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PRODUCTION OF BIOPLASTIC FROM JACKFRUIT SEED STARCH *(Artocarpus heterophyllus)* REINFORCED WITH CHITOSAN USING SORBITOL AS PLASTICIZER

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ABSTRACT

The work was aimed to determine the characteristics of jackfruit seeds and investigate the effect of chitosan and sorbitol on the physicochemical properties of bioplastics. Bioplastics were prepared from jackfruit seed starch and reinforced with chitosan with composition ratio were 7: 3, 8: 2 and 9: 1 (w/w) and using sorbitol as plasticizer with variation of 0.2, 0.25, 0.3, 0.35, and 0.40 (v/w of starch). The jackfruit seed starch has water content 6.04%, ash content 1.08%, protein content 4.68%, fat content 0.54%, starch content 70.22% with amylose content 16.39% and amylopectin content 53.83%. Gelatinization profil of jackfruit seed starch was performed by rapid visco analyzer (RVA). The starch began to gelatinize at temperature of 88.82 °C. Physical properties of bioplastic were tested through density, water uptake and FT-IR analysis. It was observed that the density increased as the amount of the chitosan increased but the water uptake decreased and indicated an increase for the OH group and the group NH on bioplastics due to the addition of chitosan and sorbitol. The best conditions of bioplastics obtained at a ratio of starch: chitosan (w/w) = 8: 2 and the concentration of plasticizer sorbitol 25% with tensile strength 13.524 MPa, elongation at break 14.67%, and modulus young 92.188 MPa. The analysis of scanning electron microscopy (SEM) showing bioplastic has a smooth fracture surface and slightly hollow compare to bioplastic without fillers chitosan and plasticizer sorbitol.

Keywords: bioplastic, chitosan, jackfruit seed, sorbitol.

INTRODUCTION

Plastics, which consist of polymers and additives, today are one of the most used versatile materials [1]. Almost all the available plastics are manufactured synthetically and they have much better properties than naturally occuring plastics but it takes 50 years to decompose in nature [2] [3]. Therefore, conventional plastic is not considered environmentally friendly because it is not biologically degraded lands and will certainly contaminate the soil [4]. Yet society is fundamentally ambivalent toward plastics, due to their environmental implications, so interest in bioplastics has sparked [1].

Bioplastics utilizing native starch derived from diverse botanical source have been widely studied [5]. Film made of starch have a significant characteristics. It has a strong and extensible characteristic than proteinaceous films [6]. These plastics can be decomposed by microorganisms naturally. Starch is a polysaccharide found in the amyloplasts of most green plants [7]. Owing to the hydrophilic properties of starch, it provides a minimal barrier to water. Nevertheless, starch films possess good barrier properties to oxygen, carbon dioxide and lipids and protect against lipid oxidation [6].

Jackfruit can contain from 100 to 500 seeds, which represent 8-15% of the total fruit weight with high starch content [8][9].

To improve the mechanical properties of bioplastics is to add plasticizer and filler. Plasticizer which is a low molecular weight nonvolatile substance, into the film to reduce biopolymer chain-to-chain interactions, thereby resulting in improved film flexibility and stretch ability [10]. Sorbitol is a plasticizer which is good enough to reduce internal hydrogen bonds that will improve the intermolecular distance [11]. Chitosan is used as a filler to reduce the hydrophilic nature (less resistant to water) bioplastics is by mixing starch with biopolymers hydrophobic (water resistant) such as cellulose and chitosan [12] [13].

The aim of this research is to obtain the effect of chitosan and sorbitol on the physicochemical properties of bioplastics.

METHODOLOGY

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 [13].

Starch Extraction of Jackfruit Seed Starch

Jackfruit seeds (100 gr) were peeled and washed with 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° C for 30 minutes. Then starch was sieved with strainer 100 mesh [13].

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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 °C for 25 minutes using hotplate. After 25 minutes, sorbitol was added with variation 20%, 25%, 30%, 35% and 40% on starch solution. After stirred for 15 minutes, the solution was cooled down and cast onto flat and dried with temperature 60 °C for 24 hours. After bioplastic dried, it was removed from the flat and saved in the desicator and ready to be analyzed [13].

