doi:10.20944/preprints201710.0134.v1

Article

Improvement in Mechanical Properties of Cantala Fiber and Short Cantala/Recycled High-Density Polyethylene Composite through Chemical Treatment

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Abstract: The improvement of mechanical properties of cantala fiber and its composites. Treatments including alkali, silane, and the combination of both were carried out to modify the fiber surface. The influence of chemical treatments on fiber properties such as the degree of crystallinity and tensile strength was investigated. A variety of short cantala fiber reinforced rHDPE composites were produced by hot press, and the effect of fiber treatment on the flexural strength of composites was observed. SEM observations also carried out to highlight these changes. The result shows that alkali treatment improves tensile strength and tensile modulus of alkali treated fiber (NF12) which was predicted as a result of the enhancement of the cellulose crystallinity. In contrast, the tensile strength and tensile modulus of silane (SF05) and alkali-silane treated fiber (NSF05) decreased compared to untreated fiber (UF) which is caused by the addition of amorphous material. The tensile strength of alkali-silane treated fiber (NSF05) was lower than alkali treated fiber (NF12), but the composites prepared with NSF05 showed the highest increment of flexural strength of 25.9%. This may be due the combination of alkali and silane treatment helped in the better formation of fiber-matrix interface adhesion.

Keywords: chemical treatment; cantala fiber; mechanical properties; recycled high-density polyethylene

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1. Introduction

In the last decade, the field of plastic composites reinforced with natural fibers experiencing rapid development. The use of natural fibers replaces the synthetic fiber as a reinforcement or filler in plastic to reduce costs, cut weight, increase toughness, enhance the biodegradability and improve the mechanical properties [1, 2]. However, for the use of fiber as reinforcement in plastic, cantala fibers similar to other natural fibers have some drawbacks, i.e. the relatively poor compatibility with polymers (3), the relatively small thermal resistance (4), and poor resistance to moisture (5).

The nature of fiber, matrix and the interfacial properties of fiber-matrix play a major role in the final properties of a natural fiber-plastic composite. To get the good mechanical properties of composites, the fibers need to be modified. One of the method to modify natural fibers is chemical treatments. Alkali and silane are the chemical treatments which are the most widely used. Alkali treatment increases the fiber roughness and enhances the exposed cellulose on the fiber surface, whereas, silane treatment creates a layer on the fiber surface to bind the matrix.

A lot of research which is related to virgin thermoplastic composites reinforced with natural fibers have been done. On the other hand, research done on the use of recycled plastic as a binder of natural fibers is still limited. The amount of plastic waste in the worldwide is increased. The World Bank predicts that Indonesia produced solid waste about 152 thousand tons per day with 10% or

15.2 thousand tons are plastic in 2025 (6). Thus, the plastic waste is a potential source in the manufacture of plastic composite.

In this study, the effect of alkali, silane, and alkali-silane treatment on the fiber properties and its composite have analyzed and compared. The effectiveness of treatment on fiber properties is investigated through the fiber properties such as crystallinity index, surface morphology, and single fiber strength. Finally, the flexural properties of composites have been studied to predict the effect of chemical treatment on the treated fiber-rHDPE composites.

2. Materials and Methods

2.1. Materials

Cantala fibers used in this study were extracted from the leaves of Agave Cantala Roxb using mechanical extraction process. rHDPE flakes derived milk jugs were obtained from CV Vanilla Plastik Sukoharjo, Indonesia. The material has a melt flow index of 2.43 g/10 min at 180°C, melting temperature range from 108.5 to 139.5°C, and a density of 1,014 kg/m^3 (7). To modify the cantala fibers by chemical treatment, Sodium hydroxide (NaOH) and Amino ethylamino propyl tri-methoxy silane (AAMS) were used as fiber treatment. NaOH was supplied by the Merck company and AAMS provided by Dow Corning.

2.2. Fiber Surface Treatment

2.2.1. Alkali treatment

The fibers were soaked in 2 wt% NaOH solution at room temperature for 12 h. Furthermore, the treated fibers were rinsed with water repeatedly until pH~7 reached. The wet fibers were dried at room temperature for 48 hours and followed by a heating process in an oven at 70°C for 10 hours.

2.2.2. Silane treatment

A solution of 0.5 wt % amino ethylamino propyl tri-methoxy silane was performed in distilled water. Acetic acid was added to a silane solution to get the pH range 3-4. Cantala fiber was immersed in the solution for four hours. After that, the fiber was washed with distilled water. Wet fibers were kept at room temperature for 48 hours. Finally, the fibers were dried at 70°C for 10 hours.

