



SYNTHESIS OF SMART PACKAGING FROM CELLULOSE ACETATE WITH THE ADDITION OF AgNO_3 AS AN ANTIBACTERIAL SUBSTANCE

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ABSTRACT

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Synthesis of smart packaging from cellulose acetate with the addition of AgNO_3 as an antibacterial substance has been investigated. The purpose of this study is to fabricate smart packaging from cellulose acetate with the addition of AgNO_3 as an antibacterial substance. The method used was solution casting. In addition, this study was also characterizing the smart packaging, they included tensile strength and elongation percent test, antibacterial test, morphology analysis using SEM, and biodegradability test. The plastics obtained were clearly yellow, not easily damaged, a little thick, and the smooth surface and slightly bubbly. The largest tensile strength of plastic was 0.0661 MPa, it found in sample No. 2, while the smallest tensile strength was 0.027 MPa, it found in sample No. 1. The greatest elongation value at break was 5% found in sample No. 4, while the smallest elongation value at break was 3.5% in sample No. 3. The result of antibacterial test reported that the freshest mangoes were shown by samples No. 1 and No. 2; while the fast-rotting mangoes were shown in the sample No. 3 and No. 4. The results of the SEM test showed the presence of nano-sized particles that spreaded in the plastic body. The greatest degradability degree were sample No. 4 about 0.0009 g/day with the percentage of mass loss about 17.38%. Furthermore, cellulose acetate can be used as a basic material for making plastics. The addition of AgNO_3 in plastic synthesis can help to delay the process of fruit spoilage caused by bacteria.

Keywords: AgNO_3 , antibacterial substance, biodegradability, cellulose acetate, smart packaging

1. Introduction

Plastic production has increased rapidly over the past 70 years, from almost 0.5 million ton in 1950 to more than 365 million ton in 2016 worldwide, and almost 66 million ton produced in Europe [1] [2]. Plastics contain several additives, including stabilizers, pigments, and toxic chemical components such as flame retardants, antioxidants, and blowing agents [3] [4] [5]. Synthetic plastic is a petroleum-based material. This substance is obtained through the refining of crude oil, this process separates and fractionates heavy crude oil into lighter components known as segments. This segment is a mixture of polymer hydrocarbon chains, one of which is naphtha, which is an important component needed to produce monomers such as ethylene, propylene and styrene to produce plastic. These monomers form plastics through polyaddition and/or polycondensation assisted by specific catalysts [6]. However, this conversion produces pollutants and greenhouse gases such as carbon dioxide (CO_2), thus contributing to environmental pollution and global warming [7]. Plastic waste is non-organic waste taking about twenty years to decompose in the soil. It can cause environmental problems, such as water pollution and toxic to organisms in the soil [8]. Plastics commonly used for food packaging are synthetic plastics made from

petroleum. Besides not being able to decompose in the soil, this plastic waste is also ineffective for burning because it will produce carbon emissions resulting air pollution [9].

One of the uses of plastic is as fruit packaging but the plastics circulating in the market are less able to maintain resistance to fruit rot caused by bacterial growth. Therefore, to increase durability and to reduce the impact on the environmental pollution. It is crucial to create an easily biodegradable plastics which also can gradually maintain the bacterial growth [7].

The problem gave rise to an idea to research the manufacture of plastic from the other sources that are eco- friendly as it can be an effective solution. One of the solutions is smart packaging, it is a new concept in food packaging as it can provide several functions that are not found in the ordinary packaging. The smart packaging can be utilized in oxygen absorption, ethanol emission, and antimicrobial activity [7]. One solution is smart packaging, which is a new concept in food packaging because it can provide several functions that are not found in ordinary packaging. Smart packaging can be utilized in oxygen absorption, ethanol emission, and antimicrobial activity. High oxygen content in food can increase the rate of oxidation and increase microbial growth so that the quality of the food will decrease. Therefore, food packaging needs to add substances that have the potential to absorb oxygen. Apart from that, smart packaging also plays a role in ethanol emissions. Ethanol is an anti-microbial substance, so the release of ethanol by smart packaging can increase food storage capacity [10] [11]. The main focus of this study is food packaging as an antimicrobial.

