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The Effect of Addition of Polyethylene Glycol (PEG) on Biodegradable Plastic Based on Bacterial Cellulosa from Coconut Water (Coconus Nucifera)

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Abstract - Plastic was the most widely used polymer in life and causes environmental pollution, plastic cannot be destroyed quickly and naturally by destructive microbes in the soil. Therefore research has been carried out on biodegradable plastics by utilizing bacterial cellulose (BC) produced by Acetobacter xylinum with coconut water media and with the addition of polyethylene glycol (PEG) plasticizers. The addition of plasticizer was done by immersion of BC in the various PEG concentration and molecular weight, namely: PEG 400 3.5%; 7%; 10.5%; 14% and PEG 4000, 6000 10.5%. The results showed that the water content and the degree of swelling of BC-PEG decreased as in created the more concentrations and molecular weights of PEG added. The degradation of BC-PEG plastics was be affected by the concentrations added to BC, the higher the concentration used then the degradation would increase. The highest quality of plastic was obtained from BC-PEG 400, 10.5% with tensile strength of 4.46 kN / m. Furthermore, the FTIR analysis of BC-PEG showed the existence of functional groups in bacterial cellulose namely: O-H, C-H, C=H,C-O-C, and there are no new functional groups appeared in the BC-PEG plastic. And small increasing in showed that the degree of crystallinity obtained from BC-PEG plastic.

Keywords — Biodegradable Plastic, Polyethylene Glykol (PEG), Acetobacter xylinum, coconut water, FTIR, XRD.

I. INTRODUCTION

Plastic has become a necessity of life that continues to increase in number. Every year around 100 million tons of synthetic plastic is produced for use in various industrial sectors which increases plastic waste every year. The plastic used today is synthetic polymer plastic made from petroleum (non-renewable) which cannot be degraded by microorganisms in the environment [1]. Plastic cannot be destroyed quickly and naturally by destructive microbes in the soil. The amount of plastic that we use results in the accumulation of waste and is a cause of pollution and environmental damage. This causes the use of plastic packaging can not be sustained widespread use because it will increase environmental and health problems in the future [2]. Thus, innovation was needed in the manufacture of environmentally-friendly plastics and raw materials obtained from materials that are easily obtained and available in nature in large quantities but can produce products with the same strength, biodegradable plastic.

Biodegradable plastic was polymers which naturally can be easily degraded either by microorganism attacks or by the weather (humidity and sunlight radiation). Biodegradable plastic is plastic that can be used like conventional plastic but will be destroyed by microorganisms activity after it is used up and discarded. Usually, conventional plastic made from petroleum, natural gas, or coal. While bioplastics are made from renewable materials, namely from compounds found in plants such as starch, cellulose, collagen, casein, protein or lipids found in animals [3].

Cellulose was very abundant, inexpensive and widely available in carbohydrate polymers, which can be extracted traditionally from plants or waste. [4]. In addition to being in plants, cellulose can also be produced by low-level animals namely bacteria [5]. Cellulose produced by bacteria was

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called bacterial cellulose (BC), the bacterium used was *Acetobacter xylinum (A. Xylinum),* this bacterium was a type of bacteria that produces cellulose with beneficial physical properties [6].

Bacteria that can synthesize cellulose are *Acetobacter, Achromobacter, Aerobacter, Agrobacterium, Alcaligenes, Azotobacter, Pseudomonas, Rhizobium,* and *Sarcina*. Cellulose from bacteria was very interesting because the physical properties of bacterial cellulose are different from plant cellulose. Bacterial cellulose has the same chemical composition as cellulose in plants but differs in terms of three-dimensional structure, degree of polymerization, and physiochemical properties [7].

Cellulose that was synthesized by microorganisms was usually called bacterial cellulose (BC). BC was made from coconut water which was inoculated with the bacterium *Acetobacter xylinum* [8]

Plasticizers used to increase the plastic properties of polyethylene glycol (PEG). Plasticizers from the ethylene glycol group are widely used in the plastics industry, especially in making polyester fibers and resins. Plasticizers from the ethylene glycol group have good physical and mechanical properties, such as good solubility in water and organic solvents, low toxic levels, and high hydrophilic properties [9].