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 [13]. Rapid Visco Analysis (RVA) was also observed in Jasa Uji Ilmu, Fakultas Teknologi Industri Pertanian, Universitas Padjajaran.

Characterization of Bioplastics

FTIR analysis of bioplastic was observed in Laboratorium Penelitian, Fakultas Farmasi, University of Sumatera Utara. Tensile strength, test is based on ASTM standard D638, density test based on ASTM standard D792-91, water uptake test based on ASTM standard D570-98. SEM analysis was observed in Laboratorium Fisika, Universitas Negeri Medan [13].

RESULTS AND DISCUSSIONS

Chemical Properties of Jackfruit Seed Starch

Based on sedimentation method [14], from 100 grams of jackfruit seed could produce jackfruit seed starch 26,67 grams or percentage of starch was 26,67% [13].

Table-1. Chemical properties of jackfruit seed starch.

Components of Starch	Percentage (%)
Starch (amylum)	70.22
Amylopectin	16.39
Amylose	58.83
Water	6.04
Ash	1.08
Fat	0.54
Protein	4.68

The chemical composition of the starch in the seeds showed protein (4.68%) and fat (0.54%) similar to those reported by Madruga *et al.* (2014) for protein (7.98%) and fat (0.59%) [8]. The starch content of jackfruit seeds were 70.22%, respectively lower than the 92.8% early study of Madruga *et al* (2014)[8]. The ash and water content of starch were 1.08% and 6.04%. These results were similar with the findings of Noor *et al.* (2014) who reported that the moisture content of jackfruit seed starch was 6.28% [15]. These results are in accordance with minimum specifications required by Indonesian Industry Standard for commercial starches used in food industry, which allows up to 14% moisture and 1.5% ash and requires at least 75% starch [16].

Pasting Properties of Jackfruit Seed Starch

The pasting properties of starch were determined by Ravid Visco Analyzer (RVA).

Table-2. Pasting properties of jackfruit seed starch.

Parameters	Result	Unit
Pasting Temperature	88.82	°C
Peak Viscosity	3276.55	cP
Hold Viscosity	2453.5	cP
Final Viscosity	5366	cP
Breakdown	823	cP
Setback 1	2912.5	cP

Increasing temperatures lead to starch gelatinisation, which increased viscosity due to the swelling of starch granules. The temperature at which granules begin to swell is called the pasting temperature (i.e., the initial gelatinisation temperature when the viscosity curve starts) [8]. Pasting temperature of jackfruit seed starch obtained at 88.82 °C which was higher than pasting temperature (83.15 °C) reported by Madruga *et al.* (2014) [8]. The difference in gelatinization temperature depends on the microstructure and degree of crystalinity within the granule and also on granule size and the amylose to amylopection ratio [17]. Starch granules begin to swell causing a rapid increase in viscosity will result in peak viscosity [18]. The peak viscosity of starch achieved was 3276.55 cP. Granule structure cracked causing a decrease in paste viscosity and low viscosity stability of the paste. After previously reaching peak viscosity, there was a decrease in viscosity to 2453.5 cP. This viscosity is called hold viscosity [18]. Final viscosity indicates the ability of starch to form a thick paste or gel after removal and cooling processes. This change in viscosity during cooling is called setback 1 [18]. Final viscosity of starch was 5366 cP and setback 1 was 2912.5 cP.

Fourier Transform Infrared (FTIR) Result

Result of Fourier Transform Infrared (FTIR) indicated jackfruit seed starch which owns O-H Alcohol

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(H-bonded), C-H alkanes (stretch) and C-H aldehydes, C-O amide, S=O sulfates, sulfonamides, sulfones and C-O ester [13]. The presence of OH group binded with hydrogen, stretching of C-H alkanes, 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 $(C_6H_{10}O_5)n$ [19].

