Effect of Gelatinization Temperature and pH and Addition of Sodium Tripolyphosphate on the Characteristics of Biodegradable Plastic from Cassava Starch (*Manihot esculenta*)

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Abstract: ***Biodegradable plastics have become a good substitute for conventional plastics because it is can be decomposed by microorganism into water, carbon dioxide, and methane. Biodegradable plastic can be made from starch and it have been widely studied and traded. Starch is one of the natural ingredients that has good properties to be formed into biodegradable plastic. However, the use of starch-based biodegradable plastic is limited due to its poor mechanical properties. Cross-linking method is one of the methods for polymer modification that is widely used to improve mechanical properties of starch-based biodegradable plastic by adding an ingredient called a cross-linking agent where it could tie together with carbon atoms from different chains of the polymer. One of the cross-linking agents is Sodium Tripolyphosphate (STTP). STTP is used because it has a low level of toxicity and is by food quality. This research aims to examine the effect of temperature and pH on the tensile strength and water absorption of the biodegradable plastic produced, to examine the effect of the concentration of sodium tripolyphosphate (STTP) on the tensile strength, water absorption, and the level of biodegradability of the biodegradable plastic. The result showed that increasing the temperature in the gelatinization process of biodegradable plastic from cassava starch can increase the tensile strength value and reduce water absorption in the resulting biodegradable plastic. Increasing the pH in the gelatinization process of biodegradable plastic from cassava starch can increase the tensile strength value and increase water absorption in the resulting biodegradable plastic. The addition of STTP levels in the process of making biodegradable plastic from cassava starch does not always increase the tensile strength value of the resulting biodegradable plastic. This can occur due to the residue of the cross-linking agent due to the addition of excess cross-linking agent so that it can interfere with the interaction between starch molecules and result in a decrease in the tensile strength value. The addition of STTP levels in the process of making biodegradable plastic from cassava starch can reduce water absorption and can increase the level of biodegradability of the resulting biodegradable plastic.***

Keywords: biodegradable plastic; cassava; cross-link; sodium tripoplyphosphate.

1. Introduction

The production of plastic waste in Indonesia is increasing. Indonesia is the largest producer of plastic waste after China [1]. Plastics are organic polymers with long carbon chains and large molecular weights [2]. This makes plastic difficult to decompose and able to last a long time in the environment [3, 4, 5]. In addition to having disadvantages, plastic has many uses. Plastics are widely used as packaging materials, both water bottles, food, medical equipment, electronic goods, construction, and so on [3]. Dependence on plastic use and poor plastic waste management are factors that cause plastic waste production to continue to increase every year [6].

In general, plastics are made using petroleum-based raw materials (petroleum based) [7]. However, the availability of petroleum as a raw material for making plastics is dwindling which allows the price of plastic to rise in the future [8].Currently, biodegradable plastic is an option to reduce the use of conventional plastics because of their effect on the environment [9]. Biodegradable plastic can be made from starch and has the advantage that it can be decomposed by microorganisms into the water, carbon dioxide, and methane [10]. This biodegradable plastic has been widely studied and traded [11]. The requirement for biodegradable plastic is to have a resistance that resembles plastic in general and can be returned safely to the ecosystem [12].

Starch is the most flexible material, in other words, it can be modified by cross-linking [13]. Cross-linking is the addition of an ingredient called a cross-linking agent to increase the viscosity and reduce the viscosity breakdown of starch [14]. One of the cross-linking agents is Sodium Tripolyphosphate (STTP). STTP is widely used because it has a low level of toxicity and is by food quality [15]. STTP is a compound containing phosphate which will increase resistance to high temperatures, and low pH, and increase the stability of starch granules. Therefore, it can increase the viscosity and texture of starch [13].

This study is based on research that has been carried out by previous workthat the manufacture of biodegradable plastic was carried out using cassava starch and glycerol as a plasticizer [1]. It indicated that the absorption value of biodegradable plastic to moisture absorption increased, the speed of plastic degradation increased, and the shelf-life also increased with the increase in glycerol concentration.

