Utilization starch of jackfruit seed (Artocarpus heterophyllus) as raw material for bioplastics manufacturing using sorbitol as plasticizer and chitosan as filler

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Abstract. Starch is a natural polymer that can be used for the production of bioplastics because its source is abundant, renewable and easily degraded. Jackfruit seeds can be used as raw material for bioplastics because it contains starch. The aim of this study to determine the characteristics of jackfruit seeds and determine the effect of chitosan and sorbitol on the physicochemical properties of bioplastics from jackfruit seeds. Starch is extracted from jackfruit seeds were then characterized to determine its chemical composition. In the manufacture of bioplastics starch composition jackfruit seeds - chitosan used was 7: 3, 8: 2 and 9: 1 (g/g), while the concentration of sorbitol used was 20%, 25%, 30%, 35%, and 40% by weight dry ingredients. From the analysis of jackfruit seed starch obtained water content of 6.04%, ash content of 1.08%, the starch content of 70.22%, 16.39% amylose content, amylopectin content of 53.83%, 4.68% protein content, fat content 0.54%. The best conditions of starch bioplastics jackfruit seeds obtained at a ratio of starch: chitosan (w/w) = 8: 2 and the concentration of plasticizer sorbitol 25% with tensile strength 13.524 MPa. From the results of FT-IR analysis indicated an increase for the OH group and the group NH on bioplastics due to the addition of chitosan and sorbitol. The results of mechanical tests is further supported by analysis of scanning electron microscopy (SEM) showing jackfruit seed starch has a small granule size with the size of 7.6 μm and in bioplastics with chitosan filler and plasticizer sorbitol their fracture surface is smooth and slightly hollow compared bioplastics without fillers chitosan and plasticizer sorbitol.

1. Introduction
Plastics as we know are used as packaging materials for various products including food. Plastic chosen as the packaging material because it is safe, robust (waterproof, light, and heat) and the price is cheap [1]. However, synthetic plastic are derived from non-renewable resources that can caused environmental damage. Most plastics are made from petroleum. Therefore, the plastic is not considered environmentally friendly because it is not biologically degraded lands and will certainly contaminate the soil [2]. Due to its shortcomings, it has been necessary to develop an ecological solution to overcome these problems, one of which is to develop biodegradable plastics (bioplastics). This means that these plastics can be decomposed by microorganisms naturally. Development of
biodegradable plastic materials using renewable natural materials (renewable resources) is highly expected [3].

One of the main component of bioplastics is starch. Starch is widely used in the form of biodegradable films in varied applications because it is a renewable, abundant and inexpensive material [4]. In a jackfruit contained approximately 100 to 500 jackfruit seeds or about 8-15% by weight of jackfruit itself with high starch content [5]. The addition of plasticizer serves to improve the flexibility bioplastic. Sorbitol is a plasticizer which is good enough to reduce internal hydrogen bonds that will improve the intermolecular distance [6]. Chitosan is used as a filler in the manufacture of bioplastics. One way to reduce the hydrophilic nature (less resistant to water) bioplastics is by mixing starch with biopolymers hydrophobic (water resistant) such as cellulose and chitosan [3].

The aim of this paper is to report a study of the utilization of jackfruit seed (Artocarpus Heterophyllus) as raw material for bioplastics manufacturing using sorbitol as Plasticizer and chitosan as filler which can be overcome environmental problems due to use of syntetic plastics.

2. The Material and Method

2.1. Materials
Jackfruit seed was obtained from fruit merchants located at Traditional Market Tanjung Rejo, Medan, Indonesia. Sorbitol, aquadest, and acetic acid were obtained from Laboratory of Chemical Process Industries, Chemical Engineering, University of Sumatera Utara.

2.2. Starch Extraction of Jackfruit Seed Starch
Jackfruit seeds (100 gr) were peeled and washed witch clean water. Seeds were cut with a size of approximately 1 cm$^2$ and then blended with 100 ml water. Starch slurry was filtered using a plastic sieve to obtain liquid filtrate (starch suspension). The resulting suspension were then deposited for 24-48 hours until the starch settles perfectly. Starch sediment was filtered using Whatman filter paper no. 1 to obtain a wet starch. Starch was dried using oven on temperature of 70$^\circ$ C for 30 minutes. Then starch was sieved with strainer 100 mesh.

2.3. Film Preparation
Number of starch and Chitosan mass wanted was 10 grams. Starch solution made with a ratio of starch : distilled water is 1 : 20 as much as 100 grams in 500 ml beaker glass. Then chitosan solution made in accordance with the volume that has been calculated on a beaker glass. Then the starch solution was heated while stirred with velocity of 400 rpm and temperature of 88,82$^\circ$ C for 25 minutes using hotplate. After 25 minutes, sorbitol was added with variation 20%, 25%, 30%, 35% dan 40% on starch solution. After stirred for 15 minutes, the solution was cooled down and cast onto flat and dried with temperature 60$^\circ$ C for 24 hours. After bioplastic dried, it was removed from the flat and saved in the desicater and ready to be analyzed.