An antimicrobial packaging is a form of smart packaging that can increase the shelf life of products and can prevent, can kill or can reduce the growth of pathogenic bacteria in packaged foods and packaging materials [12] [13]. To control the growth of spoilage-causing microorganisms on fruits such as *Escherichia coli*, *Staphylococcus aureus*, *Listeria monocytogenes*, *Bacillus cereus*, *Campylobacter jejuni*, *Clostridium botulinum*, *C. perfringens*, *Cronobacter sakazakii*, *Salmonella spp.*, *Shigella spp.*, *Vibrio spp.*, and *Yersinia enterocolitica* [14] [15], it can be worked by using packaging containing antimicrobial compound. Several studies that have been carried out include using silver nanoparticles combined with polystyrene nanocomposites as an antibacterial for Gram positive and negative bacteria and yeast [16]; utilizing a combination of silver and gold nanoparticles combined with carboxymethyl as an antimicrobial against bacteria (*S. aureus*, *P. aeruginosa*), fungi (*Aspergillus niger*) and yeast (*Candida albicans*) [17]; using silver nanoparticles combined with carboxymethyl nanocomposites which are able to inhibit the growth of *E. coli* and *Bacillus subtilis* [18]; utilizing silver nanoparticles combined with carboxymethyl which has the potential to inhibit bacterial growth [19].

Based on the data, the study utilized cellulose acetate as the basic material for fabricating bioplastics. The synthesized bioplastics were added with AgNO_3 as an antimicrobial and pluronic acid as a pore-forming material. As far as we were concerned, it has taken the proposed approach yet.

2. Materials and Methods

2.1 Materials

The materials used in the manufacture of smart packaging were cellulose acetate (Merck), pluronic acid (Merck), AgNO_3 (Merck), polyethylene glycol (PEG), acetone, and distilled water. The tools used in the study were glassware, analytical balance, ultrasonication, and SEM (Scanning Electron Microscope).

2.2 Methods

2.2.1 Fabrication of smart packaging with solution casting method

Fabrication of smart packaging with the solution casting method was done with two variations. The first variation, above 30 g and 40 g of cellulose acetate were dissolved with 60 mL and 80 mL acetone, respectively. Then, the cellulose acetate solutions were added with 5 g and 10 g of PEG, respectively. Furthermore, the mixtures were added 1.0 g of pluronic acid. Then the mixtures were sonicated at 920 rpm using sonication. After that, the sonicated mixture was added 0.5 g of AgNO_3 and they were homogenized. The mixtures were poured into a glass mold and smoothed using a glass spatula. Afterward, the mixture was dried at room temperature. After drying, they were soaked in water.

2.2.2 Tensile Strength Test and Elongation Percent Test

The tensile strength test and elongation percent test were carried out using the COM-TEN testing Machine 95T Series. Samples were tested according to ASTM D638 for plastic polymers.

2.2.3 Antibacterial Test

Smart packaging obtained could be applied as a fruit wrapper. The fruit resistance test could be carried out by comparing the use of the smart packaging and the ordinary plastic for packaging fruits within a certain period of time. If plastics can provide a longer resistance to fruit rot so the plastics are good [20].

2.2.4 Morphology analysis using SEM (Scanning Electrone Microscope)

The Morphological characteristics of smart packaging were analyzed using SEM. The smart packaging analyzed was a good sample from the more variations carried out [20].