Therefore, in increasing the utilization of coconut water to reduce the use of conventional plastic which was difficult to degrade which causes environmental pollution, the authors are interested in making research on "the effect of adding polyethylene glycol (PEG) on biodegradable plastic cellulose-based biodegradable plastic from coconut water".

II. Research Method

1.1 Preparation of Coconut Water

Coconut water was filtered using a filter and filter cloth afterward, put in a container pot.

1.2 Preparation of Bacterisl Cellulose

The BC used was obtained from the Biochemistry laboratory of the Department of Biology FMIPA UNP which was then developed on their own according to the needs of the experiment. Making a starter is done by sterilizing all equipment that used such as glass bottles and newspapers in advance in the oven at 100 ° C for 15 minutes.

For 1 liter of coconut water add 100 grams of sugar, 10 grams of urea and 20 mL of acetic acid. The medium was heated to boiling and in a hot state was transferred into a glass bottle. The medium is left to room temperature.

Then, entered the bacteria A. Xylinum to taste. then the glass bottle was closed again with newspaper and rubber bands and stored at room temperature.

1.3 Preparation of Medium Bacterial Cellulose

The aseptic coconut water medium was made by putting 600mL of coconut water into a pan, then heated to boiling. After boiling put 30 grams of sugar, 6 grams of urea, and 12 mL of acetic acid. All ingredients were stirred for ± 5 minutes. The medium was transferred into a plastic container and covered with a newspaper that has been sterilized beforehand. The medium was left to room temperature.

1.4 Preparation of bacterial cellulose

The fermentation mediums were cooled at room temperature into a plastic container, inoculated with *A. Xylinum* starter at a ratio of 10: 1 (% v / v) and fermented at room temperature to form bacterial cellulose at least 0.5cm. During the inoculation process, the container must not shake.

1.5 Purification of Bacterial Cellulose

The formed BC washed with running water for 24 hours, then soaked with 2% NaOH (% w / v) for 24 hours. After that proceed with running water until clean. The cleaning process was done so that the BC that has formed does not become smelly and rotten due to the growth of mold.

After the BC was clean, then cut into several parts and soaked into the PEG with the concentration that has been determined, soaking was done for 4-5 days, so that the PEG can enter into the SB. The PEG used was: PEG 400 by 3.5%; 7%; 10,5%; 14%; PEG 4000 and 6000 as much as 10.5%

1.6 Characteristics of Biodegradable Plastics

The obtained of biodegradable plastik was tested with the following parameters:

1.6.1 Physical Characteristics of Biodegradable Plastics

Analysis of water content in BC where cellulose with a size of 4cm x 2cm, weighed with an analytical balance. Then BC in the oven to dry with temperatures > 100 ° C. Percent of water contained could be calculated using the equation:

$$\text{water content}\% = \frac{a-b}{b} \text{ x } 100\%$$

Where:

a = initial weight (gram)

b = dry weigh (gram)

Swelling test, each plastic from BC fermentation with a various composition of the addition of PEG was then weighed to a constant weight and recorded as initial weight. Then the plastic was soaked in 20 mL distilled water and left for 24 hours. Then the plastic was removed and the surface was wiped with a tissue, then weighed. This treatment was carried out until the plastic weight is constant. Calculating the degree of swelling using the following equation:

$$\text{Swelling\%} = \frac{a-b}{b} \text{ x } 100\%$$

Where:

a = constant weight
b = initial weight

1.6.2 Mechanical Properties of Biodegradable Plastics

Tensile strength was measured using the Tensile Strenght industries model SSB 0500. Analysis of plastic tensile strength was done through data obtained from a tensometer.

