

Green synthesis of cellulose diacetate from pineapple leaf fibers to improve the mechanical properties of edible plastics made from tapioca starch

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ABSTRACT

The purpose of research is to improve the characterization of mechanical properties in edible plastic made from tapioca starch with the addition of cellulose-diacetate from pineapple fiber as a substitute of plastic-based packaging. Edible plastic was made with hydrolysis method with the comparison of 50 g of tapioca starch in 50 ml solvent. pH value used in solvent was 7. The composition of edible plastic were 7.5 g of hydrolysis product, 100 ml aquades, 45 ml of 96% ethanol, 1.2 ml glycerol and cellulose diacetate from pineapple fiber varies from 0,2%, 0,4%, 0,6%, 0,8%, 1%. In general, the plastics produced were stable enough with the power of stretch between 89,33-115,83 kgf/cm² and elasticity between 49,6-60%. This edible plastic made from pineapple fiber could react to form some new clusters which are CO-CH₂ and P-H.

Key words : Edible plastic, Tapioca, Pineapple fiber, Tensile strength, Elasticity and FT-IR, Green product

Introduction

Plastics are made from carbon polymers which are generally derived from crude oil. Due to its practical and durable functions, plastic is often used as the base material for various tools. Behind its practicality, plastic has a big problem for the environment because its waste is very difficult to decompose. At least it will take up to a hundred years to naturally break down plastic. The plastic recycling process is not an easy thing. What's more, the ability of the recycling process is not balanced with the amount of plastic waste that is produced every day. To overcome this, the use of environmentally friendly plastics continues to be developed. Eco-friendly plastics are trying to meet the needs of plastics that are du-

able, but can be more easily biodegradable (Wafiroh, 2004)..

In general, there are three types of environmentally friendly plastics that can be used, namely bioplastics, biodegradable plastics, and eco plastics. Bioplastic is a type of environmentally friendly plastic made from natural materials. Biodegradable plastic is a type of plastic that is easily biodegradable. Unlike bioplastics which are made from renewable natural materials, biodegradable plastics can be made from traditional petrochemicals with additives that make the waste easier to decompose (Mariana, 2007). Meanwhile, eco plastics are plastics that are produced from the recycling of plastic materials from petrochemicals for reuse. The recycled plastic will not be produced into the same

type of product, but rather a type of plastic with a lower quality (Wijayanti, 2016)

Bioplastics are a type of biopolymers known as natural polymers or green polymers. Edible plastics are biopolymers which have properties that can be consumed by living things. Edible plastics are made from natural materials such as polysaccharides, proteins, polystyrenes, fats and their derivatives. This causes edible plastics to quickly degrade naturally and is very effective in replacing conventional plastics in short life (Santoso, 2007).

Edible plastics can be made using natural materials, for example polysaccharide- $C_6H_{10}O_5$ (Surdia, 2005). Starch is a form of polysaccharide that can be obtained from various types of plants such as rice, corn, cassava and sweet potato. Starch derived from cassava (tapioca flour) is more developed. This is because the amount of tapioca flour in Indonesia is abundant and it is not a staple food material for Indonesian people, such as rice (Kusandini, 2016).

Based on research conducted by Kusandini (2008), edible plastic made by dissolving tapioca starch in acetate solution or ammonia solution has several drawbacks. In this study, edible plastics have a thickness between 38.34 - 42.32 kgf /cm² and elongation between 3.1 - 4.5%. If the plastic sheet is too thick it can block gas exchange, causing a buildup of ethanol which can damage the product's taste image. Thus it is necessary to do further research by adding filler to the plastic, which aims to improve the mechanical properties of the plastic, reduce the cost per unit volume, increase strength and also improve the properties of the product. Pineapple fiber was chosen as a filler with several advantages including 81% fiber content so that it can be synthesized into cellulose diacetate while also increasing the economic value of pineapple leaves in an effort to reduce waste buildup (Imamah, 2018).

Materials and Methods

The materials used in the research were pineapple leaves, tapioca starch and chemicals including sodium acetate (CH_3COONa), acetic acid (CH_3COOH), distilled water, 96% ethanol, $Ca(OH)_2$, NaOH, glacial acetic acid (CH_3COOH), acetic acid anhydride ($(CH_3CO)_2O$), concentrated sulfuric acid (H_2SO_4) and glycerol. All of these chemicals were obtained from Sigma Aldrich.