Result of FTIR indicated chitosan which owns the hydrogen bonding groups O-H, C-H alkanes (stretch), N-H amide, C=O amide, C-N amine, ether groups and C-O ester [13]. 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 3433,29 cm^{1} of wave number are able to form a strong ionic bonds, hydroxyl group (OH) which is negatively charged and the amine group (NH_2) positively charged [7] [19].

On bioplastic without adding chitosan and sorbitol could 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. On bioplastic with adding chitosan and sorbitol showed the same group also [13]. But there is an emergence of wave number 1593.20 cm^{-1} which indicated the existance of N-H (amino) group from chitosan on bioplastic with adding chitosan and sorbitol [13].

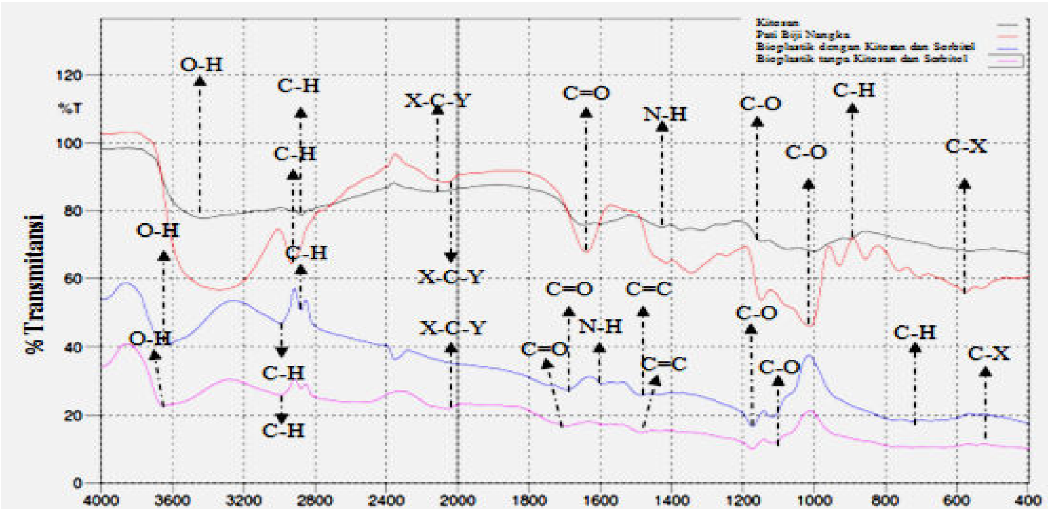

Figure-1. The result of Fourier Transform Infra Red (FTIR).

The Effect of Chitosan and Sorbitol Addition on Density Bioplastic

The highest value of density was obtained in bioplastic using 3 grams of chitosan and 20% sorbitol with a value of 1.667 gram/cm^3. While the lowest density value was obtained in bioplastic using 1 gram of chitosan and 40% sorbitol with a value of 0.765 gram/cm^3.

Figure-2 shows the density increased as the amount of the chitosan increased while the density decreased as the amount of sorbitol increased. This due to the addition of chitosan which fills and increases the density of bioplastic structure [18]. The addition of plasticizers causes network to swell and results in the decrease of network density [21].

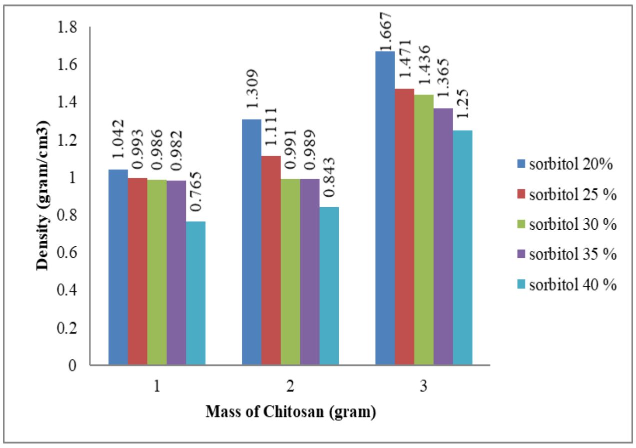

Figure-2. Density result of bioplastic.