The amount of tensile strength can be calculated by:

$$\text{Elasticity} = \frac{stress\ (MPa)}{\text{strain}}$$

The measurement of elongation strength was carried out in the same way as tensile strength testing. Elongation was expressed as a percentage, calculated by:

$$\text{Elongation}(\%) = \frac{\text{strain when breaking up (mm)}}{initial\ length\ (mm)} \text{ x } 100\%$$

1.6.3 Biodegradation

Biodegradation analysis of BC plastic sheets was done by burying plastic sheets in the soil at a depth of 15cm. The burial process was carried out for 21 days. Before being buried, the plastic was weighed in mass, then buried in the ground for 21 days at daily weighing intervals. Decomposed plastic could be calculated through the following equation:

$$\text{\% biodegradation} = \frac{a-b}{b} \text{ x } 100\%$$

Where:

a = initial weight

b = final weight

1.6.4 Molecular Structures of Biodegradable Plastic

Function Group analysis was performed using a Fourier transform infrared spectrophotometer (FTIR). Samples in the form of the film were placed into a cell holder at room temperature. FTIR analyzed the BC functional groups that were recorded using a spectrophotometer at room temperature. Before testing, the sample holder was cleaned using 70% ethanol. BC plastic samples were placed above the sample holder and the tool was ready to operate. The source gauge on the swing arm was set with a wavenumber of 90-100 cm-1 (for solid samples). Samples were moved with 4000-600 cm-1 wavenumbers. The monitor get a spectrum that analyzed to determine the functional groups contained in BC plastic. The results obtained from measurements using FTIR were spectrum, where the curves formed were curves with horizontal lines representing wave numbers and vertical lines representing% transmittance.

Plastic crystallinity analysis was carried out using X-Ray Diffraction (XRD). Cut plastic with a size of 1cm x 1 cm then put it into a sample template coated with wax. The degree of crystallinity (Xc) was determined using the following equation:

$$\text{Crystalline degree\%} = \frac{\text{a}}{\text{wide area (a+b)}} \text{ x } 100\%$$

Where:

a = wide crystalline (gram)

b = amorphous (gram)

III. Results And Discussion

1.7 Bacterial Cellulose Polyethylene Glycol

BC made from coconut water with the addition of sugar, urea and acetic acid. In making BC 2 types were done, namely by in-situ and ex-situ. The in-situ method was by directly inserting the plasticizer that used in the manufacture of BC, while the ex-situ was by adding a plasticizer after the BC was formed by immersion. In this research, the two methods have been carried out.

In the in-situ method, the addition of plasticizer which was PEG with different concentrations could affect the cellulose produced. Where the increasing concentration was used, the thickness of the cellulose also decreased. cellulose was formed only on the addition of PEG plasticizers 400: 3.5% and 7%. While the addition of 10.5% and 14% cellulose was not formed. The difference thickness in BC was caused by 2 factors, namely the process of purifying BC

by the loss of non-cellulose components (residual nutrients) and transformation of the structure in BC I to cellulose II so that the BC structure shrinks.

The next method used was the ex-situ method, in which the addition of plasticizer after cellulose was formed. Cellulose which has been cut as needed for testing was immersed with PEG with different concentrations (PEG 400 3.5%; 7%; 10.5; 14% and PEG 4000, 6000 10.5 %) for 4-5 days, resulting in a different texture. where, without PEG the cellulose produced is white and solid, whereas with the addition of PEG cellulose the color is slightly yellow, slightly springy and slippery.

BC that has been soaked with PEG for the next 4 days was made into plastic BC-PEG by ironed in a maximum temperature to dry. Heating used an iron produces a better plastic surface sheet.

