The Effect of Agricultural Waste Nanocellulose on The Properties of Bioplastic for Fresh Fruit Packaging

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Abstract. Despite the high production rate of Indonesian exotic fruits, the export rate has not been as high as desired. One of the main problems in exporting fresh fruits is the control of shelf-life during transportation and distribution. The use of active packaging has been reported to overcome this problem. Bioplastic bags are potential for this purpose, not only to extend the shelf-life, but also to create an environmentally friendly packaging. The objective of this research was to study the effects of the reinforcement of nanocellulose obtained from rice straw and palm empty fruit bunch on the properties of bioplastic. Nanocellulose from rice straw and palm empty fruit bunch were produced using top-down method and blended with bioplastic pellets to produce bioplastic bags. Bags being analyzed for mechanical properties, morphological structure and permeability. The treatment including palm empty fruit bunch Bioplastic; rice straw bioplastics; commercial bioplastic, polypropylene and low density polyethylene plastic bags. Result study showed that bioplastic bags reinforced with palm empty fruit bunch nanocellulose had 23.30% higher tensile strength and 24.76% higher elongation compared with commercial bioplastics, while rice straw nanocellulose reinforcement only increased the tensile strength by 8.51% and did not improve the elongation. The Water Vapor Transmission Rate (WVTR) of bioplastic bags reinforced with rice straw and palm empty fruit bunch nanocellulose respectively was 401% and 399% higher compared to polypropylene plastic bags. Reinforcement with palm empty fruit bunch nanocellulose produced bioplastic bags with better properties due to higher mechanical properties compared to natural bioplastics with lower WVTR.

1. Introduction
The demand of Indonesian tropical fruits in international market is high. However, despite the high production rate of these fruits, the export rate has not been as high as desired. One of the main problems in exporting fresh fruits is the inadequate shelf-life for transportation and distribution. Inadequate shelf-life causes limited volume and distance of export destination.

Packaging is a very influencing factor in quality maintenance of fresh produce. It protects the contents, and determines the convenience of transportation, storage and display of products [1]. Plastics, being low cost and convenient, has grown tremendously in packaging market trends [2]. However, its wide usage has raised concerns on environmental effects in the world [3-6]. Bioplastics are relatively new and upcoming market, emerged to answer the aforementioned concerns [3, 7]. Bioplastics are plastic that are made from renewable resources (bio-based), therefore biodegradable and have less impact on climate change and fossil resources [8]. The use of bioplastics has been reported to maintain the quality and extend shelf-life of fresh fruit [4, 9].

Our previous research showed that bags made from 100% biodegradable material were better in packaging snake fruit for export purpose, compared to polypropylene and polyethylene plastic bags. The porous property of bioplastic allowed better air flow to facilitate the respiration and transpiration process of the fruit, keeping the freshness longer [9]. Unfortunately, biodegradable packaging like bioplastics usually poor on mechanical properties such as tensile strength and elongation thus needed enhancement [10-12]. These properties are important parameters to determine the suitability of the bags for fresh fruit packaging, which may need not only good permeability but also good flexibility and strength.

Numerous exploration researches had discovered the utility of agricultural waste in the development of bioplastics and biodegradable materials, such as sugar cane bagasse, corn cob, teff straw, banana peel, cassava peel, palm empty fruit bunch, rice husk and rice straw [8, 10, 13-16]. But only a few talking about nanoreinforcement from agrowaste. The objective of this research was to determine the effect of the utilization of agricultural waste nanocellulose as reinforcement material in the production of bioplastic to the properties of the bioplastic bags.

