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Article

Characterization of Biodegradable Plastic Nata De Soya Using Glycerol and Palm Oil Addictive Substances

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Abstract. This research aimed to analyze the characterization of biodegradable plastic nata de soya (NDS). The addictive substance used to manufacture this plastic is a mixture of glycerol and palm oil with a saturated solution of arabic gum as an emulsifier. The volume variations are A (10:5:15) mL, B (10:10:30) mL, C (10:15:45) mL, control is (10 mL of 3% glycerol) and soaking time for NDS sheets with additives are 4, 6, and 8 days. The results showed that the highest degradation power of NDS is 87% in the variation of additive C with an immersion period of 8 days had a degree of crystallinity of 8.075%. The water resistance test was 318% in an immersion of NDS plastic with a variation of C for four days had a degree of crystallinity of 12.47%. The highest tensile strength value of plastic was 23.459 MPa in the composition of additive C with four days of immersion and had a degree of crystallinity of 13.172%.

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1. Introduction

Plastics play an essential role in every sector of the economy worldwide due to their wide use in agriculture, electronics, construction industry, transportation, health care, or the sport and leisure sector [1][2][3]. Plastic has the privilege of being quite strong, light, not rusty, thermoplastic, can be glued using heat, and can be labeled or printed with various creations [4]. Plastics manufacture various products, including defense materials, sanitary wares, tiles, plastic bottles, and other household items. Plastics are also used in food, medicine, detergent, and cosmetic packaging [2][5].

Global plastic production has experienced rapid growth since 1950. There has been a rapid increase in the production of commodity plastics. The worldwide production of commodity plastics exceeded 340 million metric tons produced globally in 2017, with around 46% of this coming from the packaging sector [6]. Most come from products with a short lifespan, typically around six months or less, than the building and construction sector, which has an average life of 35 years [7]. In the face of a current worldwide plastic production of almost 4 x 10^8 t annually, an enormous uptrend is noticed, especially in emerging and developing countries, characterized by boosting [8]. The rising amounts of plastics produced on our planet during the recent decades show in (**Figure 1**)[9].

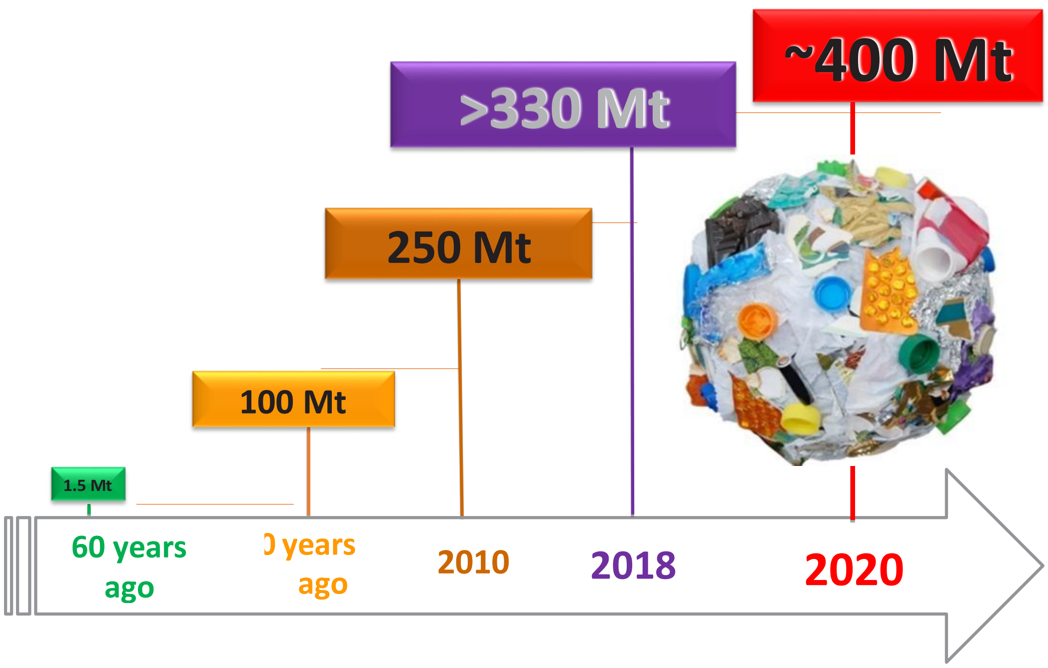

Figure 1. The rising amounts of global plastic production[9]

The use of plastic as a packaging tool can pollute the environment because of its non-biodegradable character [5][10][11][12]. The use of plastics as packaging harms health, causing cancer due to plastic monomers migrating into food if they directly contact hot or oily food. Given the dangers posed by plastic, it is necessary to have an alternative to plastic that is environmentally friendly and safe for health, such as biodegradable plastic [13][5]. Biodegradable plastics attract public attention as promising substitutes for non-degradable plastics that trigger severe plastic pollution. They are claimed to be environmentally harmless and biodegradable by microorganisms [14][15]. Biodegradable films are prepared from natural and easily renewable materials such as starch, cellulose, proteins, starch, corn, soybeans, and fats [16][17][18][19][20].