Another researcher reported that the addition of glycerol will affect the mechanical properties such as tensile strength, elongation, Young's modulus, and the ease of decomposition of the resulting biodegradable plastic. In addition, this research aims to investigate the effect of temperature and pH on the tensile strength and water absorption properties of the biodegradable plastic. The effect of sodium tripolyphosphate (STTP) concentration on the tensile strength, water absorption, and the level of biodegradability of the biodegradable plastic is also discussed in this paper.

2. Research Method

2.1 Tools and Materials Used

The materials used in this study include cassava starch obtained from PT. Organic Circle. Aquadest, NaOH, HCl, glycerol, and sodium tripolyphosphate (STPP) with variations of 1.5%, 3%, and 4.5% (w/v), and ethanol obtained from CV Indra Sari Semarang. The tools used in this study include a series of tools for making biodegradable plastic, measuring cups, Erlenmeyer, analytical balance, glass stirrers, droppers, porcelain cups, casting plates or glass, petri dish, and thermometers.

2.2 Making Biodegradable Plastics with Variations in Temperature and pH of Gelatinization

Seven point five grams of cassava starch was dissolved in a mixture of 150 ml of aquadest with 2% (v/v) glycerol. Furthermore, the addition of NaOH or HCl to adjust the pH according to predetermined variables. The mixture is stirred with a stirrer and heated at a temperature according to a predetermined variable and kept constant. After the mixture turned into a gel, the heating process was stopped and then cooled for 5 minutes at room temperature to remove any air bubbles in the mixture. Then the gel was printed on glass and dried in the sun for 3 days. The dried plastic was removed from the casting and stored in a desiccator at room temperature for further testing. In this procedure, the optimum gelatinization temperature and pH will be obtained in the manufacture of biodegradable plastics.

2.3 Production of Biodegradable Plastics with STPP Concentration

Seven point five grams of cassava starch was dissolved in a mixture of 150 ml of aquadest with 2% (v/v) glycerol. Furthermore, the addition of NaOH or HCl to adjust the pH according to the best results. The mixture is stirred with a stirrer and heated at a temperature according to the best results that have been determined and kept constant. After the mixture became gel, then STPP was added according to the predetermined variable, and stirring was continued for 5 minutes until the solution was homogeneous. Then the heating process was stopped and then cooled for 5 minutes at room temperature to remove any air bubbles in the mixture. Then the gel was printed on glass and dried in the sun for 3 days. The dried plastic was removed from the casting and stored in a desiccator at room temperature for further testing. In this procedure, the optimum STPP concentration will be obtained in the manufacture of biodegradable plastics.

2.4 Tensile Strength Test

The mechanical properties of the tensile strength (MPa) of the sample were measured at a temperature of 25^{o}C with the Mesdan Lab Testometric Machine S.p.a 25087 – Italy (Model Tenso.300, Type: 168 E) according to ASTM ID: D882-12. All film strips to be tested (5 cm × 15 cm) were pre-stored at 25^{o}C. The tensile strength can be determined by dividing the peak load by the cross-sectional area of the initial film specimen.

2.5 Water Absorption Test

This water absorption test identifies the ability of biodegradable plastic to absorb water (H_2O) which is determined by the ASTM D 570 standard. Biodegradable plastic that has previously been dried, cooled in a desiccator, then weighed and cut into 3cm x 3cm. The water absorption data of biodegradable plastic was obtained by soaking it in water for 24 hours. After that, the biodegradable plastic was dried with a cloth and immediately weighed. The water absorption capacity of biodegradable plastic can be calculated by the following equation:

$$Water\ Absorbtion\ (\%) = \frac{W_1 - W_0}{W_0} \times 100\%$$

With:

W_1 = Final weight after soaking (grams)
W_0 = Initial weight before immersion (grams)