2. Method

2.1 The preparation of nanocellulose

Nanocellulose was made of agricultural waste biomass, *i.e.* rice straw (RS) and palm empty fruit bunch (PEFB). Rice straw was obtained from rice milling unit in Karawang, Indonesia; while palm empty fruit bunch was obtained from PT. Perkebunan Nusantara VII, Lampung, Indonesia. Nanocellulose was developed by top-down method using Ultrafine Grinding Machine [17, 18]. Raw material was dried and then reduced into a 1-2 cm size. It was cooked for 8 hours at 80°C. Afterwards, 10% NaOH solution was added and the mixture was autoclaved for 15 minutes at 121°C. The next step was washing until a neutral pH was reached. The obtained mixture was added with 10% sodium hypochlorite solution and was left for 2 hours in room temperature. When the fiber became white in color, it was directly processed using ultrafine grinder in wet milling method. 2% cellulose was mixed with 2 liters of water and passed through the grinder for up to 30 cycles until a nanocellulose gel was formed. The concentration of nanocellulose gel was then increased by centrifugation. The production process was developed based on the method of Abe [19] with a modification by changing sodium chlorite with sodium hypochlorite for its much more inexpensive price, in consideration of affordability for mass production in the future.

2.2 The production of nanocellulose-reinforced bioplastic bags

The production of bioplastic bags was conducted in collaboration with PT. Intera Lestari Polimer who produces bioplastic with commercial brand of Enviplast®. It was a 100% biodegradable bioplastic with cassava as the main raw material. The formulation was developed by using the formula used in producing Enviplast® with the addition of nanocellulose biopolymer obtained in the first step in this study, as reinforcement material. The concentration of nanocellulose used in the formula was 5%. The production of the reinforced bioplastic bags consisted of biocomposite formulation, rheomix extrusion, film blowing and bioplastic sheets cutting.

2.3 The characterization of nanocellulose-reinforced bioplastic bags

Mechanical properties and morphology structure

Parameters observed were tensile strength and elongation, using UTM, based on ASTM. The film sample was measured into 20 mm long and 2 mm wide, given a 500 N load with the speed of 1 mm/minute and range of 10 mm. The analysis was done 5 times. The morphology structure was observed using Scanning Electron Microscope (SEM).
Permeability
Permeability analysis was done by the determination of Water Vapour Transmission Rate (WVTR) using Mocon Permatran based on ASTM. Polypropylene (PP), Low Density Polyethylene (LDPE) and Enviplast plastic bags were also analyzed as comparison.

3. Results and Discussion

3.1 The preparation of nanocellulose

![Figure 1](image1)

(a) crude fiber of palm empty fruit bunch; (b) fiber obtained from delignification; (c) palm empty fruit bunch cellulose; (d) nanocellulose gel obtained from wet milling using Ultrafine Grinder

Natural fibers are generally composed of cellulose, hemicellulose and lignin. The nanocellulose gel resulted in this method contained 71.17% cellulose, 12.86% hemicellulose, and 7.38% lignin [20]. This composition showed an alteration compared to that of the natural fibers of PEFB, which was reported to consist of 40-50% cellulose, 20-30% hemicellulose, and 20-30% lignin [21]. Conclusively, the method used to obtain nanocellulose in this study had successfully removed the hemicellulose and lignin components, although not completely. The removal of hemicellulose and lignin was due to the alkali treatment, as shown in other studies with various natural fiber sources [22-24]. The gel obtained in this stage was ready to be utilized in the making of bio-composite for bioplastic bags production.

3.2 The production of nanocellulose-reinforced bioplastic bags

The starch was mixed with nanocellulose and was destructed by using high temperature and pressure to transform its once semi-crystalline properties into amorph. In this condition, the rigidity would be mostly reduced, making it plastic and could be shaped like the conventional plastic material [25]. Iriani et al. [18] found that the potential formula for development was thermoplastic starch-nanocellulose formula with glycerol plasticizer and added with citric acid before mixing with LDPE polymer, maleic anhydride and dicumyl peroxide. It was also found that sometimes the nanocellulose was not mixed with the polymer matrix homogenously.

In this study, the formula was developed from hydrophilic materials. The starch-based bioplastic formula from Enviplast® was used, and was combined with the nanocellulose from RS and PEFB as reinforcement. In trial, RS and PEFB nanocellulose was added in the gel form with the concentration of 2% (dry base) to reach a composition of 5% in the whole bioplastic formula. The rest of the production stages of bioplastic bags were conducted using machinery of PT. Intera Lestari Polimer with their established Standard Operation Procedure (Figure 2).
Figure 2. The production of bioplastic bags with the reinforcement of agricultural waste nanocellulose (rice straw and palm empty fruit bunch).

