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Effect of Nanocrystal Cellulose Isolated from Empty Fruit Bunches (Elaeis Guineensis Jack) Addition on Morphological and Mechanical Properties of Natural Rubber Latex Product.

Siska Yuliani[1],Marpongahtun[2*], and Saharman Gea[3]

[1,2,3]Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Sumatera Utara, Jalan Bioteknologi No.1 Kampus USU Medan 20155, Indonesia

Abstract. The research about the effect of the addition of nano crystal cellulose (NCC) isolated from empty palm bunches (EPB) on the morphological and mechanical properties of natural rubber latex products was successfully conducted. This research was carried out in three steps: i.e isolation of α-cellulose from EPB, followed by isolation NCCs from α-cellulose using H_2SO_4 48.84% at 45ºC for 25 minutes, and manufacture of latex compound with varying filler NCC were 0, 0.6, 1.2, 1.8, 2.4, and 3.0. Manufacturing of natural rubber product was made with pour the NRL followed by vulcanized at 120ºC for 30 minutes. The sheet of NRL product was produced later characterized its mechanical properties through tensile test and surface morphology analysis by SEM. The optimum value of the tensile test result of the NRL product sheet was at the addition of 1,2 phr of NCC with values of tensile test, elongation at break, and Young's modulus was 3.771 MPa, 877%, and 0.430 Mpa respectively. In addition, surface morphology analysis showed that the NCC was spread evenly.

Keywords: NCC, Natural Rubber Latex Product, Morphology Properties, Mechanical Properties

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1 Introduction

Natural rubber latex (NRL) (*Hevea brasiliensis*) is one of the agricultural commodities that has an important role in the Indonesian economy. NRL is consisted of 94% cis of 1.4- polyisoprene obtained by tapping the bark of rubber trees. NRL has a low tensile strength, modulus, and hardness which are the most important mechanical properties required in the industrial field. Therefore, it is necessary the addition of certain materials that can increase the characteristic of the NRL till can be produced into more useful products (Stevens, 2001).

*Corresponding author at: Department of Chemistry, Faculty of Mathematics and Natural Sciences Universitas Sumatera Utara, Medan, Indonesia.

E-mail address: marpongahtun@usu.ac.id

Journal Homepage: http://jcnar.usu.ac.id

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The palm oil (*Elaeis guineensis Jack*) is native to Nigeria, West Africa. In Indonesia, palm oil has an important role in the development of national plantations. In addition to being able to create jobs and also as a source of foreign exchange. Indonesia is one of the main producers of palm oil with the largest palm oil area in the world, which is 34.18% of the world's palm oil areas (Fauzi, 2012).

The increase in palm oil which is also followed by the number of palm oil factories can cause an increase in damage to natural resources and the environment. This is due to the increasing waste disposed to the environment (Widhiastuti, 2001). Empty fruit bunches (EFB) is a solid waste obtained from palm fruit bunches (FFB) processing with an amount of 22 to 23%. EFB consists of 45.59% of cellulose, 22.84% of hemicellulose, 16% of lignin, 1.23 % of ash, 0.53% of nitrogen, and 2.41% of oil (Darnoko, 1992).

Cellulose, glucose polymer, is β-1.4, glucosidic in a straight chain. Nanocrystal cellulose (NCC) is a crystalline nanoparticle obtained from cellulose that can be used as a reinforcing agent in nanocomposites. The nanocomposite is a combination between the matrix and the filler which is nano-sized as a reinforcing element (Espert, 2004).

NRL has also low physical properties when compared to latex which has been given additional materials such as fillers. The filler material is added to the NRL formulation to improve the physical properties of NRL so that it can provide high mechanical strength and can be used for the production of raw material latex.

Many researchers have reported that have conducted research on the use of NCC isolated from bagasse as a filler material in the manufacture of rubber nanocomposite sheets, the rubber nanocomposite sheets rubber produced showed that the addition of NCC into the latex can improve the tensile strength, elongation at break and Young's modulus was 4.7 MPa, 0.65%, and 6.3 MPa, respectively (Brass, 2010). In addition, Harahap (2010) has reported that the use of banana peel flour as a filler in the manufacture of natural latex sheets which rubber nanocomposite sheets resulted in fillers of 5 g and 10 g having good mechanical properties. The properties obtained showed that tensile strength and elongation at break decreased with increasing mass of filler. In contrast, Young's modulus increased with increasing the filler.