One of the renewable resources for plastic alternatives that are environmentally friendly and safe for health is nata-based biodegradable plastic. Nata-based biodegradable plastic is a plastic synthesized from nata, which is readily decomposed by the activity of microorganisms [13]. The nata used in this study is NDS which comes from tofu-making wastewater, which is one of the sources of environmental pollution [4][5][10]. NDS is a cellulose fiber on the surface of the tofu water medium from the metabolism of the bacterium *Acetobacter xylinum*, which converts glucose into cellulose. Cellulose formed is in the form of network-structured threads randomly oriented in a gel on the culture medium. Tofu liquid waste still contains nutrients that can be used as a growing medium for *Acetobacter xylinum* bacteria to produce cellulose and reduce pollution. Therefore, Nata has the potential to be further developed to be synthesized into biodegradable plastics.

NDS-based biodegradable plastic is not as dangerous as other synthetic plastics because it uses a safe plasticizer, namely glycerol. The addition of glycerol as a plasticizer serves to weaken the stiffness of bio-cellulose. In addition, this NDS cellulose-based biodegradable plastic is expected to retain water so that the stored product is not damaged. For this reason, it is necessary to add other hydrophobic additives to reduce the water permeability of non-polar plastics. For example, the addition of palm oil reduces the water permeability of biodegradable plastics [5][21][22].

Some researches on the process of producing biodegradable films from NDS are available in the literature. Such as chitosan and glycerol as plasticizers [22], oleic acid plasticizers [4]. However, not many studies have utilized tofu liquid waste to produce biodegradable films. The application of research results is expected to be for food packaging. Therefore, the purpose of this research is to characterize NDS-based biodegradable plastic with glycerol and palm oil as additives. The characterization in this study includes the Biodegradation, Water-resistance test, Tensile strength test, and Crystallinity test.

2. Experimental Section

2.1. NDS Sample Preparation

Sample preparation was carried out by providing 1 liter of filtered tofu liquid waste added 150 grams of sugar, 10 ml of 25% acetic acid, and 5 grams of urea. The mixture is brought to a boil and allowed to cool. All ingredients are mixed into a cooking pot then boiled until boiling. Then the mixture is poured into a Petri disk with a diameter of 9 cm. Then, tightly closed and allowed to stand for a day until completely cool. 1 liter of the cold sample was added to 150 mL of starter. Samples that already contain a starter are stored for 14 days in a calm and safe place.

2.2. NDS Sample Preparation

The manufacture of nata plastic is carried out by preparing nata sheets by pressing using a tensometer. Nata sheets were dried in an oven for ± 30 minutes, at a temperature of 70 C. Nata sheets were soaked with a volume variation of A (10:5:15) mL, B (10:10:30) mL, C (10:15:45) mL and for the control was 10 mL 3% glycerol and soaking time for nata sheets. With additives is 4, 6, and 8 days. Then, the nata plastic sheet is dried.

2.3. NDS Plastic Characteristic Testing

They were testing the characteristics of NDS plastics which includes the Biodegradation test (Soil burial test), Water-resistance test (Swelling test), Tensile strength test, and Crystallinity test. Biodegradation analysis of the nata plastic sheet was carried out by burying the nata plastic sheet in the soil with a size of 4x4 cm and a soil depth of 15 cm. The burial process was carried out for ten days. Before burial, the mass of plastic was weighed then buried in the ground for ten days, weighing intervals every two days. Tensile strength is done by cutting a plastic sheet cut to a length of 15 x 5 cm. The sheet is tested using a tensometer. Analysis of the degree of crystallinity of the NDS plastic sheet was carried out using XRD.