2.6 Biodegradability Test

Biodegradable plastic is cut into 10 mm x 10 mm. Then, the biodegradable plastic was buried in soil as deep as 8 cm with a burial duration of 6 weeks with weekly analysis. Before burial, the initial mass (mass before degradation) was measured first. The final mass (mass after degradation) of the biodegradable plastic was measured afterward. Any change in mechanical properties due to the degradation process is observed and when the biodegradable plastic is completely degraded, the degradability can be calculated using the following equation:

$$Biodegradability\ (\%) = \frac{W_n - W_0}{W_0} \times 100\%$$

With:

W_n = Final weight after burial (grams)
W_0 = Initial weight before burial (grams)

3. Results and Discussion

3.1 Effect of Gelatinization pH on Water Absorption of Biodegradable Plastic

The effect of variations in gelatinization pH on the water absorption of biodegradable plastic can be shown in the following graph:

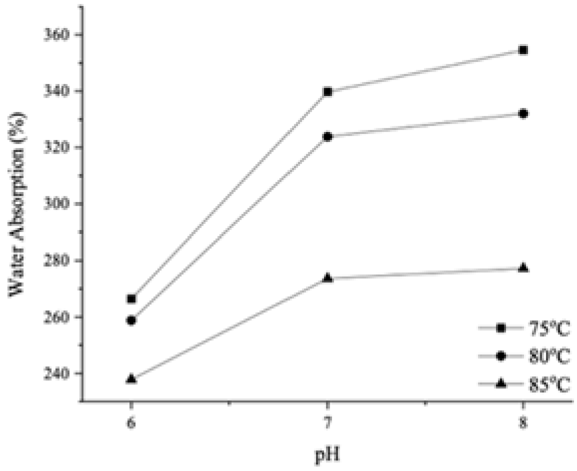

Figure 1: Effect of pH on water absorption

From Figure 1. it can be seen that the increase in pH will be followed by an increase in water absorption for each temperature variable. The lowest water absorption with a value of 237.82% was obtained at the gelatinization pH variable of 6 with a gelatinization temperature of 85°C and the highest water absorption with a value of 354.6% was obtained at the gelatinization pH variable of 8 with a gelatinization temperature of 75°C.

According to the theory, when gelatinization occurs under acidic conditions, hydrogen bonds between adjacent starch polymers are disrupted and the amorphous regions are eroded. This is what causes the swelling power or water absorption to be lower. This decrease in water absorption is caused by the entanglement of the amylopectin network viscosity in the starch crystalline region [17]. In addition, to create alkaline conditions, a NaOH solution is needed. The hydroxyl group (-OH) is hydrophilic, so the hydroxyl group will bind more water [18]. The more alkaline, the more hydroxyl groups are needed, and the more water will be bound. However, the results obtained were relatively smaller which could occur due to the addition of other substances that bind to starch, thereby reducing the ability of starch to bind water.

The recommended water absorption is around 12.41%-90.07% while the lowest water absorption is at pH 6 with a value of 237.82% [19]. Therefore, the results obtained are much higher than recommended. This can happen because during weighing there is water attached to the surface of the bioplastic, so it will increase the amount of water bound by the bioplastic itself and result in an increase in water absorption.

3.2 Effect of Gelatinization pH on Tensile Strength Biodegradable Plastic

The effect of gelatinization pH on the tensile strength of biodegradable plastic can be shown in the following graph:

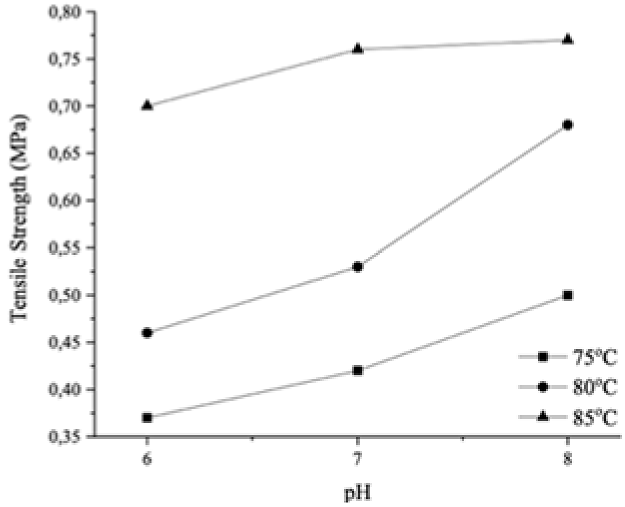

Figure 2: Effect of pH on tensile strength

It can be seen in Figure 2. shows a graph of the relationship between tensile strength and gelatinization pH. At each temperature value of 75°C, 80°C, and 85°C there was an increase in the tensile strength value along with the increase in the pH value from 6-8. Where the lowest tensile strength value of 0.37 MPa was obtained in a variable with a gelatinization pH of 6 and a gelatinization temperature of 75°C while the highest tensile strength value of 0.77 MPa was obtained in a variable with a gelatinization pH of 8 and a gelatinization temperature of 85°C.

Starch gelatinization can occur in the pH range of 4-7, if the applied pH is too high it will result in a faster gelatinization process but also retrogradation faster and vice versa if the pH is too low, will result in a slowdown in the gelatinization process. The occurrence of retrogradation will produce a stiffer polymer structure, causing the polymer to have a higher tensile strength value. It can be said that as the pH value of gelatinization increases, the tensile strength value of the biodegradable plastic obtained also increases. It can be concluded that the tennis strength data presented in Figure 2. is by the theory, that along with the increase in the pH value of gelatinization, it will affect the increase in the tensile strength value of the biodegradable plastic obtained. Previous study reported the effect of gelatinization pH on bioplastic production from cassava peel starch with a gelatinization pH range of 4-7 and at temperature of 70°C and 75°C. The lowest tensile strength value was obtained at 0.87 MPa in a variable with a gelatinization pH of 4 and a gelatinization temperature of 70°C while the highest tensile strength value of 1.18 MPa was obtained in a variable with a gelatinization pH of 5 and a gelatinization temperature of 75°C. When compared with the lowest and highest tensile strength values obtained, namely 0.37 MPa and 0.76 MPa, it can be said that the value is lower.

3.3 Effect of Gelatinization Temperature on Water Absorption of Biodegradable Plastic

The effect of variations in gelatinization temperature on the water absorption of biodegradable plastic can be shown in the following graph:

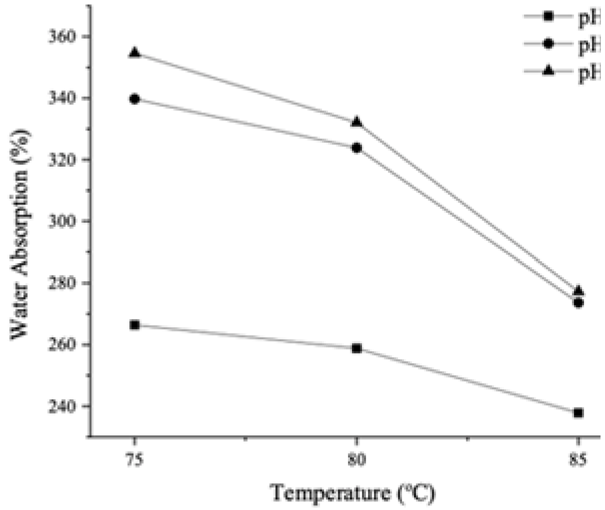

Figure 3: Effect of temperature on water absorption

From Figure 3. it can be seen that the increase in temperature will be followed by a decrease in water absorption for each temperature variable. The lowest water absorption of 237.82% was found in the variable with a gelatinization temperature of 85°C and gelatinization pH of 6 while the highest water absorption of 354.60% was obtained in the variable with a gelatinization temperature of 75°C and gelatinization pH of 8.