Fillers added to NRL can improve the vulcanization of natural rubber so that the tensile strength and other mechanical properties also increase (William, 2008). Each type of filler provides certain properties as a result of its specific chemical surface. A filler with nanoparticle size can increase the polymer reinforcement value compared to large fillers (Leblanc, 2002).

Therefore, it is necessary to conduct research to make NRL product sheet loaded NCC that can accelerate the morphological and chemical properties through vulcanizing the NRL. The formed nanocomposite was characterized using a scanning electron microscope (SEM), tensile testing, swelling tests and transmission electron microscopy (TEM), and fourier transform infrared spectroscopy(FT-IR).

2 Materials and Methods

2.1 Equipments

In this study, equipments used were an oven, glassware, printing plate, analytical balance, dialysis membrane, centrifugation, hot plate, a set of TEM, SEM, FT-IR, and tensile testing.

2.2 Materials

The main materials used were empty fruit bunches, HNO_3, $NaNO_2$, NaOH, Na_2SO_3, NaOCl 1, H_2O_2, H_2SO_4, concentrated latex HA 60%, KOH, $Ca(NO_3)_2$, Metanol, sulfur 50%, wing stay 55%, ZDBC 50%, $CHCl_3$, and $CaCO_3$.

2.3 Preparation of Empty Fruit Buches (EFB)

EFB fibers were separated from the fruit, then washed with water to remove sand and oil from the fibers. Dried under the sun until dry and then cut into pieces of EFB to form fine fibers.

2.4 Isolation of α-Cellulose from EFB

As much as 75 g of EFB was placed into a beaker glass, then added 1 L of a mixture of 3,5% HNO_3 and 10 mg of $NaNO_2$ while heating on a hot plate at 90ºC for 2 hours. After that, it was filtered and the pulp was washed until the filtrate reach neutral. Next added 750 mL of a solution containing NaOH 2% and Na_2SO_3 2% and heated at 50ºC for 1 hour, filtered and the fibers washed until the filtrate reach neutral. Then, the fibers obtained were bleached with 250 mL of 1.75% NaOCl at 70ºC for 30 minutes, filtered, and washed by using distilled water to a neutral filtrate. After that, the purification of the α-cellulose from the sample with 500 mL of 17.5% NaOH solution at 80ºC for 30 minutes, filtered, and washed until the filtrate reach neutral. Furthermore, bleached with H_2O_2 10% at 60ºC, filtered, and washed. Finally, the fibers were dried in the oven at 60ºC (Ohwoavworhua, 2005).

2.5 Isolation of NCC from α-Cellulose

As much as 1 g of α-cellulose was dissolved in 20 mL of H_2SO_4 48.84% at 45ºC for 25 minutes. Then cooled and added with 25 mL distilled water. Next, allowed to stand for one night to form a suspension. The formed suspension was centrifuged at 10,000 rpm for 20 minutes to adjust pH to neutral. Then, ultrasonicated for 10 minutes, and after that placed into the dialysis membrane that has been soaked in 100 mL distilled water. Then, allowed to stand for 8 days while stirring with a magnetic stirrer. After that, evaporated at 70ºC till the NCC was obtained.

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2.6 Fabrication of Natural Rubber Latex (NRL) Product Sheet

NRL product sheet was made on the printer plate. Firstly, the printer plate was soaked in 10% acetic acid and 10% KOH, then washed with water till clean. The clean printer plate was dried for 5 minutes. After that, washed with $Ca(NO_3)_2$ and methanol, then dried (Harahap, 2010). Next, the NRL sheet was made by mixing the 100 Phr latex HA 60%, with 2.9 Phr KOH 10%, 1.7 Phr sulfur 50%, NCC, 1.2 Phr wing stay 50%, 3.5 Phr ZnO 60%, and 2.5 Phr ZDBC 50% and then stirred by the magnetic bar for 2 hours. After that, the latex compound was pre-vulcanized at 70ºC, and the maturation stage of the latex compound was determined using the $CHCl_3$ number technique. Then, the latex compound pre-vulcanized was matured for 24 hours and filtered. After the maturing process, the compound obtained was poured into the prepared printer plate. Then, the plate was dried at room temperature. Next, the compound was vulcanized at 120ºC for 30 minutes. Then, dried, and removed from the printer plate (Harahap, 2010).