2.4. Characterization of Starch
The procedure of water content and ash content of jackfruit seed starch is based on AOAC standard. Analysis of starch, amylose, amylopectin, fat and protein was observed in Jasa Uji Ilmu, Fakultas Teknologi Industri Pertanian, Universitas Padjajaran.

2.5. Characterization of Bioplastic
FTIR analysis of bioplastic was observed in Laboratorium Penelitian, Fakultas Farmasi, University of Sumatera Utara. Tensile strength test is based on ASTM standard D638. SEM analysis was observed in Laboratorium Fisika, Universitas Negeri Medan.

3. Results and Discussion

3.1. Characterization of jackfruit seed starch
Based on sedimentation method [7] from 100 grams of jackfruit seed could produce jackfruit seed starch 26.67 grams or percentage of starch was 26.67%.

Characteristics of jackfruit seed starch was aimed to know the percentage of each component contained in the starch produced, include starch (amylum), water, ash, fat, protein, amylose and amylopectin content so that known the quality of starch produced [8]. The result of jackfruit seed starch was provided inside the Table 1.

**Table 1. Results of jackfruit seed starch**

| Components of Starch | Percentage (%) | Standard of Indonesia Industry (%) [9] |
|----------------------|----------------|----------------------------------------|
| Starch (amylum)      | 70.22          | Min 75                                 |
| Amylopectin          | 16.39          | -                                      |
| Amylose              | 58.83          | -                                      |
| Water                | 6.04           | Max 14                                 |
| Ash                  | 1.08           | Max 1,5                                |
| Fat                  | 0.54           | -                                      |
| Protein              | 4.68           | -                                      |

3.2. Fourier-transform infrared spectrometry

The FTIR result of starch, chitosan, bioplastic without adding filler and plasticizer was provided in figure 2 below.

![Figure 2. The result of Fourier Transform Infra Red (FTIR)](image)

From the analyzing result of FTIR can be seen that jackfruit seed starch which owns O-H Alcohol (H-bonded), C-H alkanes (stretch) and C-H aldehydes, C-O amide, S=O sulfates, sulfonamides, sulfones and C-O ester. The presence of OH group binded with hydrogen, stretching of C-H alkane, stretching of C=O amide and stretching of C-O ether had represented the content of jackfruit seed starch which is consisted of amylose and amylopectin and reducing glucose (C\(_{6}\)H\(_{10}\)O\(_{5}\))\(_{n}\) [10].

From the analyzing result of FTIR chitosan which owns the hydrogen bonding groups O-H, C-H alkane (stretch), N-H amide, C=O amide, C-N amine, ether groups and C-O ester. Chitosan has a hydroxyl group (OH) which is negatively charged and the amine group (NH\(_{2}\)) positively charged, so that chitosan is able to form a strong ionic bonds [11]. The existing of those groups had shown the characteristic of chitosan which is characterized by the presence of amino group at wave number of absorption peak 1315.45 cm\(^{-1}\) and 1145.72 cm\(^{-1}\), and the presence of hydroxil group at wave number 3433.29 cm\(^{-1}\).
From the analyzing result of FTIR bioplastic without adding chitosan and sorbitol can be seen the presence of alcohol group O-H, alkanes group, aldehydes, C-H alkenes, amide C=O, aromatic group C=C and C-O ester.

From figure 2 also can be seen the FTIR result of bioplastic with adding chitosan as filler and sorbitol as plasticizer showed the groups of O-H alcohol, alkenes, aldehydes, C-H alkenes, C=O aromatic, the primer and secondary groups of amine, C=C aromatic and ester C-O. The differences result between bioplastic without adding filler chitosan and plasticizer sorbitol and bioplastic with adding filler chitosan and plasticizer sorbitol could be seen from the emergence of the N-H amino group of a functional group which is owned by chitosan with wave number 1593,20 cm$^{-1}$.

From the result, can be seen that bioplastic with adding chitosan as filler and sorbitol as plasticizer had increasing wave number of OH group in which the increase of O-H group is due to the increasing of hydrogen bonds formed on bioplastics. The hydrogen bonds consist of bonds between chains of amylose-amylose, amylose-amyllopectin, chitosan-chitosan, and amylose-chitosan-amyllopectin [8].

3.3. Tensile strength of Bioplastic
The following graphs shows the effect of chitosan and sorbitol addition on tensile strength bioplastic.