Previous work produced bioplastics from MuluBebe banana peel starch with variations in the addition of glycerol (1%; 2%; 3%) obtained water absorption results in the range of 12.407%-90.066% [19]. The best results were obtained with a glycerol variation of 2% with a water absorption capacity of 12,407. The water absorption obtained in this study is lower because the gelatinization temperature used is 80-90°C so that the temperature is higher than the temperature used in this study. The lower water absorption capacity of biodegradable plastic, the higher the resistance of biodegradable plastic to water and the reduced swelling (swelling) of biodegradable plastic [21]. The swelling power of starch is influenced by several factors, namely the amylopectin structure. Other factors that can affect the swelling power are the ratio of amylose to amylopectin, the degree of chemical cross-linking in the granules, and non-carbohydrate substances such as fats, phosphates, and proteins. When a starch has reached its maximum swelling power, it will reach peak viscosity. Peak viscosity is the maximum viscosity before the start of cooling the sample during stirring. High amylose content and low amylopectin can cause a decrease in peak viscosity because swelling of starch granules and pastes is associated with amylopectin level and structure. However, it is also possible that the swollen granules become very brittle and break when they start to swell [22]. Damage to these granules can affect the water-binding process when the film is formed. Another work reported that the gelatinization temperature to reach peak viscosity is 71.8ºC [22]. While the temperatures used in this study were 75ºC, 80ºC, and 85ºC so that starch granule damage can occur. This can trigger a decrease in water absorption every time the gelatinization temperature increases.

3.4 Effect of Gelatinization Temperature on Tensile Strength Biodegradable Plastic

The influence of gelatinization temperature on the tensile strength of biodegradable plastic can be shown in the following graph:

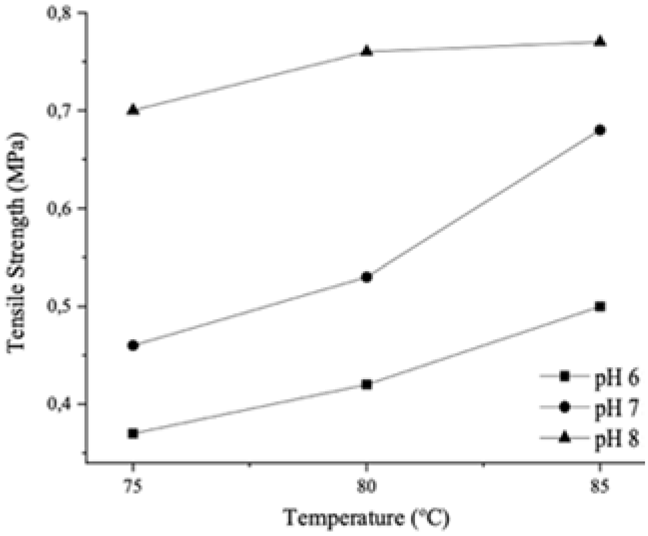

Figure 4: Effect of temperature on tensile strength

It can be seen in Figure 4. shows a graph of the relationship between tensile strength and gelatinization temperature where in general there is an increase in the tensile strength value along with the increase in gelatinization temperature at pH 6 – 8. The lowest tensile strength value of 0.37 MPa is obtained in the variable with a temperature gelatinization of 75°C and gelatinization pH of 6 while the highest tensile strength value of 0.77 MPa was obtained at a variable with a gelatinization temperature of 85°C and a gelatinization pH of 8.

Previous research reported that the higher the gelatinization temperature applied, the higher the tensile strength value of biodegradable plastic obtained will also be higher [23]. This could be due to the weakening of the intermolecular bonds of starch as the gelatinization temperature increased. The weaker the intermolecular bonds of starch, the more easily the bonds between the amylose molecules are broken. The higher gelatinization temperature can also cause the components of the biodegradable plastic to be more homogeneous and create a more compact structure so that the resulting tensile strength value is greater [23]. It can be said that the tensile strength value presented in Figure 4. is by the theory obtained, namely, the greater the gelatinization temperature applied, the greater the tensile strength value of the biodegradable plastic produced. Another study also stated the effect of gelatinization pH on the manufacture of bioplastic from cassava peel starch with a gelatinization pH range of 4-7 at gelatinization temperature conditions of 70°C and 75°C [20]. The lowest tensile strength value was obtained at 0.87 MPa in a variable with a gelatinization pH of 4 and a gelatinization temperature of 70°C while the highest tensile strength value of 1.18 MPa was obtained in a variable with a gelatinization pH of 5 and a gelatinization temperature of 75°C. When compared with the lowest and highest tensile strength values obtained, namely 0.37 MPa and 0.77 MPa, it can be said that the value is lower.