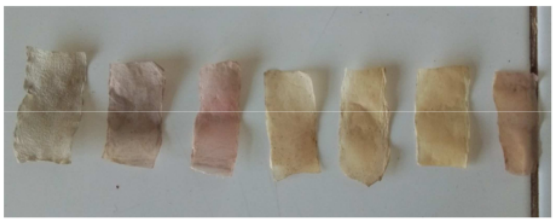

Fig. 3.1 Bacterial Cellulose Polyethylene Glykol Plastics

1.8 Physical Characteristics of Biodegradable Plastics

1.8.1 Water Content

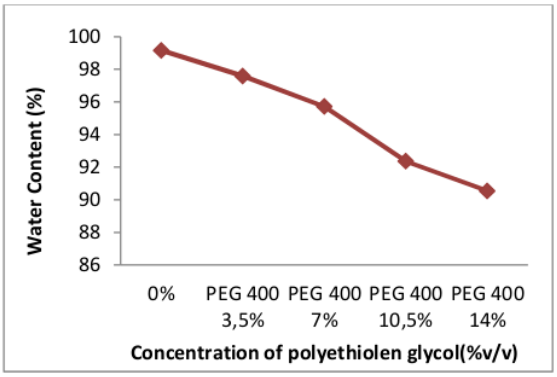

Fig. 3.2 Effect of plastic water content of bacterial cellulose polyethylene glycol on the concentration of polyethylene glycol 400

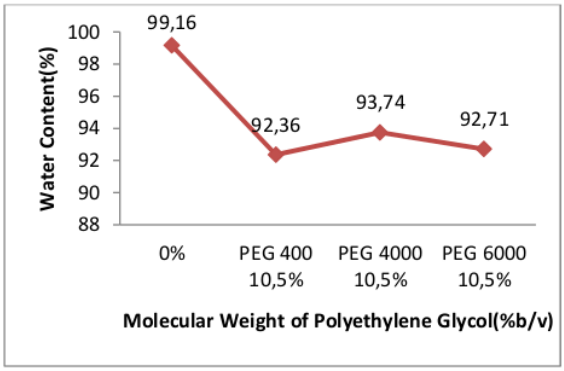

Fig. 3.3 Graph of the influence of polyethylene glycol cellulose plastics on differences in molecular weight polyethylene glycol

The increasing concentration used, the water content contained in cellulose decreased. In BC without using PEG (0%) was the highest percentage that is 99.16%.This decrease was caused by PEG which was a polymer consisting of glycol monomers having a high ability to absorb water by forming hydrogen bonds.

1.8.2 *Swelling Test*

The degree of swelling only interferes with intermolecular bonds and does not cause chemical bonds to break in the chain of a polymer. Effect of adding PEG to the percentage degree of swelling carried out for 24 hours / until a constant weight (for 5 days)

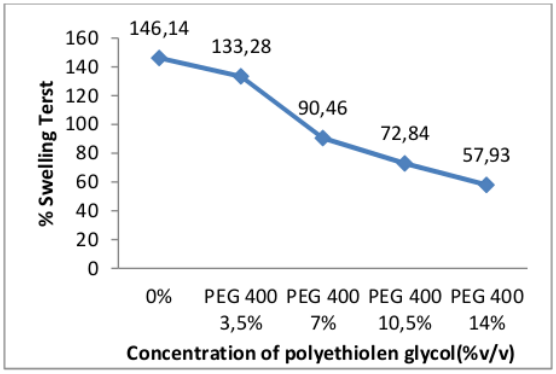

Fig. 3.4 Graphs of the effect of different concentrations of polyethylene glycol on the degree of plasticity of plastic cellulose polyethylene glycol

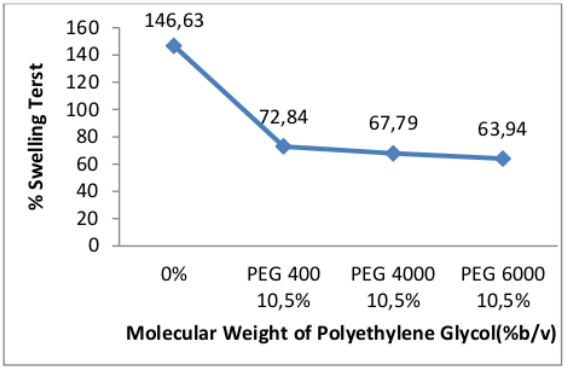

Fig. 3.5 the effect of molecular weight of polyethylene glycol on the degree of plasticity of plastic cellulose polyethylene glycol

PEG could affect the degree of swelling BC , where the high concentration used then the degree of swelling decreased, and the difference also in molecular weight of PEG with the same concentration. In this reduction, the adsorption process was occur that the interaction between adsorbent, in this case, the active substance was PEG and adsorbent was BC. The bond that occurs in the adsorption process was weak because it was a physical bond (only on the surface) so that this bond was easily broken and then the water was released again from the plastic pores.