The effect of Chitosan and Sorbitol Addition on Water Uptake Bioplastic

The maximum water uptake value of bioplastic was obtained in bioplastic with 1 gram of chitosan and 40% sorbitol with a value of 68.96%. While the minimum water uptake value if bioplastic was obtained in bioplastic with 3 grams of chitosan and 20% sorbitol.

Figure-3 shows as the amount of chitosan increased, the absorption of water decreased, this is due to chitosan is a hydrophobic compound [18]. While water

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uptake increased as the amount of sorbitol increased, due to sorbitol absorb moisture over time which is likely due to the hydrophilic nature of sorbitol [22].

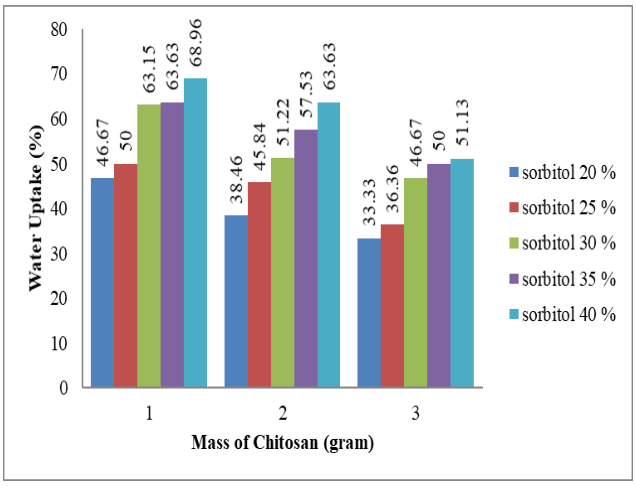

Figure-3. Water Uptake result of bioplastic.

The Effect of Chitosan and Sorbitol Addition on Tensile Strength Bioplastic

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% [13].

Figure-4 shows as the amount of chitosan increased, the tensile strength value also increased. The addition of chitosan affects the chemical bonds constituent of bioplastic during the mixing process, thus improved its mechanical strength. It is depending on the amount and type of chemical bonds (covalent bonds, hydrogen and van der walls) [23]. 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 [13] [22].

A drop tensile strength value is shown for bioplastic with chitosan content 3 grams and concentration of sorbitol 25% [13]. Susilawati *et al.* (2019) reported that the decrease tensile strength can be caused by too much chitosan being used so that the chitosan formed a very thick film [25]. High levels of fillers may reduce the interaction between the filler and the polymer matrix due to the agglomeration and non-uniform dispersion of the filler. The results of this test suggest that there is an optimum concentration of the filler to make the maximum tensile strength of the biocomposites films [26].

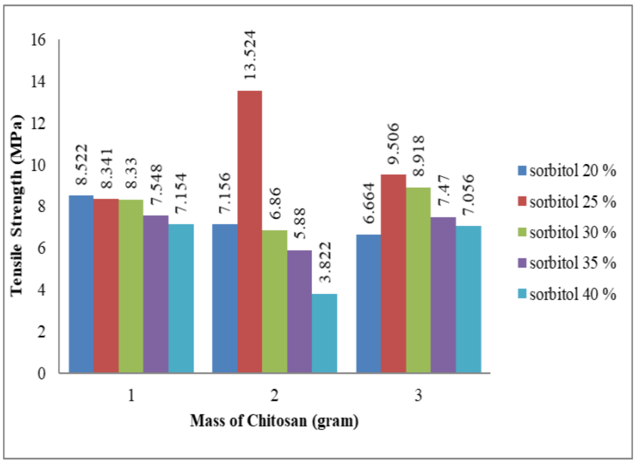

Figure-4. Tensile strenght result of bioplastic

The Effect of Chitosan and Sorbitol Addition on Elongation at Break Bioplastic

The highest elongation at break value is the addition of 1 gram of chitosan and sorbitol 40% which is equal to 22.09% while the lowest elongation at break is the addition of 3 gram of chitosan and sorbitol 20% which is 12.69%.

