

# The Effect of Nano Crystalline Cellulose-Filled to Tensile Strength of Oil Palm Trunk Starch-Based Adhesive

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## Abstract

Has conducted research on the effect of nanocrystals of cellulose produced from oil palm trunk. Nano crystalline cellulose-field into the oil palm trunk starch modified. There was a increase in the quality of oil palm trunk starch which be used to as adhesives. Their modified starch added DSTB. Nano crystalline cellulose produced by oil palm empty fruit bunches used isolation method. Adhesive characterized by tensile strength, and SEM. Durability and adhesion strength of the nanocomposites were compared to those of the Indonesian Standard for wood adhesives which there have been gave us a new information for adhesive. Nanocrystalline cellulose added 2% into nanocomposite were the optimal yield.

**Keywords:** modified-starch, nanocrystalline cellulose, tensile strength, and SEM

## 1. Introduction

The use of additives in technology continues to grow rapidly around the world and research in polymer science continues to evolve rapidly, producing a wide range of new adhesive (Hon, 2003). Adhesive made from flour generally derived from plants such as corn, potatoes, cassava, corn, wheat, rice, and soybeans (Lubis, 2012). In the trunk of sago palm starch contained 4.7% moisture content of 10.65% of the total 2 meters shoots palm trunks. Palm starch composition has a fat content of 0.37%, ash 0.68%, and 1.78% fiber, whereas amilosanya content of 28.76%. Palm starch has a gelatinization temperature 720°C, while the degree of whiteness of 83.02%. Pasta clarity 15.4% T is smaller than the commercial starch. Amilografi pattern of palm starch categorized normal starch, has a viscosity higher end of the commercial starch which indicates the oil starch more susceptible to retrogradation and very well applied as an adhesive (Ridwansyah, et al., 2012).

In general effort to strengthen a filler material is influenced by three main characteristics that the particle size and surface area, shape and surface structure, as well as the activity and chemical properties of the surface. At the same time reinforcing fillers generally have a small particle size, surface active chemicals. Which has a porous surface and a non-uniform shape can improve adhesion (Hanafi, et al., 2005).

Borax (disodium tetraborate decahydrate or abbreviated as DSBT) in a small amount of sodium hydroxide is widely used as an additive for starch-based adhesive. As the dextrin, borax function increases the viscosity and acts as a stabilizer adhesive sticker (tackifier) and viscosity. The effect is particularly important in the application of the adhesive to menyubstrat machine. When used in adhesives, borax is often added in an amount up to 10% dry starch before the starch is cooked. Sodium hydroxide is added to convert enough of borax into sodium metaborate, where there is an active boron species in the thickening (thickening). Metaborate able to implicate two starch molecules at once, forming a complex (Pizzi, 2003).

If the addition of sodium hydroxide is added, the complex compounds will be split; the viscosity of the suspension will begin to decline with increased sodium hydroxide (Baumann and Conner, 2003).

Prioritizing the use of natural raw materials than synthetic raw materials is an environmental issue that has long been developing, including the development of adhesive raw materials. This relates to some of the advantages of natural raw materials, such as more environmentally friendly and quite a lot of potential and can be renewed. Currently development is pursued adhesive glue that little or no formaldehyde and adhesives with little or no use of water-based solvent so that the negative impact on the environment will be reduced. Research and development of the adhesive continues to explore new natural adhesive quality is high and the negative impact on the environment is low. Weakness synthetic adhesives such as Urea Formaldehyde, Phenol Formaldehyde and Formaldehyde Methanol is the availability of sources of raw materials and adhesive diminishing generate formaldehyde emissions from the product material adhesion results for the environment. The emission of formaldehyde can cause dizziness, headache and insomnia (Umemura, 2006).