1.9 Mechanical Properties of Bacterial Solulose Polyethylene Glycol Plastics

1.9.1 Tensile Strength

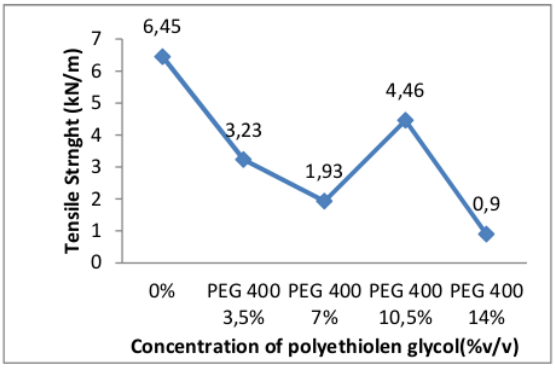

Fig. 3.6 Graphic Effect of different concentrations of polyethylene glycol on the tensile strength of bacterial polyethylene glycol plastic cellulose

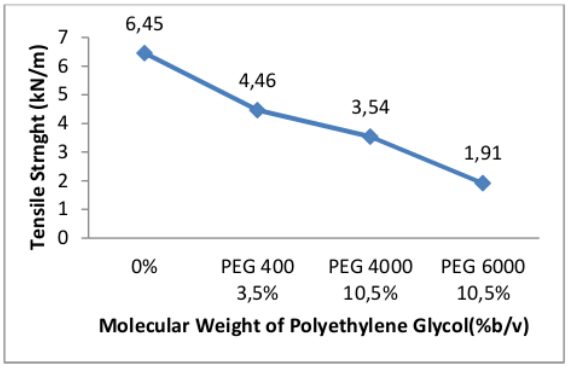

Fig. 3.7 Graph of the effect of molecular weight of polyethylene glycol on the tensile strength of polyethylene glycol bacterial cellulose plastic

The concentration of PEG to the tensile strength value was inversely proportional. Based on Rika Amalia's research (2017) [10], that tensile strength decreases along with the increasing number of plasticizers added. This decrease was related to the presence of space that occurs because the bond between polysaccharides was broken by plasticizers. This causes the bonds between molecules in the plastic film to weaken. The increase in the addition of plasticizer concentration which causes the reduced value of the tensile strength

1.9.2 Elongation

The elongation was the increase of percent the length of film material which measured from tensile strength until elongation breaking up.

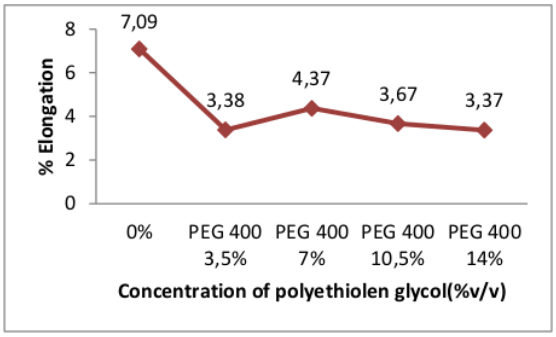

Fig. 3.8 Effect of polyethylene glycol concentration on elongation ability of bacterial cellulose plastics.