Figure-5 shows as the amount of chitosan increased, the elongation at break value decreased. The percentage of elongation is inversely proportional to the addition of fillers, so the more fillers the percentage of elongation will decrease [27]. Meanwhile, increasing the amount of sorbitol can cause the elongation at break value of bioplastic also increase. The addition of plasticizers serves as a giver of elastic properties in bioplastics, so the more plasticizers given will increase the value of plastic extension [27].

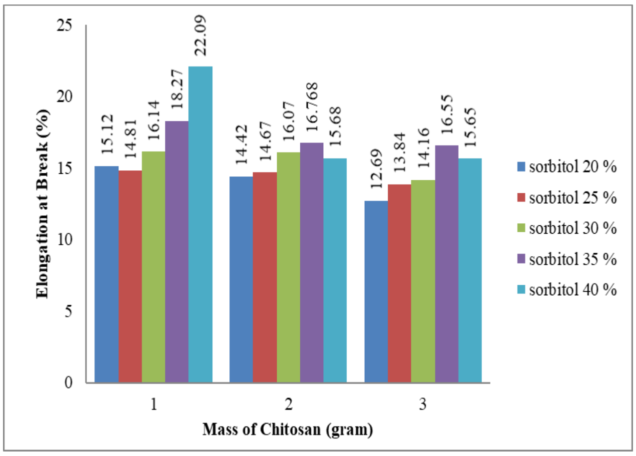

Figure-5. Elongation at break result of bioplastic

The Effect of Chitosan and Sorbitol Addition on Modulus Young Bioplastic

Modulus young determined the stiffness of the bioplastic, where the modulus young is the ratio between tensile strength and elongation at break [18]. The highest modulus young value is the addition of 2 gram of chitosan and sorbitol 25% which is equal to 92.188 MPa while the

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lowest modulus young value is the addition of 2 gram of chitosan and sorbitol 40% which is 24.375 MPa.

Figure-5 shows as the amount of chitosan increased, the modulus young value increased while the amount of sorbitol increased, modulus young decreased. The higher of adding chitosan can cause the enhancement on the value of Modulus Young. This thing can be caused by higher the compactness of intermolecular bond in bioplastic because of hydrogen bonds when the adding of chitosan, so the formed of bioplastic becomes stronger and more rigid [23].

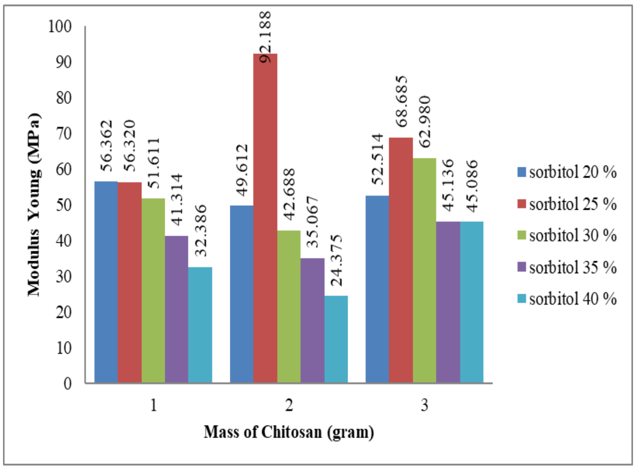

Figure-6. Modulus young result of bioplastic.

Scanning Electron Microscopy (SEM) Result

Figure-7 shows the result of analyzing SEM on jackfruit seed starch. Jackfruit seed starch have a variance in 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 spherical 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 [8][13]. Also Suryadevara *et al.* (2017) obtained jackfruit seed starch exhibited spherical free flowing with low dense form of starch grains [28].

From Figure-8 can be seen the presence of intact starch granules (non-gelatinized starch granules) of jackfruit seed starch of bioplastic without adding chitosan as filler and sorbitol as plasticizer which means that the starch was not fully gelatinized during the film forming process [29].