Oil palm trunk is an abundant solid waste in palm oil plantations especially during the replanting season which possesses valuable starch content but has not been utilized commercially. On the other hand, the oil palm trunk starch may be used as wood adhesives but the mechanical and adhesion strengths were low due to its high water absorption. In this work the oil palm trunk starch was extracted using presipitation in water, and modified using disodiumtetraborax (DSTB) to promote crosslinking reactions within the starch main chains which in-turn improve its adhesion strength on the substrate surface. Mechanical strengths as well as durability of the starch matrix were also improved by addition of nanocellulose as a filler to produce a nanocomposite adhesives for wood substrates. The weight ratios of nanocellulose to the composites were varied from 1%, 2%, 3%, 4%, and

5% respectively. The nanocomposites were then characterized for their physical and mechanical properties using tensile strength, as well as morphological properties using scanning electron microscopy (SEM). The nanocellulose improved the quality of the composite. Compositions and loading of the nanofillers against modified-starch matrixes were also varied and their durability and adhesion strength were compared to those of the Indonesian Standard for wood adhesives.

Oil palm trunk have used to thermoplastic board by the treatment of reactive impregnation treatment technique by using the recycle of polyolefine and then called as a polyolefine impregnation on the palm trunk which its did at around palm board surface only and didn't to be into the center of palm trunk (Wirjosentono, et al., 2001).

Oil palm trunk so have used to the raw material of the particles board by using the adhesive from polypropilene degraded which to be fungsionalized by benzoyl peroxide, maleat anhidride, and di-vinyl benzene (Nasution, 2011).

Starch can be obtained by extracting from oil palm trunk. Palm trunk contains starch that can be obtained by performing the extraction, which contained the highest starch yield on the shaft of oil in the 1 meter from the top of the stem with a yield of 3.3% (Ariansyah, et al., 2011).

The modified starches synthesized in the present study are found to be hydrophobic and can biodegrade in composting. Blends of a modified starch and poly(ethylene-co-vinyl alcohol) (EVOH) were prepared by melt blending. Phase diagrams of the blends exhibited a lower critical solution temperature. The nanocomposites based on a modified starch and EVOH were prepared. Natural clay (montmorillnite, MMT) and two commercial organoclays (Cloisite 30B and Cloisite 15A) were employed to investigate how the functional groups in the modified starch influence the dispersion characteristics of nanocomposites. Anionically modified starch was found to be very effective in exfoliating organoclay aggregates due to the presence of ionic interaction, as determined by Fourier transmission infrared (FTIR) spectroscopy, between the anionic group in the modified starch and positively charged  $N^+$  in the surfactant residing at the surface of an organoclay. For the nanocomposites based on a cationically modified starch and EVOH, the preparation methods had a large influence on the dispersion characteristics of the nanocomposites. The aggregates of MMT have a very high degree of dispersion characteristics in the nanocomposites prepared by solution blending but poor dispersion characteristics in the nanocomposites prepared by melt blending. FTIR spectroscopy has indicated that the ionic interaction between the cationic group in a modified starch and negatively charged surface of silicate sheets of MMT could be formed in the nanocomposites prepared by solution blending but not in the nanocomposites prepared by melt blending. We have found that an improvement in the tensile properties of nanocomposites can only be obtained if the nanocomposites have a very high degree of dispersion of the aggregates of clay, and there is strong attractive interaction between the clay and the matrix (Lin Song, 2010).

Investigation of effect of nano- $Al_2O_3$  on adhesion strength of epoxy adhesive and steel, journal showed that the highest adhesion strength was obtained with 2%wt nano- $Al_2O_3$  in epoxy adhesive, being almost four times higher than that of the unmodified. As the adhesion strength increased, the locus of failure changed from interfacial to the mixture of interfacial and cohesive (Zhai, et al., 2007).

