was buried in the soil at a depth of 5 cm. The sample was taken out from the soil every three days and the weight was measured. This process was repeated for 22 days. Fig. 2 gives the pictures of the bioplastic taken before and after soil degradation. It took about 7 days for the degradation to begin. The final weight of the sample was measured. The amount of biodegradation was calculated by

$$\text{Percentage degradation} = \frac{\text{Initial weight} - \text{Final weight}}{\text{Initial weight}} \times 100$$

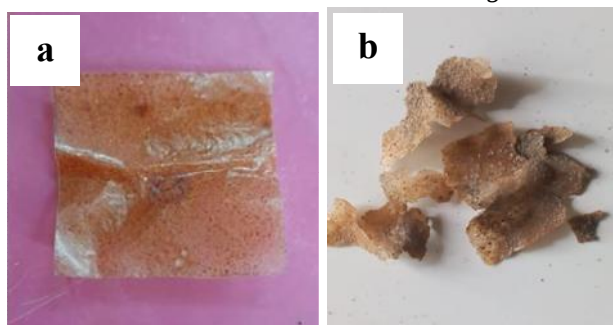


Fig.2 Bioplastic a) before b) after soil degradation

Air degradation

A small piece of bioplastic of length and width of 5 cm was cut off from the sample, weighed and was kept in air at room temperature. The weight was measured every 3 days for 22 days. Appearance of fungus on the sample was also noted. Fig. 3 shows the picture of the bioplastic before and after air degradation. And the percentage degradation was calculated using the above equation.

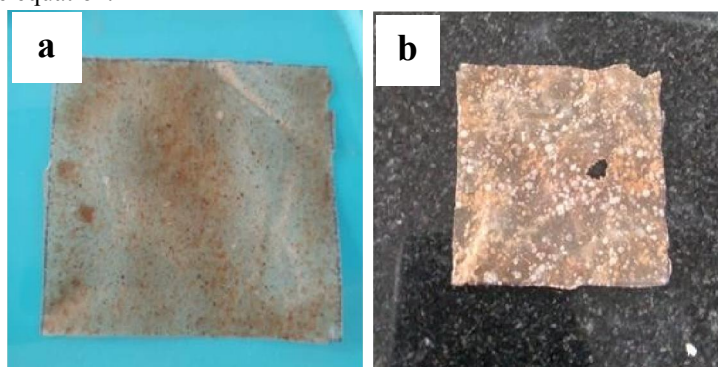


Fig. 3 Bioplastic a) before b) after air degradation

Water degradation

Previously weighed piece of bioplastic of length and breadth 5 cm from sample was dipped in water. The degradation of bioplastic in water was studied by noting the changes in the sample. Fig. 4 gives the pictures of the bioplastic before and after water degradation. The changes were noted every three days and the percentage degradation was determined.

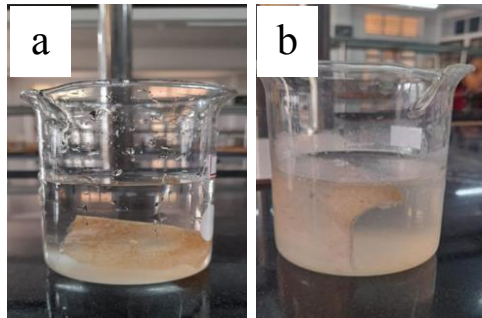


Fig. 4 Bioplastic a) before and b) after water degradation

Bacterial degradation

We carried out bacterial degradation studies of the prepared bioplastic film using different types of bacteria. The bacterial degradation of the sample was studied the following seven different types of bacteria: *E. coli*, *Mycobacterium smegma*, *Enterococcus*, *Staphylococcus aureus*, *Klebsiella*, *Staphylococcus iniae*, *Vibrio parahaemolyticus*. We cultured the bacteria in different test tubes. A small piece of bioplastic of length and width 1cm was cut off from the sample and weighed and put into each test tube. The changes were noted every three days and the final weight was measured on 7th day. Percentage degradation was calculated. Fig. 5 depicts the pictures of bacterial degradation of bioplastics.