2.2.5 Biodegradability test

The biodegradability test was carried out to determine the time required for smart packaging to degrade. The biodegradability test was carried out using the soil burial test technique. Smart packaging was buried in the ground and left exposed to the open air without being covered with glass. The observations on samples were carried out for seven days with observations once a day. The observations were made by looking at changes in the weight of the smart packaging film [20]. The rate of degradability can be calculated by the following calculation:

$$\text{Rate of degradability} = \frac{W_1(\text{g}) - W_2(\text{g})}{7 \text{ hari}} \quad (1)$$

Which W_1 is weight sample before burial and W_2 is weight sample after burial.

3. Result and Discussion

This research synthesizes smart packaging made from cellulose acetate so that this packaging can be degraded in the environment. Apart from that, with the addition of AgNO_3 as antibacterial agent, this packaging was expected to last longer compared to other commercial packaging.

3.1 The smart packaging with solution casting method

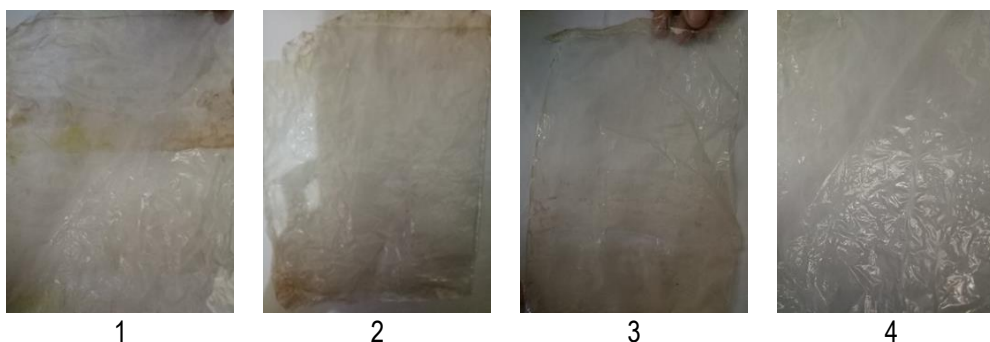


Figure 1. Smart packaging synthesized

There were four plastic samples that have been made with casting method. The principle of this method is to dissolve a polymer in a solvent containing salt particles, which are evenly distributed to a certain size, and the solution is then placed into a container according to the desired shape, then the mixture is allowed to solidify [21] [22]. The plastic produced comes from biodegradable materials, in this case cellulose acetate. This material is containing nutrients needed by microorganisms in the soil. Microorganisms utilize cellulose as a source of carbon which acts as material for cell -forming. Through the process of metabolism cellulose using enzymatic reaction can be converted into simple molecules (CO_2), water (H_2O), and other organic acid [23] [24]. These compounds are certainly not harmful to environment. In general, Figure 1 represented the four samples. The plastics obtained generally were clearly yellow, not easily damaged, a little thick, and the smooth surface and slightly bubbly. Based on the report [25], plastics synthesized from jackfruit seed starch produced a clear brownish-brown plastic, easily damaged, the slightly rough surface and slightly thick, and bubbly. The bubbles produced were still more numerous than the bubbles produced in this study. The difference in these results could be seen from the basic compound, this study used pure cellulose, while [7] used starch from natural compound. According to [26], bioplastics synthesized from starch were not resistant to heat, were not resistant to microorganisms and air, and had low mechanical properties. Therefore, plastics made from starch are easily damaged. That is because that starch is hydrophilic so that it affects its stability and mechanical properties. As a result, the plastic has a short shelf life so it is less optimal because water vapor and microbes that enter through the film will damage food [27].

3.2 The Tensile strength forces

The mechanical properties of plastics are the main characteristics that determine their quality. The mechanical properties of plastics include hardness, compressive strength, elongation at break, tear resistance, abrasion resistance, and temperature resistance. In this study, the mechanical properties tested were compressive strength and elongation at

break. Compressive strength is an indication to quantify maximum number of force required to make the plastic damaged or torn while the elongation at break is an indication of the length of the plastic that can be stretched until it breaks [28].