Empty fruit bunches (EFB) is used to be the flour of pulp EFB esterificated as the reinforcement in the composite matrixes of polyetilene have studied about their adhesion. SEM analysis showed that the flour of esterified EFB pulp had the improvement adhesion force which reacted with polyetilene. Its effected by the change of surface from the hydrofilic to be the hydrofobic (Daulay, 2009)

Nano crystalline cellulose (NCC) isolated from empty fruit bunch (EFB) using a solvent mixture of DMAC/LiCl, were passed on activated dialysis membrane after centrifugated to release it from the solvent. The purpose of additional GMS antistatic is to lower the volume resistivity of nanocomposite PS/NCC which usually containing static electricity either due to the nature of the material or by the fabrication process itself. XRD analysis showed that the NCC formed was nanocellulose I with an average size of 72 nm diameter, because of its high surface are, the addition of NCC were able to improve the mechanical properties of the nanocomposite but did not affect the nature of the resistivity (Adriana, 2014).

Tensile test results obtained in the presence of nano particles increase in natural zeolite, the composition of 2%wt with calcination process give 8 Mpa and 6%wt without calcination give 7.7 Mpa, while without the nanozeolite give 6.6 Mpa tensile strength (Bukit, 2011).

Production of cassava starch-based adhesive studied investigate possible improvement methods of the properties of the adhesives produced by studying the effects of borax and temperature on the viscosity, density and pH of adhesive and the use of two different modifiers (HCl and NaOH) in the production of the adhesive; thus provide a range of conditions for producing starch-based adhesives for diverse applications depending on the required properties and industrial applications (Gunorubon, 2012).

## 2. Experimentals

The first, raw material like the oil palm trunk starch produced by oil palm tree 22th years old and was do

extraction and presipitation processes. NCC produced by oil palm empty fruit bunches used isolation process. The second, the oil palm trunk starch, DSTB, NCC, NaOH/HCl has prepared and then blending in the variety mass on hotplate at temperture 338K. DSTB used 5%weight for each sample. The third, characterization by tensile strength and SEM.

### 3. Results And Discussions

The minimum strain value is 4th sample, so did for Mo value and stress value is 1st sample which be showed on the Table 1.

Table 1. Average of tensile strength for adhesive by 2nd replications

No	Sample	Strain	Stress	Mo
1	No DSTB (A)	1,072.290	97.16,923	0.182,492
2	With DSTB (Ab)	1,102.355	92.28,203	0.169,220
3	Ab+1%NCC	1,078.033	87.95,288	0.167,577
4	Ab+2%NCC	955.764	95.41,088	0.241,536
5	Ab+3%NCC	1,074.752	87.16,509	0.177,085
6	Ab+4%NCC	1,066.606	85.92,613	0.201,391
7	Ab+5%NCC	1,024.963	82.08,150	0.163,934

Characterization of yield using SEM showed on figure 1 and 2 in the following:

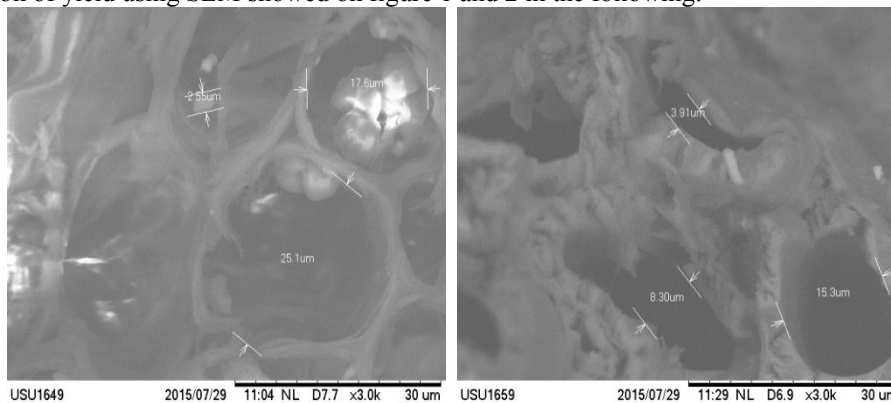


Figure 1: characterization of SEM for adhesive without DSTB and filler (a) and for adhesive added DSTB without filler (b) respectively with scale 1:3000.