3 RESULT AND DISCUSSION

3.1 Isolation of α-Cellulose from EFB

Through a series of processes of the delignification, bleaching, and purification process, obtained white-colored cellulose as shown in figure 1. a. At the stage of isolation was used as much as 75 g of EFB was and the α-cellulose obtained is about 30.24 g (40.32% of the initial weight of EFB). Moreover, α-cellulose obtained was then hydrolyzed using H_2SO_4 48.84% to obtain NCC as presented in figure 1. b. The isolation process was carried out through hydrolysis and dialyzed for 8 days using a dialysis membrane, and then the NCC obtained is about 0.18 g (18% of the initial mass of the α-cellulose).

a) **b)**

Figure 1. a). α-cellulose and b). NCC

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3.2 Natural Rubber Latex Product Sheet

The NCC obtained was mixed into concentrated NRL with a ratio of 0, 0.6, 1.2, 1.8, 2.4, and 3 phr. The mixing process was conducted with stirring for 2 hours and then carried out pre-vulcanized at 70ºC for 30 minutes and matured for 24 hours. After that, the matured compound was poured into the printer plate with a size of 15 cm x 7 cm and then vulcanized at 120ºC for 30 minutes. NRL product sheets were indicated in figure 2.

Figure 2. NRL product sheets

3.3 FT-IR (Fourier Transform-Infrared) Analysis

The FT-IR test results indicated in figure 3 show that the three typical bands commonly found in α-cellulose were the band at 3335 cm^{-1}, which indicated the presence of OH groups, the band at 2916 cm^{-1}, which showed C-H bonds, and the band at 1031 cm^{-1}, which assigned C-O-C bonds. In addition, the three typical bands also found in NCC were the band at 3429 cm^{-1}, which indicated the presence of OH groups, the band at 2885 cm^{-1}, which showed C-H bonds, and the band at 1095 cm^{-1}, which assigned C-O-C bonds (Silverstein, 1981). These typical bands show that α-cellulose and NCC showed no significant difference. Due to both being derived from cellulose.

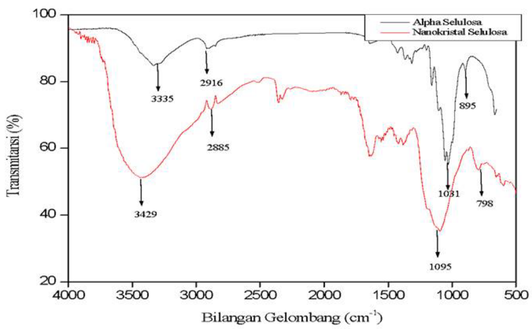

Figure 3. FT-IR spectra of α-cellulose and NCC

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3.4 TEM (Transmission Electron Microscopy) Analysis

Morphological analysis of NCC was carried out using TEM JEOL JEM-1400 with a scale of 200 nm. The TEM test result shown in figure 4 shows that the NCCs produced have different particle sizes. Based on the measurement, obtained the length of the diameter of NCCis about 47.46 nm, where the NCC obtained and analyzed has met the criteria of nanotechnology that has a scale of 1 to 100 nm.

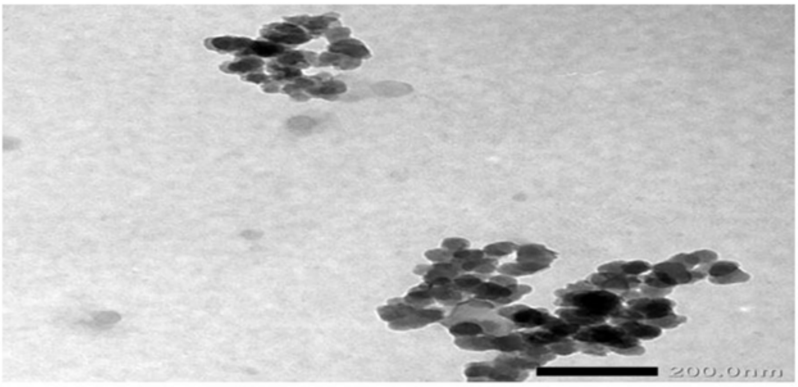

Figure 4. Morphological analysis of NCC

3.5 Swelling Index and Total Solid Content Analysis

The swelling index value was determined in accordance with ASTM D 3615. NRL product sheets are formed roundly with a diameter of 38 mm by the method of immersion in chloroform for 25 minutes.