The synthesis of edible plastics in this study can be divided into two stages, namely the synthesis of

cellulose diacetate from pineapple fibers and synthesis of edible plastic from tapioca which is given pineapple cellulose diacetate fiber. The first stage, cleaned pineapple fibers then soaked in distilled water for 2 weeks until the pineapple skin is soft and the fibers separate. The pineapple fiber is then washed thoroughly and then dried in the open air (Wijanji, 2006). Furthermore, 20 g of pineapple fiber was added with $Ca(OH)_2$ 2.5% (w/v) 150 ml and soaked for 3 days. After that it was washed with distilled water and then put into a round bottom flask which was previously filled with 300 mL of 17.5% (w/v) NaOH solution, then refluxed for 4 hours (Puspasari, 2008).

After chilling, the pineapple fibers were washed until they were NaOH-free and then blended and molded into pulp sheets and dried in an oven at 60 °C. The pineapple fiber pulp was added with glacial acetic acid and shaken at 40 °C for 1 hour. After that, a mixture of 60 ml glacial acetic acid and 0.5 mL concentrated sulfuric acid was added and then shaken again for 45 minutes at the same temperature. The mixture was cooled to a temperature of 18 °C and acetic anhydride was added at 40 °C. The mixture was added with 67% (w/v) acetic acid as much as 30 mL dropwise for 3 hours at 40 °C and then shaken. Furthermore, it is hydrolyzed for 15 hours and precipitated by adding distilled water drop by drop and stirring to obtain a powder in the form of a precipitate. The precipitate is filtered and washed until it is neutral. The precipitate is dried in an oven at 60–70°C and filtered using a micro filter.

The second stage, making edible plastics is made from mixing tapioca starch and solvent manually in a container (beaker glass) with a composition of 50 grams of tapioca starch and 50 ml of solvent. Then the mixing is done using a heater equipped with a stirrer. The edible plastic that will be made is a composition of 7.5 g of the result of mixing a solution that has been dried and refined, 100 ml of distilled water, 45 ml of 96% ethanol, 1.2 ml of glycerol and variations of 0%, 0.2%, 0.4% , 0.6%, 0.8%, 1% cellulose diacetate from powdered pineapple fiber. Furthermore, mixing is carried out using a heater and magnetic stirrer with a temperature of less than 70 °C and a rotation speed of 60 rpm until the mixture thickens. The resulting mixture was then printed on plexiglass and cooled to room temperature. The resulting edible plastic is formed into a thin sheet in such a way that it suits your needs

Results and Discussion

The cellulose diacetate obtained from pineapple leaves was analyzed for its functional groups using FTIR, which results were shown in Figure 1a. Compared with standard cellulose diacetate (Figure 1b) can be obtained as follows. The synthesized cellulose diacetate had a wide and sharp O-H peak at the wave number 3492.86 cm^{-1} . Whereas in standard cellulose the wave number is 3491.4 cm^{-1} . In the synthesized cellulose diacetate there is a peak at the wave number 1747.93 cm^{-1} which indicates the presence of a carbonyl group (C=O), while in standard cellulose at the number 1743.2 cm^{-1} . In the synthesized cellulose diacetate there is a peak at the wave number 1239.03 cm^{-1} which indicates the presence of a C-O ester bond, while in standard cellulose there is a wave number of 1242 cm^{-1} . In the synthesized cellulose diacetate, there is a sharp peak at the wave number 1046.68 cm^{-1} which indicates the presence of C-O glycoside bonds and C-O bonds of pyranose bonds, while in standard cellulose the wave number is 1042.8 cm^{-1} .

This microstructure analysis to determine morphology, structure, and various forms including grain, phase, attached phase, and attached particles was carried out using Scanning Electron Microscopy as shown in Figure 2. From the SEM photos it appears that both compositions have bubbles. This is due to the material mixing process carried out in open air. The results of edible plastic without mixing cellulose diacetate (Figure 2a) appear smooth and not bright, whereas in the sample B edible plastic from mixing starch with the addition of cellulose diacetate from pineapple fibers (Figure 2b) appears to have a smooth surface but the bubbles are very

visible clumps of filler particles. This is due to the imperfect mixing process, which affects the mechanical strength of the edible that is formed. This is the reason why the addition of cellulose diacetate is 0.2%. has the lowest tensile strength of edible plastic.

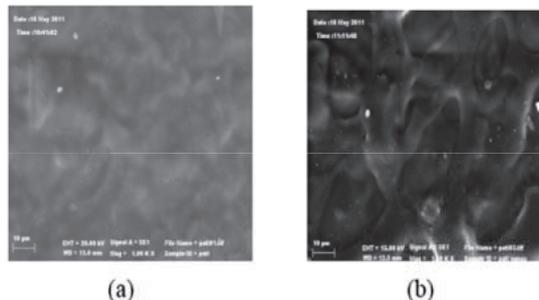


Fig. 2. SEM of (a) Starch tapioca plastic (b) starch tapioca – cellulose plastics