3. Results and Discussion

3.1. NDS Plastic Biodegradation Test

The biodegradation test is the most superficial characterization. Nevertheless, it is essential to know how much plastic is degradable [23]. The method used is to determine the biodegradation of plastic with a burial time of 10 days. The biodegradable film having –COO,–OH, and –COOH functional groups as flexible active sites can undergo degradation with high rates since these active groups enabling the film to bind on enzyme sites faster than the case with rigid biodegradable films [24]. The biodegradable film produced was easily decomposed. It is caused the raw material used was a raw material that easily interacted with water and microorganisms and was sensitive to physicochemical influence [25]. Data on the percentage of biodegradation of NDS plastic mass buried for ten days with variations A, B, and C, and 3% glycerol (as a control) during immersion variations 4, 6, and 8 days shown in (**Figure 2**).

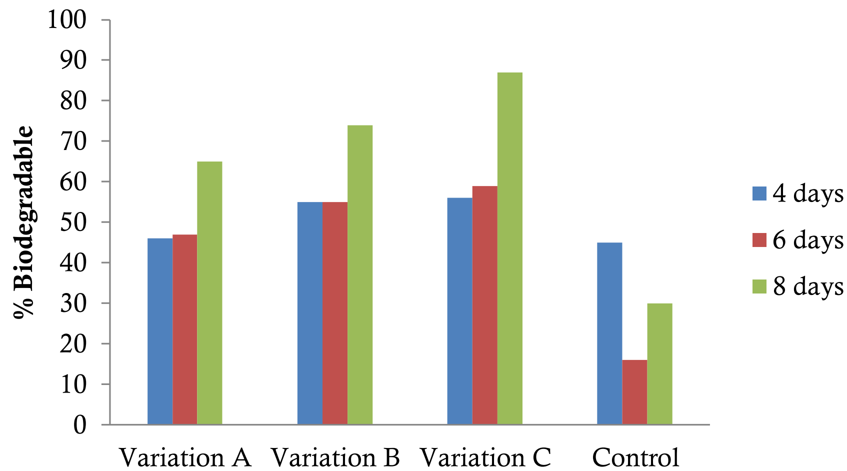

Figure 2. Graph of Relationship between Additive Composition of Variations A, B, C and 3% glycerol 10 mL (Control) for 4, 6, and 8 Days with Percent Degradation (degraded mass) of NDS Plastic with a Burial Time of 10 Days

It can be observed that each of the buried NDS plastic samples has a pretty good degradation power. The longer the immersion time with additives, the higher the plastic degradation power. NDS plastic soaked in additives for eight days had the most negligible residual mass after ten days, in variations A, B, and C. The NDS plastic sample was a control, which was only soaked with 3% glycerol. The immersion time did not show identical results. It shows that the effect of variations in the mixture of additives is large enough for the plastic to be readily degraded (**Figure 2**).

The longer the immersion time, the greater the percentage of the plastic mass degraded [4]. In addition, the greater the volume of additives added to the NDS plastic, the greater the mass of the degraded plastic. It can be seen in plastic soaked with variation C, which has the most significant percentage of decomposed mass than plastic soaked in other variations. NDS plastic with additives soaking variation C for eight days showed the highest percentage of biodegradation, which was 87%. This percentage is influenced by the addition of hydrophilic glycerol additives [5][22]. This NDS-based biodegradable plastic with glycerol, palm oil, and arabic gum additives has a semi-crystalline structure. It is indicated by the presence of a pointed peak, namely a crystalline peak and a blunt, amorphous one, with the highest degree of crystallinity in the plastic sample. The biodegradation test results were 8.075%.

3.2. NDS Plastic Water Resistance Test

The water resistance test is carried out to determine how much the plastic absorbs into water. Plastic is expected to absorb only a little because the ultimate purpose is for food packaging. One of the additives added to plastic is palm oil. Palm oil was chosen because of its high content of saturated and unsaturated fatty acids. The function of adding it to plastic is to reduce water entering the plastic because it is hydrophobic [5][21][22]. Data regarding the relationship between additive composition and control with water absorbed by NDS plastic can be seen in (**Figure 3**).