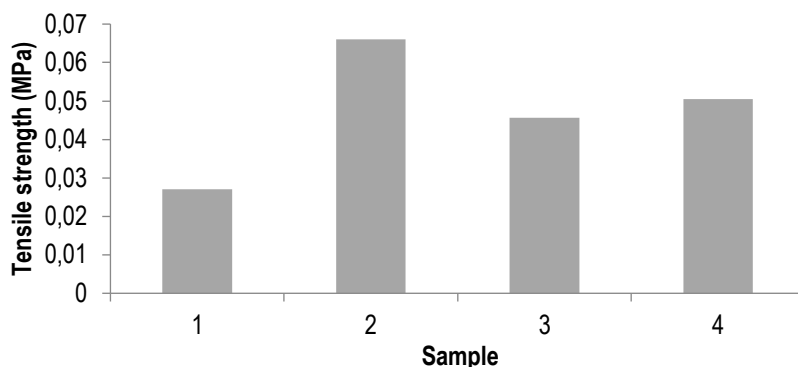


Figure 2. Value of the tensile strength power for each sample

The compressive strength values of the four synthesized samples showed an increasing value along with the increase in the amount of cellulose acetate, polyethylene glycol (PEG), and pluronic added. The largest compressive strength of plastic was 0.0661 MPa, it found in the second sample (sample 2) with a composition of cellulose acetate 40 g, PEG 10 g, pluronic 1 g. The smallest compressive strength was 0.027 MPa, it found in the first sample (Sample 1) with a composition of cellulose acetate 30 g, PEG 5 g, pluronic 1 g. It is caused by the amount of cellulose acetate, the more cellulose used the better the compressive strength. The addition of cellulose can improve polymer adhesion interactions and strengthen cohesion [29]. The cellulose acetate used has also been previously converted into nano form so that it can increase the degree of crystallinity and make interactions between polymers increase and indicate a more regular structure so that the compressive strength will increase [30]. The increase in compressive strength was also caused by an increase in the amount of PEG used due to the function of PEG as a plasticizing agent. The plasticizer used will position itself between the polymer molecules so as to interfere with polymer-polymer interactions and increase the flexibility of the film and also because of the physical properties of PEG which will be flexible when in the form of a solution [31]. Sample 4 has tensile strength compared to samples 1 and 3 because the amount of cellulose acetate in sample 4 was greater than samples 1 and 3. Meanwhile, sample 3 has a different PEG and plurony composition from sample 1 even though the acetate composition is the same. This has a significant tensile strength effect.