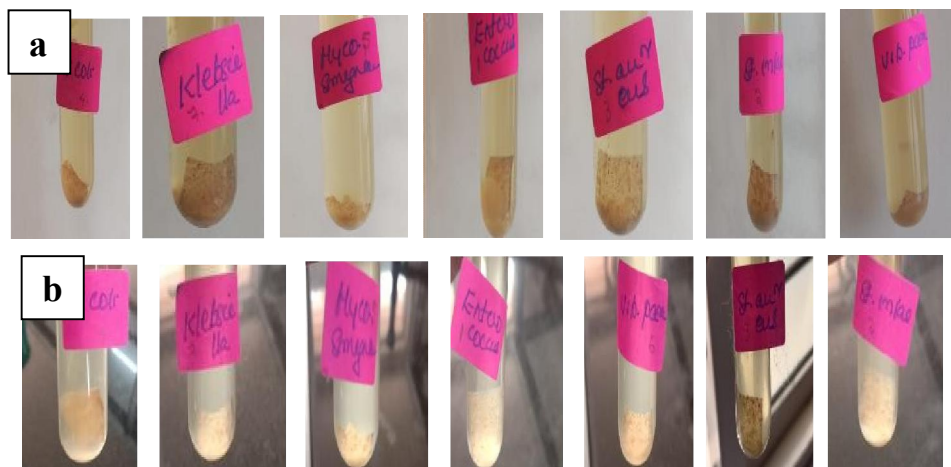


Fig. 5 Bioplastic before bacterial degradation on a) day 1 b) day 22

III. RESULTS AND DISCUSSIONS

3.1 Mechanical Properties

Mechanical properties of the bioplastic may be considered as the most important of all its other physical properties in view of commercial applications. The tensile strength of the bioplastic was tested as described in methods section. Tensile strength is the amount of load or stress that can be handled by a material before it stretches and breaks. Tensile strength of a material can be affected by several factors including the relative comparison between the matrix and the reinforcement materials in composite material. The graphical representation of the tensile strength of the samples with varying amounts of plasticizer (glycerol) is shown in fig: 6.

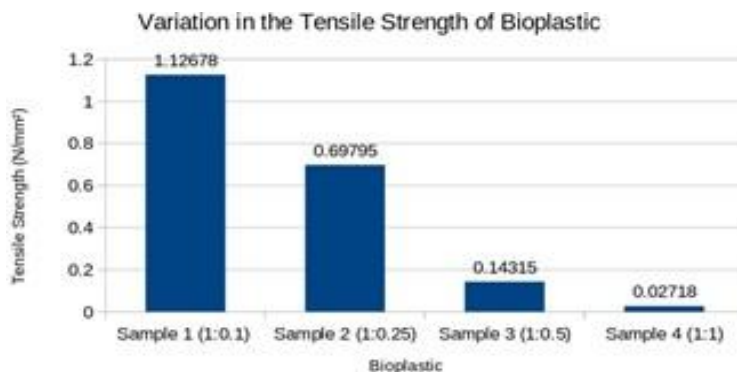


Fig: 6 Tensile strengths of the prepared bioplastics samples

From fig. 6, it is clear that as the amount of glycerol increases, the tensile strength of the bioplastic decreases. This is in accordance with previously reported studies [15]. Fig. 6 shows that sample 1 (1:0.1 ratio of Jackfruit seed starch and glycerol) has highest tensile strength of 1.12698 N/m². This sample i.e., Sample 1 was used for further studies.

Thermogravimetric Analysis

Thermal stability of the bioplastic sample (sample 1) was studied using thermogravimetric analysis. Thermal stability of bioplastic is defined as the ability of the material to resist the action of heat and to maintain its properties such as strength, toughness, elasticity at given temperature. So, the thermal stability of the sample helps to find out the temperature at which degradation of the sample take place. DTG graph gives clear idea about the decomposition of bioplastic at specific temperature with the help of peaks during DTG analysis. TGA graph of sample 1 (1:0.1) is shown in the fig. 7 and the thermal degradation characteristics are presented in table 2.