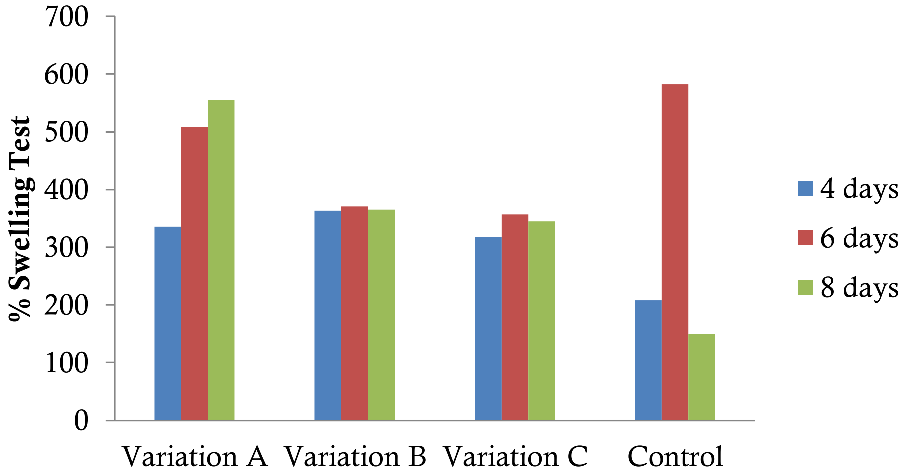

Figure 3. Graph of Relationship between Additives Composition and Control with Percentage of Water Absorbed by NDS Plastic

The results of the water-resistance test, as can be seen in (**Figure 3**). it shows that the larger the volume of palm oil added to the additive mixture, the smaller the percentage of water absorbed by the plastic. It is because the addition of lipids can reduce water permeability. After all, lipids can increase the distance traveled by water molecules that are absorbed by the plastic [5][21][22]. The duration of plastic immersion shows the percentage of water that rises on the 6th day of immersion and falls back on the 8th day. The longer the immersion, the lower the hydrogen bonds between glycerol and cellulose in the plastic. NDS-based biodegradable plastic has the most significant water resistance (most minor percent water resistance), 318% by immersion treatment with variation C additive with a composition of 10 mL glycerol, 15 mL palm oil, and 45 mL arabic gum for four days (**Figure 3**).

In control, it was only soaked with 3% glycerol as much as 10 mL. It turned out to have a minor percent water resistance (more excellent than plastic soaked with a mixture of 3% glycerol additives, palm oil, and gum arabic). It was 150% during immersion eight days. In the mixture of additives, arabic gum is also added as an emulsifier which is hydrophilic arabic gum (**Figure 3**). The water resistance of a molecule is related to the basic properties of its constituent molecules. The cellulose material used in this study is hydrophilic; that is, it likes water. Glycerol and arabic gum also have hydrophilic properties, so that it further adds to the hydrophilic properties of this plastic.

In contrast, the interaction between additives, especially palm oil and plastics, occurs only by physical adsorption. The additives are not effective in reducing the swelling value of cellulose plastics [26]. Biodegradable-based plastics have a semi-crystalline structure, with a degree of crystallinity in the plastic sample. The highest water resistance test results were 12.47%.

3.3. NDS Plastic Tensile Strength Test

The plastic base material influences mechanical properties and additives added to the plastic. Tensile strength is the maximum tensile force that the film can withstand during the measurement until it breaks. Thus, the tensile strength of a film greatly affects the quality of the film[27]. The tensile strength test aims to determine the value of the tensile strength of the synthesized polymer. The relationship between the tensile strength of NDS plastic and the composition of additives and controls can be seen in (**Figure 4**).

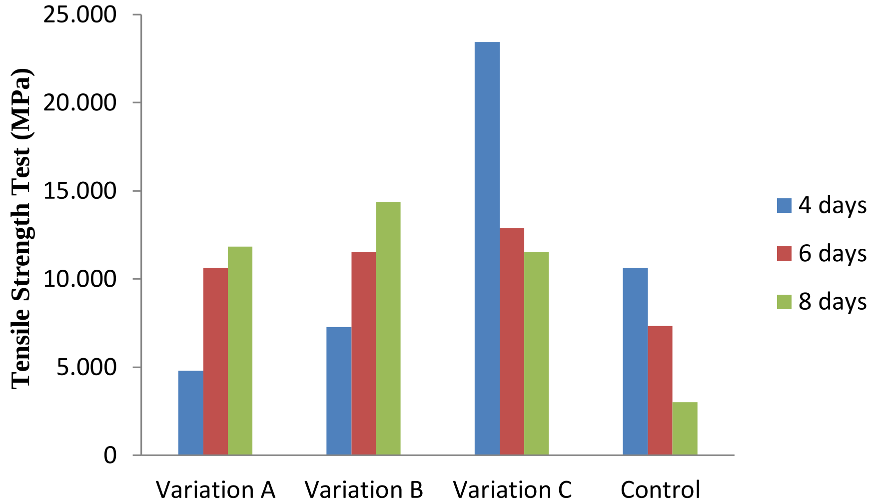

Figure 4. Graph of Relationship between Additive and Control Substance Composition with NDS Plastic Tensile Strength Value

This NDS-based biodegradable plastic with the addition of glycerol, palm oil, and Arabic gum additives has a pretty good tensile strength, with the best results being in plastic soaked with a mixture of C variation additives with a composition of 10 mL glycerol, 15 mL palm oil and 45 mL arabic gum for four days with a tensile strength of 23.459 MPa (**Figure 4**). The plasticizer in the edible film affects the physical and mechanical properties of the edible film. The addition of 3% glycerol is used as a plasticizer that increases the flexibility of NDS plastics.