From Figure-9 shows that jackfruit seed starch and chitosan completely dissolved in bioplastic solution and produced a homogeneous bioplastics. From the analysis of SEM also can be seen the presemce of grooves on bioplastic produced. The occurance of grooves may be explained by the presence of microbubbles formed during the gelatinization process [29]. Figure-9 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 [13].

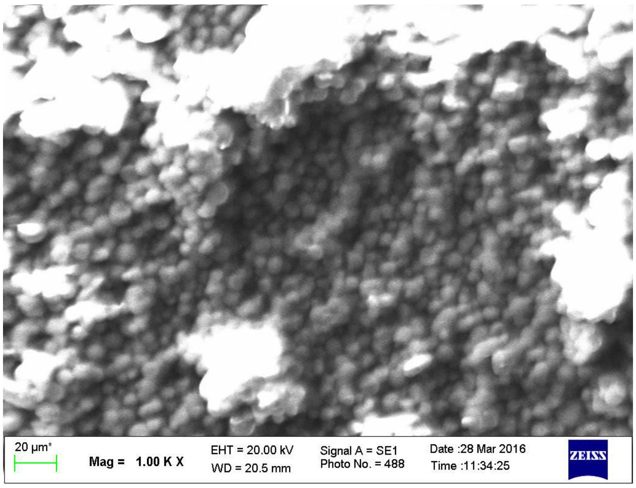

Figure-7. Jackfruit seed starch with magnification 1000x.

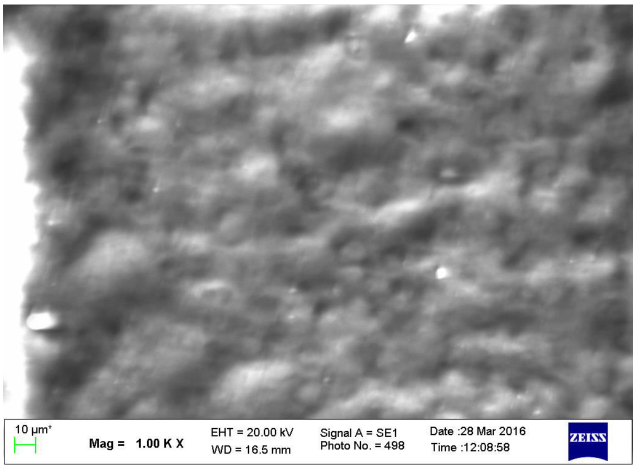

Figure-8. Bioplastic without adding chitosan as filler and sorbitol as plasticizer with magnification 1000x.

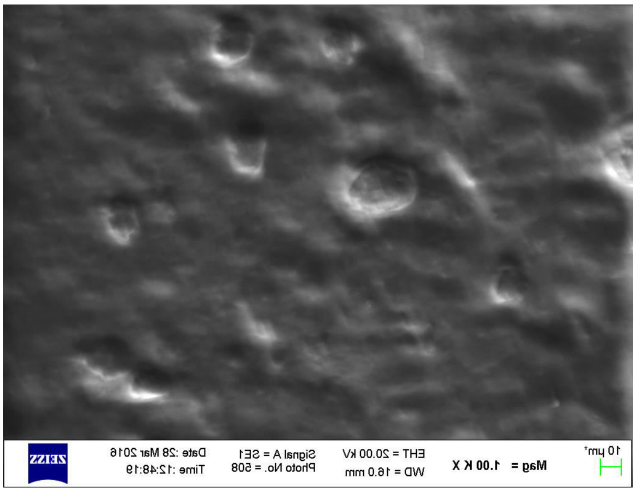

Figure-9. Bioplastic with chitosan content 2 grams and sorbitol 25% with magnification 1000 x.

CONCLUSIONS

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 %, amylose 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, elongation at break 14.67%, and modulus young 92.188 MPa. Bioplastic with adding chitosan and sorbitol has a smooth fracture surface and slightly hollow compare to bioplastic without fillers chitosan and plasticizer sorbitol.

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