2.2.3. Alkali and silane treatment

In alkali-silane treatment, before silane treatment, cantala fibers were treated with NaOH solution at 2%, at room temperature. After that, fibers were immersed in silane solution using the same method as for silane treatment of cantala fibers. In this study, all the procedures that were applied to cantala fibers were listed in Table 1.

Table 1. Coding for various treated and untreated cantala fibers

Material code	Description
UF	Fiber without treatment
NF12	Fiber treated with 2 wt% NaOH aqueous solution for 12 hours
SF05	Fiber treated with 0.5 wt% silane aqueous solutions
NSF05	Fiber treated with 2 wt % NaOH aqueous solution for 12 hours and then with 0.50 wt% silane aqueous solutions

2.3. rHDPE Fiber Preparation

rHDPE fiber was derived from flakes of milk jugs. The first, flakes were crushed into small pieces with a diameter of about 7-8 mm. Then, the material pieces were processed into rHDPE fibers using a gas atomizer. Finally, rHDPE fibers collected and were stored in a sealed plastic bag, before the material was processed further.

2.4. Composite and Specimen Preparation

Cantala fiber reinforced rHDPE composites were obtained by mixing the rHDPE matrix with each type of untreated and treated cantala fiber using household blender Kirin model KBB-515 SG. First, the dried cantala fibers chopped to length around 10 mm using the sharp scissors. Then, chopped fibers were mixed with rHDPE fibers in a weight ratio of 30% : 70%, respectively. Afterward, the mixtures were placed in a rectangular aluminum box with a dimension of 170 x 110 x 17.5 mm. Next, the hot-press was used to press panels under 50 bar pressure for 25 min at 150°C. Finally, the panels were cooled in the hot-press until they reached a final temperature of 40°C. For impact testing, the nominal sample thickness was 4 mm, while the nominal thickness of the bending strength specimens was 3.2 mm. For the composite name, the first letter represents the type of fiber used as reinforcement, followed by recycled high density polyethylene matrix is denoted as ''rHDPE''. For example, NF12/rHDPE means the composites with fiber alkali-treated in a 2% NaOH solution for 12h as reinforcement and rHDPE as a matrix.

2.5. X-ray Diffraction

X-ray diffraction was used to investigate the effect of chemical treatment on the fiber crystallinity. XRD analysis was carried out with PanAnalytycal type X-Pert Pro Diffractometer with copper radiation. The CuKα (λ=1.54$\acute{\mathrm{A}}$) was operated at a voltage of 40 kV and a current of 30 mA. The samples were examined in powder form. The diffraction intensity of sample was scanned in the range 10-70° with a speed of 2°/min. The crystallinity index (CrI) of the samples was estimated using the Segal empirical method [5, 8].

$$CrI = \frac{I_{002} - I_{am}}{I_{002}} \ x \ 100 \qquad (1)$$

Where I_{002} is the maximum intensity of crystalline area 002 at 2θ angle in the range 22-23° and I_{am} is the intensity of the amorphous region at 2θ = 18-19°.

2.6. Tensile test of fiber

The tensile test of single cantala fibers was carried out at room temperature according to ASTM C1557 using the universal testing machine JTM-UTS 510. The fiber was mounted and fastened on a cardboard tab with a drop of glue with a gauge length of 12 mm. Fiber diameter was measured by stereo zoom microscope SZX7 at three different locations along the fiber length. The mounted fibers were placed in the grips, and then the middle portion of the papers was cut using scissors. After that, the fiber was loaded until it was broken. The crosshead speed was set 10 mm/min at ambient conditions. Thirty samples for each variation and the average value was taken as the tested value.

2.7. Flexural test of composites

The three-point bend test of composites performed with a universal testing machine JTM-UTS 510. The test was carried out by the ASTM D-790. The width and thickness of the specimens were measured and recorded. The specimen uses a span of 51.2 mm and a crosshead speed of 5 mm/min. For each variation, it used at least five samples. The testing value was taken from the average value of the test result. The entire test was carried out at room temperature. The flexural strength of composite was calculated from this test.

2.8. Scanning Electron Microscopy (SEM)

Scanning electron microscopy (SEM), VEGA 3 TESCAN was used to investigate the fiber surface morphology. These observations were made to observe the effects of chemical treatment on the morphology of the fiber surface.