3.5 Effect of STTP Level on Water Absorption of Biodegradable Plastic

The effect of variations in STPP levels on the water absorption of biodegradable plastic can be shown in the following graph:

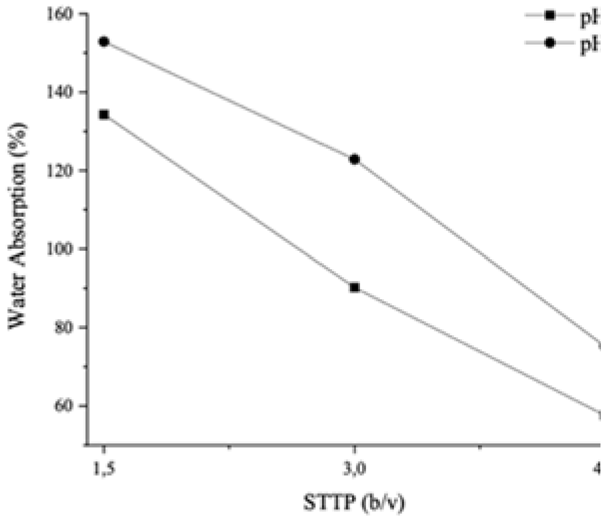

Figure 5: Effect of STPP levels on water absorption

From Figure 5. it can be seen that the increase in STPP levels added to the process of making biodegradable plastic will be followed by a decrease in water absorption in the resulting biodegradable plastic. The lowest water absorption capacity of 57.64% was found in the variable with an STPP content value of 4.5% (w/v) and gelatinization pH of 6 while the highest water absorption capacity of 152.88% was obtained in a variable with an STPP level of 1.5% (w/v) and the gelatinization pH was 8.

The addition of a cross-linking agent in the polymerization process of biodegradable plastic will cause the formation of a three-dimensional bond. Increasing levels of cross-linking agents will also result in smaller pore sizes formed on biodegradable plastics. This causes a decrease in the water capacity that can be absorbed by biodegradable plastic [24]. Increasing levels of cross-linking agents will affect the formation of polymer structures that are close together where the structure is not easy for water molecules to enter [25]. The recommended water absorption capacity of biodegradable plastic is around 12.41% -90.07% [19], so it can be concluded that the value of water absorption in the variables used is partly to the range of water absorption values used. It is recommended that the range of water absorption values in biodegradable plastic obtained is in the range of 57.64%-152.88%. Another work also discussed the effect of adding glycerol and citric acid as cross-linking agents to biodegradable plastics from cassava starch [26].It was explained that at the same gelatinization temperature and absorption time conditions, respectively 75°C and 80 hours where the variable with a citric acid content of 0% has a higher water absorption value of 32.5% while the variable with a citric acid content of 7.2% has an absorption value of 22.5% [26]. These results when compared with the water absorption value obtained, can be said that the value is greater.

3.6 Effect of STTP Level on Tensile Strength Biodegradable Plastic

The effect of variations in STPP levels on the tensile strength of biodegradable plastic can be shown in the following graph:

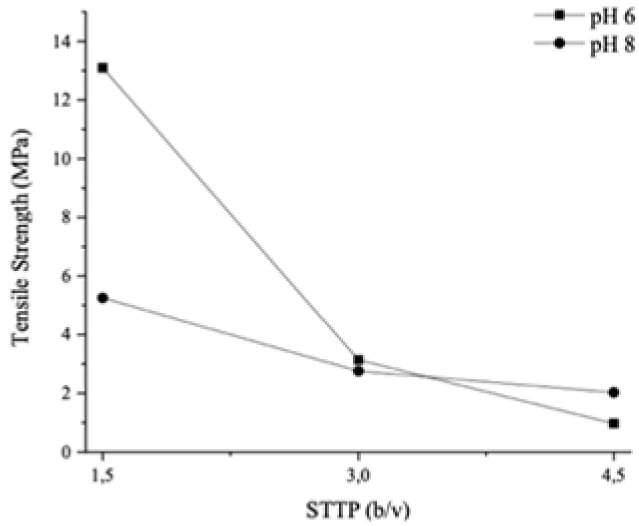

Figure 6: Effect of STPP content on tensile strength

From Figure 6. it can be seen that the amount of STPP added to the process of making biodegradable plastic will affect the tensile strength value of the biodegradable plastic produced under gelatinization pH conditions of 6 and 8. and gelatinization pH of 6, the lowest tensile strength value obtained was 0.97 MPa while the variable with STPP content added of 1.5% (w/v) and gelatinization pH of 6 obtained the highest tensile strength value, namely of 13.1 MPa.

The greater the level of STTP used, the greater the tensile strength value obtained. This is due to the nature of the cross-linking agent itself, which can form cross-links that can link starch molecules to each other [27]. Novitasari et al. (2016) [28] stated that the addition of a phosphate group from STPP replaces the hydroxyl group of starch by cross-linking the amylose and amylopectin molecules. This is what increases the cohesiveness of the polymer molecule, thereby increasing the tensile strength value. According to prior work, the formation of cross-links during the process of forming biodegradable plastics can increase interactions between amylose molecules to form stronger hydrogen bonds and result in the biodegradable plastic matrix being formed to be thicker, and denser [29]. In line with this explanation, the rigidity and strong hydrogen bonds between amylose molecules in biodegradable plastic can affect the tensile strength value [29]. From some of these explanations, it can be said that the STTP level will be directly proportional to the tensile strength value obtained. However, the data shown in Figure 6. shows a phenomenon that does not match the explanation obtained. Where the value of tensile strength decreased along with increasing levels of STTP at pH 6 and 8. Another study explained that the addition of an excess of the cross-linking agent can produce residues of the cross-linking agent itself so which can interfere with the interaction between starch molecules [30]. If the interaction between starch molecules is disturbed, it can lead to a weakening of the biodegradable plastic structure that is formed so that it can reduce the tensile strength value. In addition, the optimization of the manufacture of biodegradable plastic from cassava peel starch with the addition of 1% citric acid as a cross-linking agent of 5, 10, 15, and 20 ml, the lowest tensile strength value was 1.19 MPa in the variable with the large addition of citric acid [30]. One percent of 20 ml and the highest tensile strength value of 2.12 MPa was obtained in the variable with the addition of 1% citric acid of 5 ml. When compared with the lowest and highest tensile strength values of 0.97 MPa and 13.1 MPa respectively, it can be said that the value is smaller for the lowest tensile strength value and greater for the highest tensile strength value.

3.7 Effect of STTP Level on Biodegradability of Biodegradable Plastic

The effect of variations in STPP levels on the biodegradability of biodegradable plastics can be shown in the following Figure 7. It can be seen that the increase in STPP levels at pH 6 and 8 will be followed by a decrease in % biodegradability along with the time of burial of biodegradable plastics. Under conditions of gelatinization pH of 6 and burial time of 6 weeks, the variable with STTP level of 4.5% (w/v) obtained the lowest % biodegradability of 70% and the variable with STPP level of 1.5% (w/v). obtained the highest % biodegradability of 75%. Under conditions of gelatinization pH of 8 and burial time of 6 weeks, the variable with STTP level of 4.5% (w/v) obtained the lowest % biodegradability of 64%, and the variable with STPP level of 1.5% (w/v) obtained the highest % biodegradability of 78%.