In Figure 1 above shows the DSTB minimize cavity - cavity in the adhesive without addition DSTB and filler. Or in other words, DSTB occupies most of cavity - the cavity is in adhesive before being added DSTB.

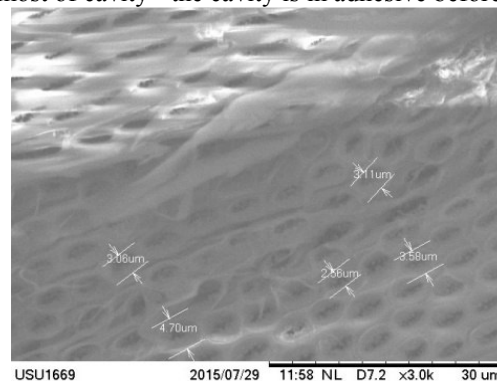


Figure 2: SEM for adhesive added DSTB+2%NCC with scale 1:3000.

In Figure 2 above shows that the addition of STB and 2% NCC similar result occurs woven ties, which allegedly is the crosslinking.

While the morphology of the adhesive with the addition of borax and NKS shows the adhesive surface of the closer it is because cellulose nanocrystal distributed on the surface of the adhesive so that it can enter the cavities can even cover the cavities seningga resulted in increasingly dense adhesive surface. For SEM results on the adhesive with the addition of borax and nanozeolit show denser and solid surface, this is due nanozeolit particles distributed on the surface of the adhesive.

### Adhesive characteristics with SNI

Adhesive obtained in this study were tested to determine whether the adhesive obtained sesuai Indonesian National Standard. SNI were used in this study SNI 06-0060-1998 Urea formaldehyde liquid for glue plywood that covers the quality requirements; pH (25°C) = 8-10; density (25°C) = 1200-1240; Formaldehyde-free maximum = 2%; gelatinasi period (100°C) minimum = 60; Dry bonding strength of plywood minimum = 10 kg/cm<sup>2</sup>. The test results on the adhesive quality standards obtained are shown in the table below:

Based on pH indicators which pass SNI adhesive is the adhesive with the addition of borax and adhesives with the addition of borax and NKS 1% with a pH of 9. As for the second adhesive gelatinasi time and temperature not exceeding SNI gelatinasi so it is still categorized according to SNI, when viewed from the Test adhesiveness, then the adhesive with the addition of borax and NKS 2% (11.15 kg/cm<sup>2</sup>) meet the criteria of SNI.

The most important indicator of the adhesive is tacky adhesive test, based on the above results adhesive that best meet the criteria of SNI adhesive is the adhesive with the addition of borax and NKS 2%.

### 4. Conclusion

Physical, mechanical, and morphological characteristics of the nanocrystalline cellulose-filled modified oil palm trunk starch nanocomposites are reported.

Durability of the nanocrystalline cellulose-filled modified oil palm trunk starch nanocomposite to heat base on SEM. Durability and adhesion strength of the nanocomposites were compared to those of the Indonesian Standard for wood adhesives which there have been gave us a new information for adhesive. Nanocrystalline cellulose added 2% into nanocomposite were the optimal yield.