3.3 The characterization of nanocellulose-reinforced bioplastic bags

3.3.1 Mechanical properties and morphology structure

The tensile strength of conventional plastic bags/enviplast was found higher than that of bioplastic bags. However, it was shown that the reinforcement of nanocellulose into commercial bioplastic bags had succeeded to increase the tensile strength by 23.30% (PEFB) and 8.51% (RS) (Figure 3). This is a promising result to expand the utilization of bioplastic bags. Nowadays, bioplastic bags have only been used for delicate and light product due to its low tensile strength. The improved tensile strength may arise a new opportunity to using bioplastic bags for other products.

Figure 3. Mechanical properties of different conventional plastic and bioplastic bags.

Based on the elongation, LDPE had the highest elongation compared to other samples (Figure 3). Between the bioplastic bags, reinforcement of nanocellulose from RS did not improve the elongation of the bioplastic bag, while that of PEFB had succeeded to increase elongation by 24.76%. The analysis of the mechanical properties of the reinforced bioplastic bags proved that PEFB nanocellulose can improve the rigidity of bioplastic bags by increasing their tensile strength and elongation. Cellulose derived from PEFB had been reported to have lower density and high specific strength. The concentration of PEFB nanocellulose influenced the tensile strength, as was shown by Lani et al [21], where highest tensile strength was obtained with the higher content of PEFB nanocellulose in PVA and starch solution in the production of nanocomposites. However, more than 10% of nanocellulose content had caused a decreased tensile strength, though still much higher than that of the unreinforced film. This trend applied also for elongation, where a 5% of nanocellulose was the best concentration to gain the best elongation at break.
Das et al [26] elaborated that the improvement of mechanical properties was due to the presence of intermolecular interactions between nanocellulose and starch. On the other hand, an excessive addition of nanocellulose caused reduction of tensile strength and elongation because the increased intermolecular interaction might compete with interactions between polyvinyl alcohol, starch and nanocellulose.

The surface morphology of the reinforced bioplastic studied by SEM was shown in Figure 4 informed that nanocellulose from rice straw gave better homogenous appearance compare to nanocellulose from PEFB. The SEM results also showed that the rice straw and PEFB had become nano fibrillated cellulose that entrapped at matrix polymer. This condition explained why the nanocellulose addition on the proper amount can improve the mechanical properties of the bioplastics.

![Figure 4. Structure of bioplastic with the reinforcement of rice straw nanocellulose (left) and palm empty fruit bunch right)](image)

3.3.2 Permeability

The result of permeability analysis showed that the WVTR on all types of bioplastic bags were very high compared with that of PP and LDPE (Figure 5). The highest WVTR was shown by Enviplast® bags. The reinforcement of agricultural waste nanocellulose into the Enviplast® bioplastic bags was succeeded to maintain the WVTR well over that of conventional plastic bags/petroleum-based plastic bags. This result proved that nanocellulose reinforcement can improve mechanical properties without compromising the rate of water vapour transmission into and from the bags. This property is an advantage for the utilization of bioplastic bags as packaging material for products that needs a certain rate of water vapour transmission such as fresh horticultural produces.

![Figure 5. Permeability of different plastic based on Water Vapour Transmission Rate (WVTR)](image)
The WVTRs obtained in this study were considerably lower compared to those reported by Tengrang et al [27] where the WVTRs of different types of bioplastics made of renewable resources were ranged from 692-1390 g/m²/24h, with the highest WVTR resulted in a prolonged storage life of fresh rambutan fruit. However, the comparison of WVTR of bioplastic (with or without nanocellulose reinforcement) with that of PP and LDPE showed a promising potential in bioplastic utilization as fresh fruit packaging.

4. Conclusion
The Reinforcement of nanocellulose obtained from agricultural waste (rice straw and palm empty fruit bunch) in the production of bioplastic bags can improve mechanical and permeability properties of the bag. Reinforcement using palm empty fruit bunch resulted better properties than rice straw.

5. References

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