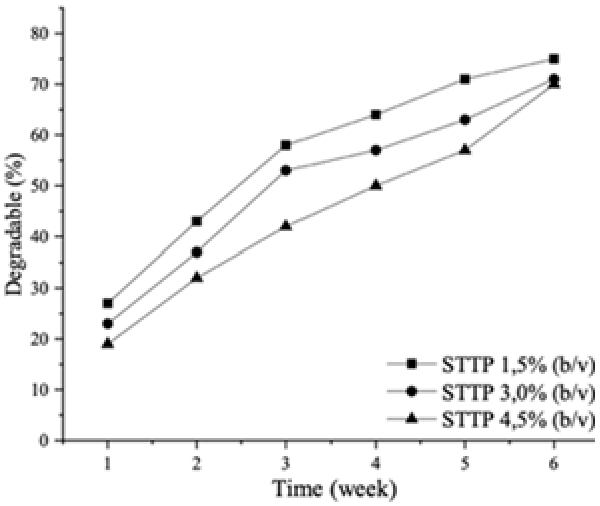

Figure 7: Effect of STPP concentration on biodegradability at pH 6

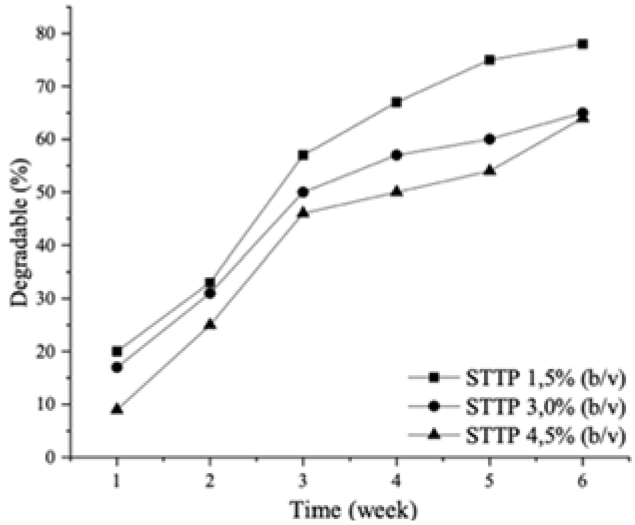

Figure 8: Effect of STPP concentration on biodegradability at pH 8

Previous study explained that the addition of a higher cross-linking agent (STPP) and plasticizer can affect the number of hydrocolloid and plasticizer structural bonds formed so that it will have an impact on the longer time it takes microbes to break the bonds of hydrocolloid structures and plasticizers [30]. In the degradation, process hydrolysis occurs which will result in the polymer matrix with more hydroxyl groups being decomposed more quickly into small pieces which will take longer to break down [31]. The addition of a higher cross-linking agent (STPP) and plasticizer can affect the number of hydrocolloids and plasticizer structural bonds formed will have an impact on the longer the time needed for microbes to break the bonds of hydrocolloid and plasticizer structures [30]. Another work also reported that the effect of adding glycerol and citric acid as a cross-linking agent to biodegradable plastic from cassava starch, it was stated that in two variables with the same gelatinization temperature and burial time, respectively of 30 days [26]. The variable with 0% citric acid content takes 12 days to decompose significantly while the variable with 7.2% citric acid content takes 18 days to decompose significantly. From this explanation, it can be concluded that the amount of % biodegradability obtained is following the theory obtained.

4. Conclusion

Increasing the temperature in the gelatinization process of biodegradable plastic from cassava starch can increase the tensile strength value and reduce water absorption in the resulting biodegradable plastic. Increasing the pH in the gelatinization process of biodegradable plastic from cassava starch can increase the tensile strength value and increase water absorption in the resulting biodegradable plastic. The addition of STTP levels in the process of making biodegradable plastic from cassava starch does not always increase the tensile strength value of the resulting biodegradable plastic. This can occur due to the residue of the cross-linking agent due to the addition of excess cross-linking agent so that it can interfere with the interaction between starch molecules and result in a decrease in the tensile strength value. The addition of STTP levels in the process of making biodegradable plastic from cassava starch can reduce water absorption and can increase the level of biodegradability of the resulting biodegradable plastic.

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