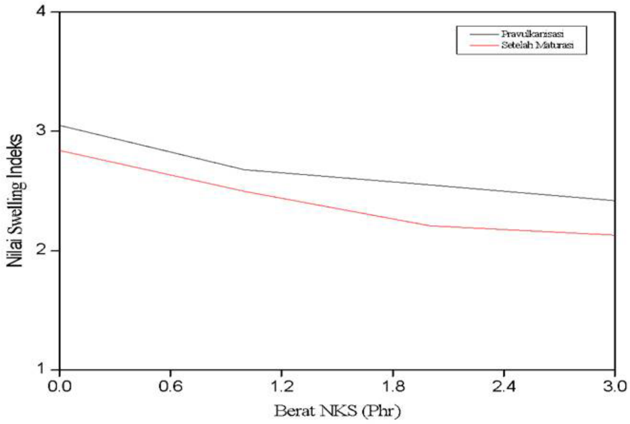

Figure 5. Graph of a swelling index value

The graph of swelling index values presented in figure 5 shows that the swelling index of the sheets produced a decrease followed by the increasing number of NCC. It indicated that the sheets resulted have experienced good cross-linking.

Furthermore, the total solid content (TSC) was determined by drying at 70ºC for 16 hours or at 100ºC for 2 hours (Ompusunggu, 1989). The highest value of TSC was in the variation of 1.2 Phr of NCC. The TSC value of the latex compound determines the quality of the latex product produced. When the value of TSC is low, the tensile strength of the product is also lower. By ASTM D 1076 and ISO 2004 concentrated latex quality, the minimum TSC value is 61.5%.

3.6 Mechanical Properties Analysis

The NCC as a filler material in the polymer matrix can improve the mechanical properties of the composite. Due to the strong chemical bonds and hydrogen bonds that occur between the NCC and the polymer matrix. In addition, NCC has a high homogeneity on its particle size, till it can be dispersed well and evenly in the polymer matrix. The mechanical properties of the composite both stress, strain, and Young's modulus are influenced by the comparison of NCC and polymer matrices.

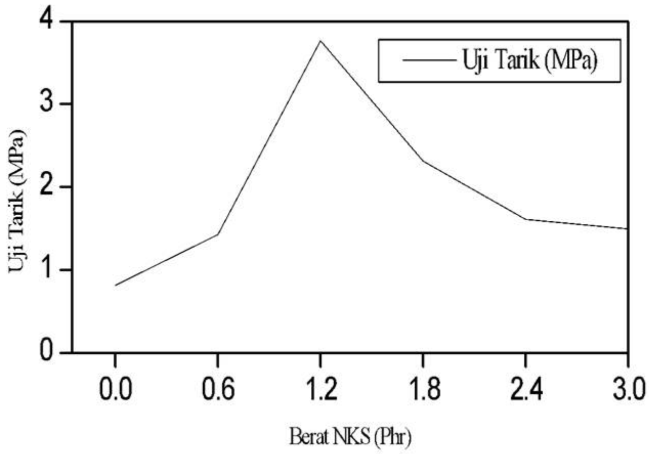

Figure 6. of tensile strength with the mass of NCC

The improvement of mechanical properties of NRL sheets, when added to the NCC, is 1.2 Phr. Due to NCC is as an amplifier, it can improve the strength in the interface area of the filler and matrix. Further, a filler can withstand the voltage applied to the matrix. The decreasing of tensile strength properties on nanocomposites, due to the increasing quantity of filler while the quantity of the matrix decreases causes the interface area to become weak, causes the tensile strength possessed by the product sheet to receive the voltage decreases, and causes the agglomeration of the filler so that there is a decrease in voltage transfer from the matrix to the filler as indicated in figure 6.

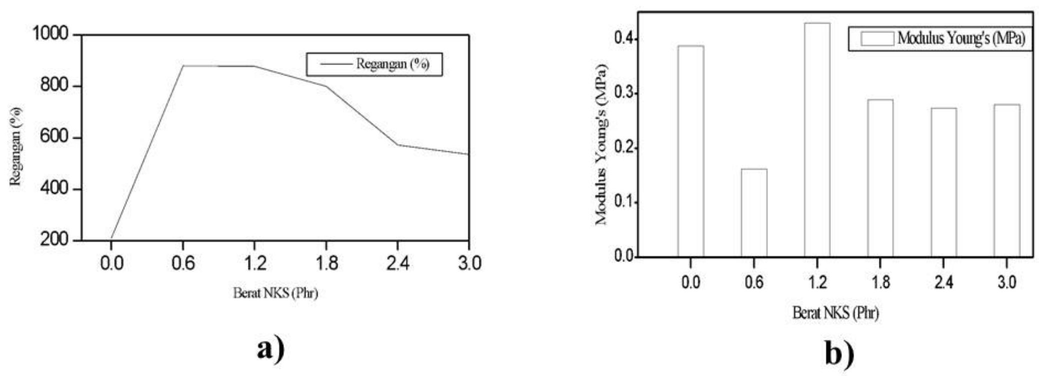

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Figure 7. Relationship of strain with a mass of NCC