Based on the tensile strength value results on the control plastic (which only added 3% glycerol), there was a decrease in the tensile strength value. The longer it was soaked, the lower the tensile strength. The addition of glycerol decreases the plastic tensile strength of NDS because the plasticizer reduces the inter-chain force and decreases the tensile strength [28]. Plasticizers in high amounts can cause the material to become elastic to reduce the material's tensile strength [29]. Variation C of biodegradable plastic has a semi-crystalline structure. The highest tensile strength test results were 13,171%.

3.4. NDS Plastic Crystallinity Test Results

A plastic (polymer) crystallinity can be determined using an X-Ray Diffraction (XRD). The plastic samples produced are semi-crystalline, which means they have amorphous or crystalline parts. NDS-based biodegradable plastic with the addition of glycerol additive has a semi-crystalline structure indicated by the presence of sharp peaks, namely crystalline peaks, and the presence of blunt ones, namely amorphous ones. The relationship between the duration of immersion of NDS plastic with 3% glycerol and the degree of crystallinity can be seen in (**Figure 5**).

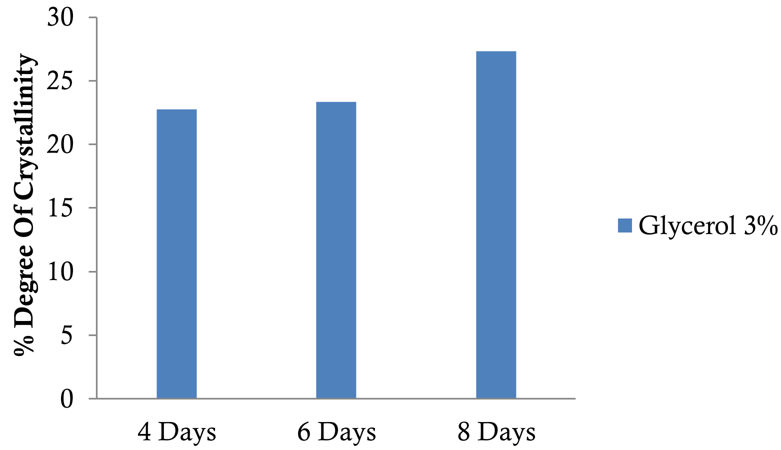

Figure 5. Graph of the relationship between the duration of immersion of NDS plastic with 3% glycerol and its degree of crystallinity

The best biodegradation test results, the crystalline degree is poor, namely 8.075%. It indicates the number of amorphous structures, which causes this plastic to have the highest degradation power because the more amorphous it is. Thus, it is easier to degrade soil due to the weak bond and irregular structures easily damaged by soil microbial activity. Microorganisms attack the less regular or amorphous parts more quickly than attack the crystalline parts [30]. Plastics from the best results of the water-resistance test have a reasonably large degree of crystalline, namely 12.47%, which means that amorphous rather than crystalline structures dominate this plastic. This plastic has a higher degree of crystallinity and has the best result of the biodegradation test. It causes this plastic to have the lowest percentage of water resistance than other plastics because the higher the crystallinity, the tighter the cellulose structure so that water is difficult to penetrate it.

Plastics from the best tensile strength test results showed a degree of crystallinity of 13.172%, which means that the amorphous structure was 86.828%. It turns out that this plastic is still quite strong, with a tensile strength value of 23.459 MPa. In general, the results of the crystallinity of NDS plastics prove that this plastic is quite strong. The highest degree of crystallinity is in plastics soaked with 3% glycerol additive for eight days, 27.334%. Compared to commercial plastic, HDPE (High-Density Polyethylene) has a crystallinity percent of 62.95%-70.34% [31]. This plastic is still not strong enough for packaging.

4. Conclusion

The results showed that, to begin with, the highest degradation power of NDS is 87% in the variation of additive C with an immersion period of 8 days had a degree of crystallinity of 8.075%. In addition, the water-resistance test was 318% in an immersion of NDS plastic with a variation of C for four days had a degree of crystallinity of 12.47%. Furthermore, plastic's highest tensile strength value was 23.459 MPa in the composition of additive C with four days of immersion and had a degree of crystallinity of 13.172%.

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