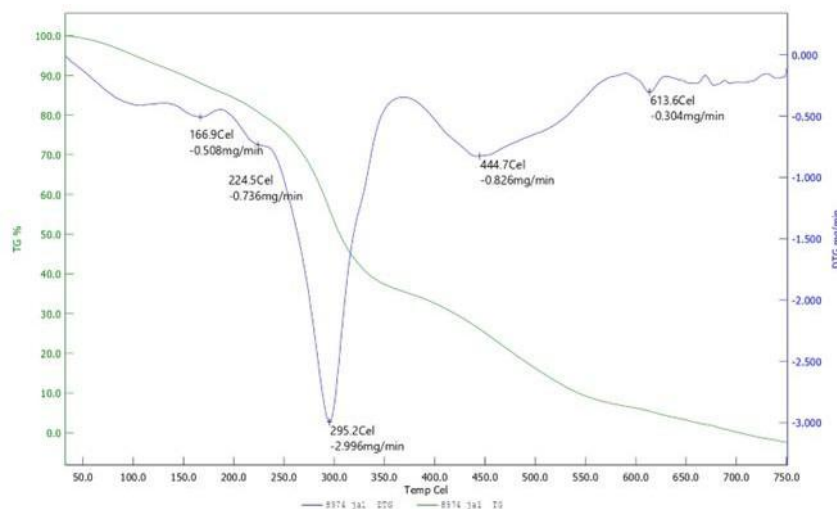


Fig. 7 TGA of sample 1

The DTG graph shows five degradations. Around 100°C, there is a small weight loss due to the loss of moisture. The second degradation maximum occurs at 166.9°C which is the onset temperature of degradation. Thus, the sample is thermally stable up to 166.9 °C. The third degradation is the one where maximum degradation occurs. The peak degradation temperature is 295.2 °C. The fourth degradation maximum occurs at 444.7°C and the last occurs at 613°C. 50 % weight loss occurs around 300 °C. Thus, the sample can be used for commercial applications. Yet, it can undergo thermal degradation easily.

Table 2: Thermal degradation characteristics of sample 1

Feature	Value
Onset temperature (°C)	166.9
Peak degradation temperature(°C)	295.2

Weight remaining at Peak degradation temperature (%)	~50
Temperature at 50 % weight loss (°C)	300

Biodegradation Studies

Soil degradation:

On the 22nd day, the percentage degradation was 38.73. This means that the prepared bioplastic will degrade slowly in soil.

Air degradation

The sample 1 was kept in open for 22 days. On the 15th day, fungal attack was observed. Hence the sample had a shelf-life of 15 days. The percentage degradation on the 22nd day was only 16.07. This means that the prepared bioplastic can be used without much degradation until 15 days.

Water degradation:

Biodegradation of any substance occurs in the presence of water and bacteria. The sample exhibited a gradual dissolution in water as expected due to degradation of the sample. The percentage degradation on 22nd day was 73.22. Hence the prepared bioplastic will easily degrade in water.

Bacterial degradation:

The degradation of sample 1 in presence of 7 different types of bacteria was monitored. The percentage degradation of the sample in different bacteria is listed in table 3. All bacteria were successful in degrading the sample. The maximum degradation of 99.25 % occurred in *Vibrio parahaemolyticus*. The least degradation was observed in *Enterococcus* with 52.98 % degradation.

Table 4: Percentage degradation of sample 1 in different types of bacteria

Bacteria	Percentage degradation
<i>E coli</i>	91.04
<i>Mycobacterium smegma</i>	88.05
<i>Enterococcus</i>	52.98
<i>St. aureus</i>	82.83
<i>Klebsiella</i>	96.26
<i>St. iniae</i>	88.80
<i>Vibrio parahaemolyticus</i>	99.25

From the above observations, it is clear that the bioplastic is completely biodegradable in all soil, air, water and in bacteria.

IV. CONCLUSION

Different samples of bioplastics were prepared from jackfruit seed starch with varying amounts of glycerol as plasticizer. The tensile strength of the prepared samples were measured. It was observed that as the amount of glycerol increases, the tensile strength of the bioplastic decreases. The sample with starch powder: glycerol ratio 1:0.1 had the highest tensile strength of 1.12678 N/mm². Further studies were done with this sample.

Thermogravimetric analysis demonstrated that this sample was stable up to 166.9 °C. The peak degradation temperature was 295.2°C. 50 % weight loss occurs around 300 °C. Thus, the sample can be used for commercial applications. Yet, it can undergo thermal degradation easily.

Percentage degradation of bioplastic in soil, water and air was found to be 38.73%, 73.22% and 16.07 %, respectively on 22nd day. Hence the bioplastic is completely biodegradable in all of the above conditions. In air, fungal attack appeared on 15th day. Hence the shelf life of the bioplastic is 15 days, which is reasonable for commercial applications like preparation of disposable carry bags or garbage bags.

Bacterial degradation studies show that bioplastic can be broken down by practically all types of bacteria within 7 days. The maximum degradation of 99.25 % occurred in *Vibrio parahaemolyticus*. The least degradation was observed in *Enterococcus* with 52.98 % degradation.

Thus, jack fruit seed powder could be an effective substrate for the production of starch-based bioplastics and it can be a suitable replacement for petroleum-based plastics for commercial applications like making of disposable carry bags or garbage bags.

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