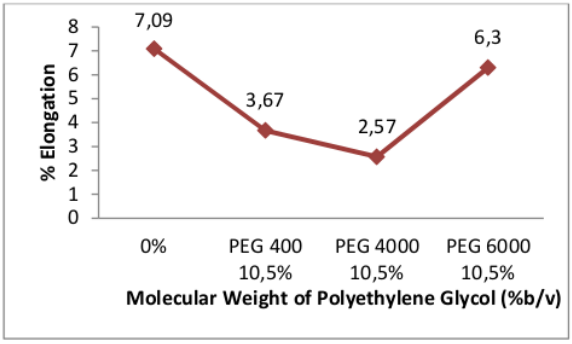

Fig. 3.9 Graph of molecular weight effect of polyethylene glycol on elongation of plastic cellulose bacterial polyethylene glycol

Based on the graph shows the elongation value decreases with the addition of PEG. This is due to the occurrence of dispersion and plasticizer interactions not only in the cellulose ring in the form of hydrogen bonds but also occur with glycoside bond chains. The steric hindrance that occurs was higher. This causes the glycoside bonds that connect between cellulose molecules to weaken because in the cellulose chain there was also a hydrogen bond. If it was subject to a pulling load, the oligosaccharide bond cannot maintain the strength of the cellulose chain and results in low elongation.

1.10 Biodegradation

Biodegradation was a process by which microorganisms could degrade or break down the natural polymers (lignin, cellulose) and synthetic polymers (polyethylene, polistire). Each microorganisms have different degradation characteristics and therefore differs from one microorganism to another microorganism.

Biodegradation test of BC-PEG plastic whith buryed bioplastics in the soil, the occurrence of biodegradation was marked by breaking the polymer chain which was marked by weight loss. The soil used for biodegradation testing was plantation soil. The burial was held for 21 days.

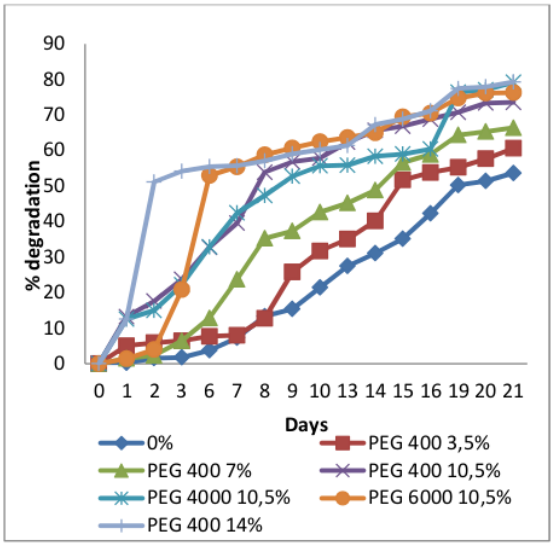

Fig. 3.10 Graph Effect of Degradation Time Against Percent Loss of Cellulose Polyethylene Glycol Plastic

PEG was a hydrophilic plasticizer. Hydrophilic plasticizers increased the rate of biodegradation, and the addition of hydrophilic plasticizer concentrations increased the rate of enzymatic degradation. Increasing the amount of hydrophilic plasticizer increasde the number of polar groups (-OH) in the sample, and cause an interaction between the polar groups with water molecules in the hydrolysis process, a process that causes biodegradation. As a result of the interaction between polar groups found in plasticizers and water molecules, the degrading microorganisms found in the media also more easily interacted with the polymer surface and have greater access to attach to the polymer surface, so that the biodegradation process can be faster.

1.11 Function Groups of Polyethylene Glycol Plastic Bacterial Cellulose

Analysis of functional groups contained in BC-PEG plastics was carried out with Fourier Transform Infrared Spectrophotometer (FTIR).