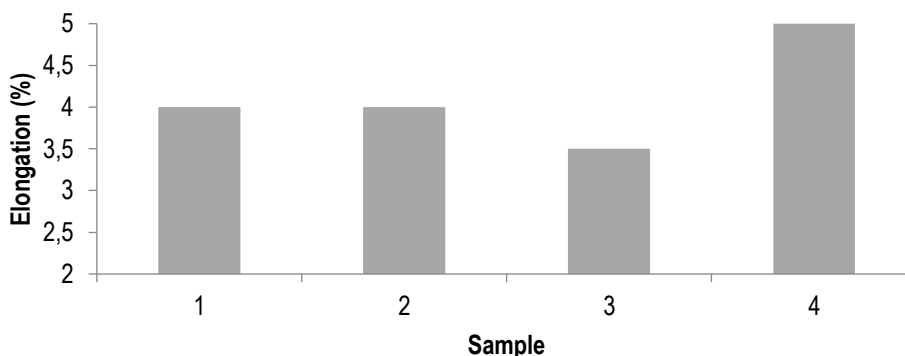


Figure 3. Value of elongation percent for each sample

The greatest elongation value at break was 5% found in sample 4 with the composition of cellulose acetate 40 g, PEG 10 g, pluronic 1.5 g. The smallest elongation value at break was 3.5% in sample 3 with the composition of cellulose acetate 30 g, PEG 10 g, pluronic 1.5 g. The value of elongation at break that did not match the compressive strength of the synthesized film could be caused by the amount of pluronic added were too much and the amount of plasticizer (PEG) used were small. Therefore, sample 3 had a low elongation at break. The plasticizer used aimed to provide flexibility to the synthesized film, but the presence of excess pluronic could reduce its flexibility because the use of pluronic as a pore-forming agent could interfere with the interaction between polymer molecules and PEG. [32] researched on PS-Pluronic membranes, the best pluronic solubility was obtained at 50 °C, but the use of this temperature in this study could cause rapid evaporation of the acetone solvent used. Inappropriate elongation values could also be caused by the presence of pluronics that had not completely dissolved in the printed film. The elongation value for samples 1 and 2 was 4%. Each sample has a different composition of cellulose acetate and PEG. The difference in composition does not provide a different

elongation effect. The elongation of samples 1 and 2 was greater than sample 3 because the same pluronic composition (1 g) in samples 1 and 2 makes its flexibility greater than sample 3 which has less flexibility due to excess pluronics (1.5 g).

3.3 The Antibacterial power

The biodegradable plastics have been widely used as packaging for food, fruits, to prolong life after harvest, reduce changes in quality and quantity, and inhibit microbial growth [33]. Plastic from the resulting cellulose is applied as a packaging for mangoes. Mango fruit was chosen because it is cheap and its spoilage is easy to observe. Observations were made for 3 days both with and without packaging. In addition to plastic from cellulose, commercial plastic is also used to package mangoes for comparison.



Figure 4. Comparison of the smart packaging samples (1; 2; 3; 4) with commercial plastic (PK)

Film coatings (biodegradable plastics or commercial plastics) can reduce respiration rates because they limit contact with oxygen in the air and increase internal carbon dioxide which can further slowly the ripening process [33]. The physical appearance of mangoes after being stored for three days can be seen in Figure 5. The freshest mangoes were shown by samples 1 and 2 packaged in biodegradable plastic; while the fast-rotting mangoes were shown in the sample 3 and 4. Although all samples had with added AgNO_3 as an antibacterial agent, samples 3 and 4 had the same pluronic composition making the flexibility even less. This makes room for air so the shelf life of the fruit decreases. As a result, the fruit in samples 3 and 4 rotted faster than samples 1 and 2.

3.4 Morphology of the smart packaging

The surface morphology of the synthesized plastic was observed using SEM (Scanning Electron Microscope). It aims to determine the surface structure, cracks, and surface smoothness of the plastic [34].