From Figure 3, it can be seen that the tensile strength of plastic edibles with the addition of cellulose diacetate from pineapple fibers is higher than that of plastic edibles using only tapioca starch. The addition of 1% cellulose diacetate had the highest tensile strength value, namely 115.8 kgf/cm^2 ,

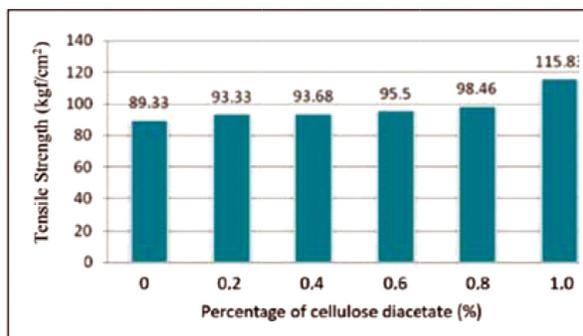


Fig. 3. The tensile strength of samples

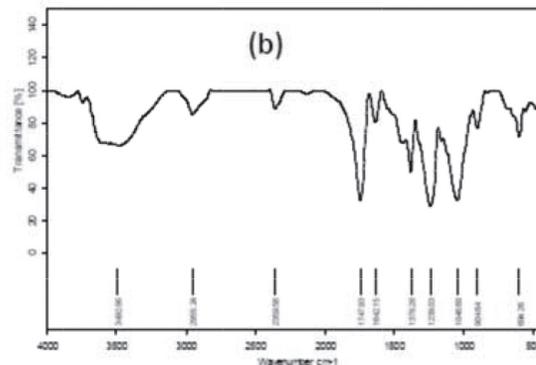
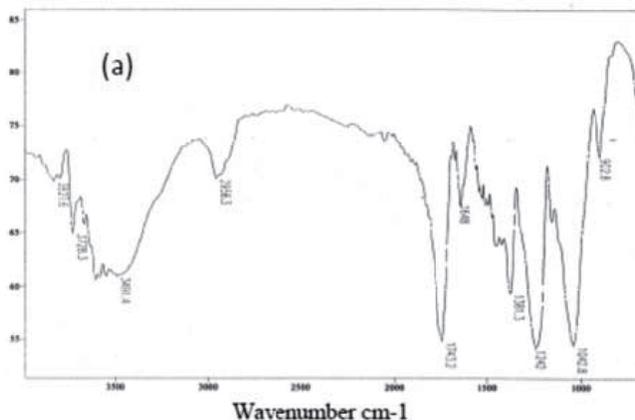


Fig. 1. FTIR spectrum of (a) Standard cellulose (b) cellulose isolated from pineapple leaf fibers

whereas without the addition of cellulose diacetate, the plastic edible tensile strength value was relatively low, namely 89.3 kgf /cm². Likewise in terms of the percentage of elongation, the effect of addition The high cellulose diacetate resulted in the plastic being more elastic so that the elongation value reached 60% while the edible plastic made from starch-tapioca had lower elongation, namely 49.6% (Figure 4). The difference in values obtained in the plastic edible is due to differences in mixing.

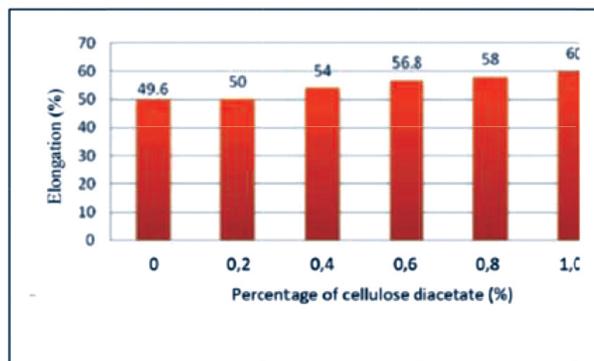


Fig. 4. The elongation of samples

In general, edible plastic has sufficient stability with a minimum thickness of 41.72 μ m, tensile strength values between 27.01 - 217.7 kgf/cm², elongation between 2.55 - 62.89 %. Judging from the stability standard, it can be concluded that the edible plastic produced in this study is quite stable with tensile strength values between 89.33 - 115.83 kgf/cm², and elongation between 49,6 – 60 %.

Conclusion

The three conclusions of this study are as follows. Firstly, the addition of cellulose diacetate from pineapple fibers used in the starch hydrolysis process with acetate solvent has an effect on the mechanical

properties of edible plastics. Secondly, the edible plastic produced by the addition of cellulose diacetate from pineapple fibers with the addition of 1% has the best mechanical properties. Finally, in general the edible plastic produced is quite stable with a tensile strength value between 89.33 - 115.83 kgf /cm², and an elongation between 49.6 - 60% and is environmentally friendly.

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