![Figure 3](image)

**Figure 3.** The effect of chitosan and sorbitol addition on tensile strength bioplastic from jackfruit seed starch

Figure 3 shows bioplastic with chitosan content 2 grams and 25% sorbitol provided the maximum tensile strength for 13.52 MPa. While, bioplastic with chitosan content 2 grams and 40% sorbitol provided the minimum tensile strength for 3.82%.

From figure 3 can be seen that with increasing mass of chitosan cause the increasing value of tensile strength. The addition of chitosan affects the chemical bonds constituent of bioplastic thus improved its tensile strength. With increasing concentrations of chitosan, there will be more hydrogen bonds contained in the bioplastic so that the chemical bonds of bioplastics will be stronger and difficult to break up, because it requires a large energy to break the bond. This is caused by the physics changes of bioplastic particles, so that plastics are increasingly homogeneous and has a dense structure [12].

A drop tensile strength value is shown for bioplastic with chitosan content 3 grams and concentration of sorbitol 25%. Buzarovska et al (2008) reported that a decrease in the results of tensile strength is caused also by uncompletely distribution of each constituent of bioplastic. Plasticizer has two OH groups that are used to interact with the starch molecule and interacts with the amine group of chitosan that will form a new hydrogen bonds [13].

Meanwhile, figure 3 also shows that with increasing concentrations of sorbitol, cause the value of tensile strength of bioplastics decreased. This case shows the addition of sorbitol inversely proportional to the tensile strength of bioplastics produced. When sorbitol is added, the molecules of the plasticizer in the solution lies between biopolymer bonding chain and be able to interact by forming hydrogen bonds in the bond between the polymer chains resulting in weak interactions between molecules biopolymers [14].
3.4. Scanning Electron Microscopy (SEM)

Characteristic of SEM jackfruit seed starch, bioplastic without chitosan as filler and sorbitol as plasticizer and bioplastic with adding chitosan as filler and sorbitol as plasticizer are shown on figure 4 below.

![Figure 4. Jackfruit seed starch with magnification 1000x](image)

![Figure 5. Bioplastic without adding chitosan as filler and sorbitol as plasticizer with magnification 1000x](image)

![Figure 6. Bioplastic with chitosan content 2 grams and sorbitol 25% with magnification 1000x](image)

Figure 4 shows the result of analyzing SEM on jackfruit seed starch. The morphological jackfruit seed starch didn’t have a uniform particle size. This is due to sieving process carried out at a size of 100 mesh, so that particles with a size smaller than 100 mesh also passed the sifting process. The surface morphology of starch varies depending on the type of starch source. SEM analysis results of jackfruit seed starch had a semi-round shape granules. Marta et al., (2014) showed the similar results with observations using 1000x magnification electron micrograph. The jackfruit seed starch had a spherical shape with an average size of granules 6-13 μm [15].

The SEM analysis results of jackfruit seed starch shows that starch granules had a size of 7.6 μm. In the classification of starch, there are two types of starch granule size, ie small granule size (5-10 m) and large granule size (25-40 m) [16]. Based on the classification, jackfruit seed starch had a small granule size.

From figure 5 can be seen the presence of white granules of jackfruit seed starch of bioplastic without adding chitosan as filler and sorbitol as plasticizer which indicates that jackfruit seed starch do not evenly mixed in the bioplastics produced. However, there are no void fill found in bioplastic without adding chitosan as filler and sorbitol as plasticizer. Sorbitol in bioplastics will form a large cavity of bubbles. This is because sorbitol is hydrophilic so that the bioplastics is easy to fragile. This proves the above analysis of tensile strength, with increasing concentrations of sorbitol causing decreased tensile strength bioplastic [12].

From figure 6 shows that jackfruit seed starch and chitosan completely dissolved in bioplastic solution and produced a homogeneous bioplastics. From the analysis of SEM is also can be seen the void fill on bioplastics produced. This is due to the the void that is trapped in bioplastics when mixing. Figure 6 shows the surface of bioplastic looked smooth and more compact because the existing of adding plasticizer and a little bit found space for the existing of adding chitosan as filler so when the tensile strength was observed, the product will have better capability [8].
4. Conclusion

Based on the results of research can be concluded that the analysis of jackfruit seed starch obtained moisture content 6.04 %, ash content 1.08 %, starch content 70.22 %, amylpectin content 16.39 %, amylopectin content 53.83 %, protein content 4.68 %, and fat content 0.54 %. The best condition of bioplastic from jackfruit seed starch obtained at comparison of starch : chitosan (w/w) = 8:2 and concentration of sorbitol 25 % with tensile strength 13,524 MPa.

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