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### References

- Adriana (2014). The Polimer of Nanocomposites base on Polystyrene and Nanocrystalline Cellulose (NCC) from Empty Fruit Bunches (EFB) by using of The Antistatic Aditives from Glycerol Monostearate to be Technique Raw Material. *Disertation, The Doctoral Programme of Chemistry, Faculty of Science and Mathematics, University of Sumatera Utara, Medan.*
- Ariansyah, F., Laga, A., and Mahendradatta, M. (2011). A Study of Starch Extraction Base on The Highness of the oil palm tree (*Elaeis guineensis*). *Journal, University of Hasanuddin. Bandung.*
- Baumann, MGD., and Conner, A.H. (2003). Carbohydrate Polymers as Adhesives: Forest Products Laboratory, USDA–Forest Service. *Handbook of Adhesive Technology, Revised and Expanded, Chapter 22, Second Editions, Madison, Wisconsin, U.S.A.*
- Bukit, N. (2011). The Treatment of Natural Zeolite as The Nanocomposite field in Polypropilene and natural rubber SIR-20 by using Compatibilizer from Maleic-Grafted-Polypropilene Anhydride. *Disertation, The Doctoral Programme of Chemistry, Faculty of Science and Mathematics, University of Sumatera Utara, Medan.*
- Daulay, L.R. (2009). The Adhesion of The Reinforcement of The Pulp Flour from Empty Fruit Bunches (EFB) Esterified with The Matrixes of Polyietilene Composite. *Disertation, The Doctoral Programme of Chemistry, Faculty of Science and Mathematics, University of Sumatera Utara, Medan.*
- Gunorubon, A.J. (2012). Production of Casava Starch-Based Adhesive, Department of Chemical/ Petrochemical Engineering Rivers State University of Science and Technology, Port-Harcourt, Rivers State, Nigeria, Research. *Journal in Engineering and Applied 1(4) 219-214, Nigeria.*
- Hanafi, I., Halimatuddahlia, and Hazizan. (2005). Properties of Polypropylene/Ethylene- Propylene Diene Terpolymer/Natural Rubber (Pp/Epdm/Nr) Ternary Blend: The Effect of Dynamic Vulcanization. *Solid State Science and Technology, Vol.13, No.1 & 2.*
- Hon, S, and David, N. (2003). Analysis of Adhesives. *Handbook of Adhesive Technology, Revised and Expanded, Chapter 14, Second Editions, South Carolina, USA.*
- Lin Song (2010). Chemical Modification Of Starch And Preparation Of Starch-Based Nanocomposites, *Disertation, The Graduate Faculty Of The University Of Akron.*
- Lubis. (2012). Breadfruit Starch Hydrolysis with H<sub>2</sub>SO<sub>4</sub> Catalyst for Development Adhesives, *Department of Chemical Engineering, Faculty of Engineering, UNSYIAH, Banda Aceh.*
- Nasution, D.Y. (2011). The Fungsionalization of Polypropilene Degradated by using Benzoyl Peroxide, Maleic and Divynyl Benzene Anhidride as The Adhesive of The Particle Board of The Oil Palm Trunk. *Disertation, The Doctoral Programme of Chemistry, Faculty of Science and Mathematics, University of Sumatera Utara, Medan.*

- Pizzi, A. (2003). Principles of Polymer Networking and Gel Theory in Thermosetting Adhesive Formulations. Handbook of Adhesive Technology, Revised and Expanded, Chapter 8, Second Editions, Ecole Nationale Supérieure des Technologies et Industries du Bois. *Universite' de Nancy I, Epinal, France.*
- Ridwansyah, Nasution, M. Z., Sunarti, T.C. and Fauzi AM. (2012). Characteristics of Physico-Chemical properties Pati Palm, Department of Technology Agriculture, Faculty of Agriculture USU, Medan and Technology Department Industri Pertanian. *Faculty of Agricultural Technology IPB, Bogor.*
- Umamura, K. (2006). Wood-Based Materials and Wood Adhesives. Recent Trend in Japan. Cibinong: Article Wood Science School di UPT Biomaterial LIPI.
- Wirjosentono, et al. (2001). Production of Thermoplastic Board from Oil Palm Trunk with Reactive Impregnation Technique of Polyolefine Recycle. *Domestic Collaboration Research, The Department of Chemistry, Faculty of Science and Mathematics, University of Sumatera Utara, Medan.*
- Zhai, et al. (2007). Effect of nano- $Al_2O_3$  on Adhesion Strength of epoxy adhesive and steel, *Journal, College of Materials Science and Chemical Engineering, Zhejiang University, Hangzhou 310027.* People's Republic of China.