Figure 7 illustrates that the strain value decreased with the increase in the weight of the NCC was added into a mixture of NRL. When the addition of the filler was increased, the chemical interactions between phases occur that caused the chain to be hard to move and become stiffer. The resistance to mobility or movement of this rubber molecular chain will cause the vulcanized rubber to break.

NRL products with the addition of 1.2 Phr of NCC is a product with Young's modulus optimum value is 0.430 MPa. Meanwhile, the NRL product with the addition of 0.6 Phr of NCC is a product with the lowest Young's modulus value is 0.162 MPa. Due to the voids produced being larger caused the composite is easily broken during the tensile testing. The strength of the composite is inversely proportional to the number of voids. Moreover, voids can also affect the bonds between the fillers and the matrices, that is the presence of gaps or imperfect forms in the filler can cause the matrix is not able to fill the space in the mold. If the composite receives a load, the voltage area will move to the voids area so that it will reduce the strength of the composite. Composite tensile testing will result in the escape of the filler material from The Matrix. This is due to the strength of the interfacial bond interfacial between the matrix and the filler being less large (Schwartz, 1984).

3.7 SEM (Scanning Elektron Microcopy) Analysis

Morphological analysis was carried out using SEM to observe the surface of the nanocomposite sheet based on concentrated NRL. The SEM test results displayed in figure 8 show differences in the surface of concentrated NRL nanocomposite sheets with no filler and with NCC filler. Morphology with NCC showed a larger aggregate compared to morphology with no filler. This is due to the presence of aggregates from NCC that coat the surface of the natural rubber matrix, thereby also gave an influence the mechanical properties of nanocomposite sheets.

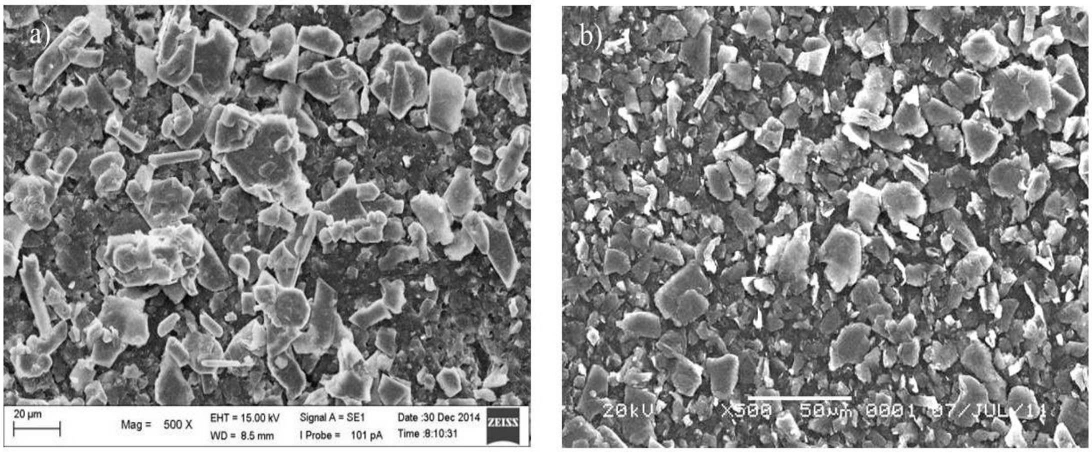

Figure 8. Surface morphology of NRL sheet a). without NCC and b). with NCC

4 Conclusion

In conclusion, the NRL product sheet has been successfully fabricated. This is supported by the characterization results obtained from FT-IR, TEM, and swelling index value. In this study, tensile testing showed that the most optimum mechanical properties were obtained with the addition of 1.2 Phr NCC with tensile strength, Young's modulus, and strain are 3.771 MPa, 0.430 MPa, and 877%, respectively. Based on the morphological observations using SEM showed that NRL products that were given NCC filler showed the aggregates formed is larger compared to NRL products with no NCC filler.

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