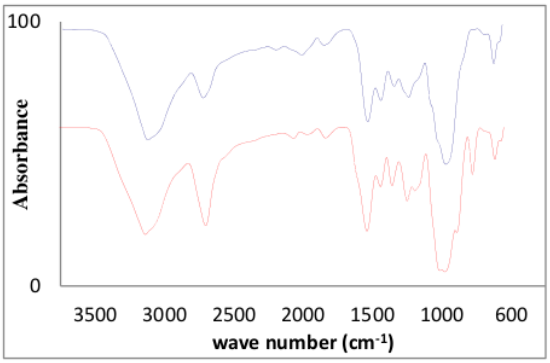

Fig. 3.11 FTIR spectrum of bacterial cellulose and bacterial cellulose polyethylene glycol

From the graph, the blue spectrum was the BC spectrum and the red spectrum was BC-PEG 400 10.5%. In the spectrum BC-PEG, there is no addition new spectrum of BC, and the spectrum shift occurs not too far, because the functional groups contained in BC and PEG were the same and do not have many different wavelengths.

Table 3.1 Values of Wavelengths and Functional Groups in Each Sample

sample	Peak (cm^{-1})			
	O-H	**C-H**	**C=H**	**C-O-C**
0%	3338.70	2911.15	1639.35	1035.18
10,5%	3343.89	2881.77	1646.78	1055.90

1.12 Plasticity of Bacterial Cellulose Polyethylene Glycol Plasticity

Crystallinity test was carried out to determine the degree of crystallinity of BC-PEG plastic**.** This analysis was carried out using an X-ray Diffractogram (XRD). In the XRD analysis the samples used were BC and BC-PEG 400 10.5%. BC-PEG 400 10.5% was chosen because it showed the optimum breaking strength results, while BC as a control.

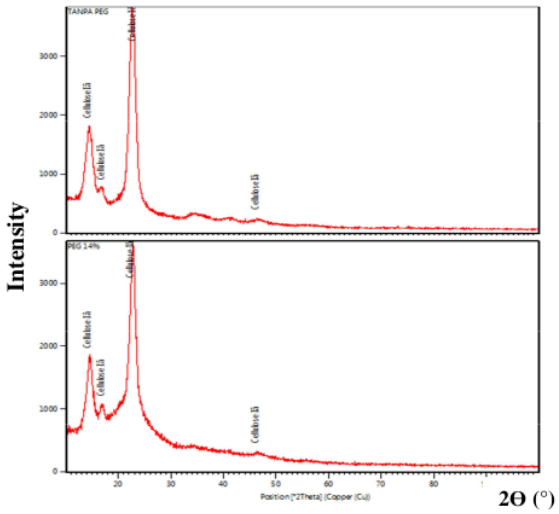

Fig. 3.10 Diffragram XRD bacterial cellulose and bacterial cellulose polyethylene glycol 400 10%

Based on the Jayme-Knollemethode method used in determining the degree of crystallinity of cellulose, according to PH Hermans and A.Weidinger, this crystallinity test was carried out by copying traces of photometer lines on transparent paper or millimeter paper that are known to weigh per unit surface, cutting images, weigh paper and take grades. The percentage value of the degree of crystallinity can be seen in Table 3.2 below:

Table 3.2 % Degrees of Crystallinity of bacterial cellulose and bacterial cellulose polyethylene glycol 400 10.5%

Sample	**m_{total} (gram)**	**m_{armorf} (gram)**	**$m_{crystal}$ (gram)**	**%crystallinity**
BC	0,053	0,01	0,043	81,14%
BC-PEG 400 10%	0,04	0,007	0,033	82,5%

The addition of polyethylene glycol as a plasticizer can increase crystallinity because PEG molecules that function as filters of BC plastic.The addition of PEG as a filter in the preparation of BC plastic has been proven to reduce the value

of the degree of crystallinity of BC. This is due to reduced molecular order (amofr) by PEG plasticizer.

IV. CONCLUSION

Immersion of BC-PEG could affect the texture and color of cellulose.

The addition of PEG can affect the water content and degree of swelling, where the more concentrated and molecular weight of PEG, the lower the water content and the degree of swelling BC-PEG plastic. The highest quality plastic was obtained from PEG 400 10.5% BC plastic based on tensile strength.

Based on FTIR analysis showed the presence of functional groups in BC but there are no new functional groups in BC-PEG. whereas, the analysis of crystallinity showed that the addition of PEG will increase the crystallinity of BC plastic.

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