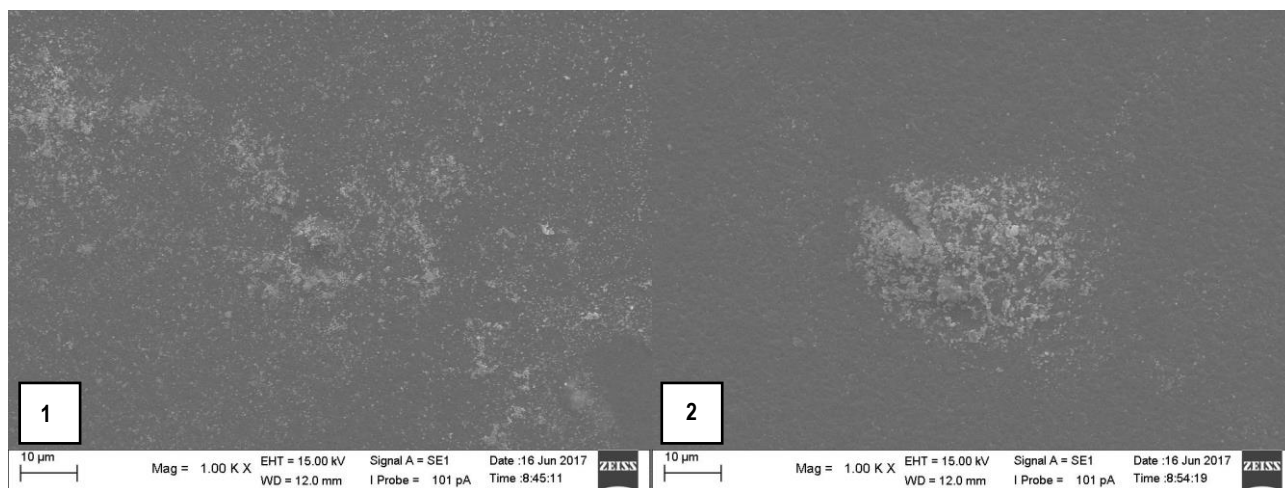


Figure 5. Morphology of the smart packaging samples 1 and 2

Based on the results of SEM testing that has been carried out on sample 1 and 2, the pores of the plastic under test were not visible. The results of the SEM test showed the presence of nano-sized particles that diffused in the plastic body. The spread of nano-sized particles in sample 1 was smoother than the particle distribution of sample 2. The particles in the sample 2 were generally larger and more lumpy in the center of the plastic. These nano-sized particles were most likely cellulose acetate particles.

3.5 The biodegradability power

The biodegradability test of polymer-based plastics aims to determine the ability of plastic to decompose as an environmentally friendly plastic application. Biodegradability can be done by hydrolysis (chemical), bacteria/fungi, light (photodegradation) and others [35]. In this study, the biodegradability test was carried out using a soil burial test technique where the sample was buried in the soil in an open space, with the help of bacteria/fungi in the soil.

Table 1 Percentage of mass lose and degradation degree

Sample	Percentage of mass lose (%)	Degradation degree (g/day)
1	5.66	0.0005
2	11.8	0.0007
3	6.62	0.0004
4	17.38	0.0009

Note: Sample 1 is plastic with a mixture of CA 30 g, PEG 5 g, and pluronics 1 g; sample 2 is plastic with a mixture of CA 40 g, PEG 10 g, and pluronics 1.5 g; sample 3 is plastic with a mixture of CA 30 g, PEG 5 g, and pluronics 1 g; sample 4 is plastic with a mixture of CA 40 g, PEG 10 g, and pluronics 1.5 g

In table 1, it can be seen the difference in the biodegradability of plastic samples 1 and 4. Plastic with a mixture of pluronic 1.5 g has the greatest degradation ability, namely 11.08% (sample 2) and 17.38% (sample 4), compared to pluronic 1 g which is 5.66% (sample 1) and 6.62% (sample 3). This plastic based on cellulose acetate is a hydrophilic component, namely from starch and PEG content. The presence of glycerol in plastic will bind moisture in the air. This causes bioplastics to easily absorb water in the soil so that the swelling power (plastic swelling by the presence of water) becomes greater which causes the plastic to be more easily broken into fragments that will be easily degraded by bacteria/fungi in the soil. The pluronic content in the plastic causes the formation of pores, the plastic with the most pore content will experience the greatest biodegradability because it helps plastic contact with air so that bacteria / fungi will be easier to decompose. In addition, presence of PEG increased rate of biodegradability for the samples [36].

The large molecular weight of the polymer affects the length of the degradation time because it is difficult to break the long chain alkane bonds in its structure. The greater the molecular weight (the longer the chain), the longer the polymer will be degraded [37]. Another factor that helps the decomposition process is environmental conditions, namely the presence of rain and heat will affect plastic degradation. [38] stated that in the biodegradability test, water must be able to enter to penetrate the structure of the material and assist the biological (microbial) activity of the material [39].

4. Conclusion

Cellulose acetate can be used as a basic material for making plastics. The addition of AgNO₃ in plastic synthesis could help to delay the process of fruit spoilage caused by bacteria. Therefore, further research is recommended to find alternative solvents that are resistant to temperature so that the pluronic dissolution process is better.

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