3. Results and Discussion

3.1. Surface Modification of Fiber

Figures 1a-d show the surface features of untreated and treated fibers. The existence of wax, fat, and impurities cause the UF surface heterogenous. On the alkali modification, the treatment effectively eliminated impurities and non-cellulosic material, so that NF12 looked clean and had many grooves (Figure 1b).

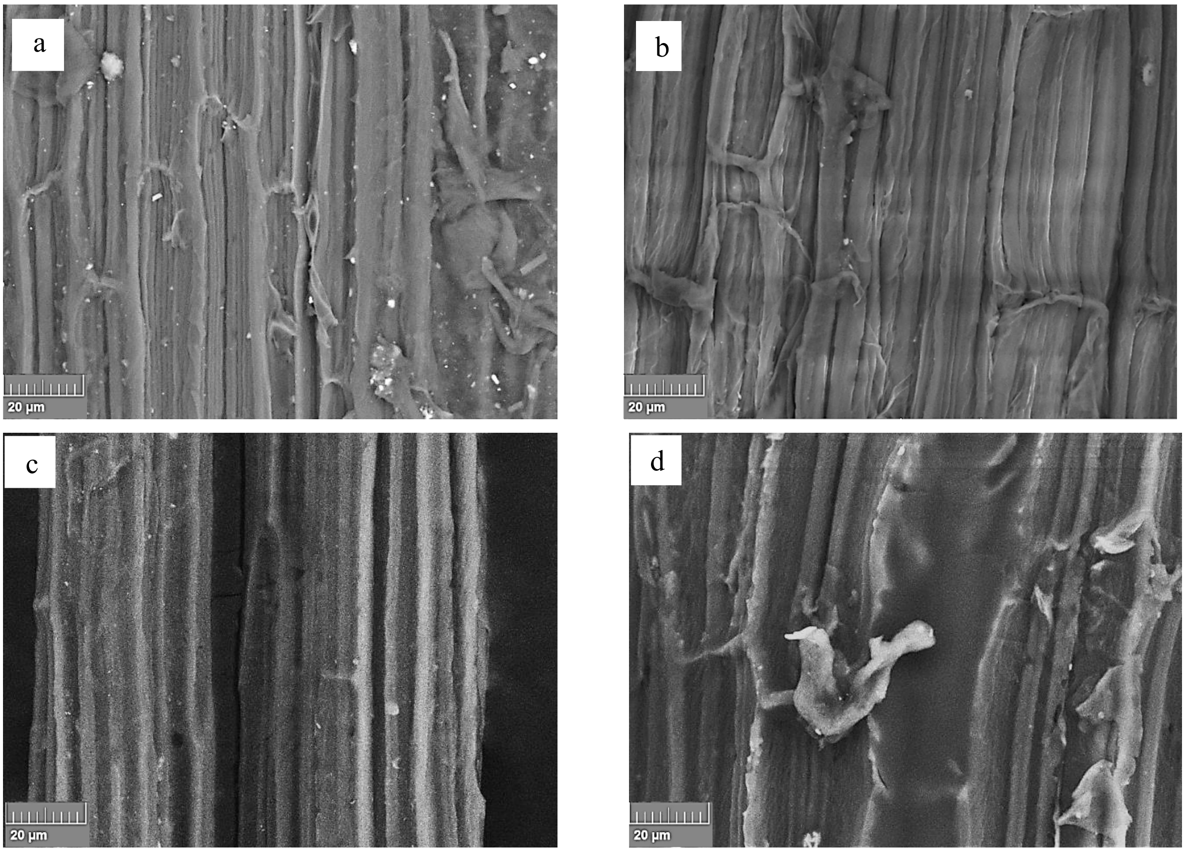

Figure 1. SEM micrographs of untreated and treated; a) untreated, b) alkali treated, c) silane treated, d) alkali-silane treated.

After the silane and alkali-silane treatment (Figures 1c-d), a thin layer appeared on the fiber surface. This was due to the occurrence of the link between silane molecules and the fiber constituents during the treatment. A similar result was obtained by [9-11], in which, silane was effectively deposited on the fiber surface.

3.2. XRD Analysis of Fiber

Figure 2 shows the X-ray diffraction pattern of untreated and treated cantala fibers. The major intensity peak of each profile was located at a 2θ value of around 22°. This peak corresponds to the reflections of (002) crystallographic plane, which represents the crystalline peak of cellulose [12, 13]. On the alkali treatment, it can be seen that the peak at 22° was increased significantly. Meanwhile, the crystalline peak of SF05 and NSF05 did not change much. This fact suggests that the NaOH treatment removes amorphous material more than others.

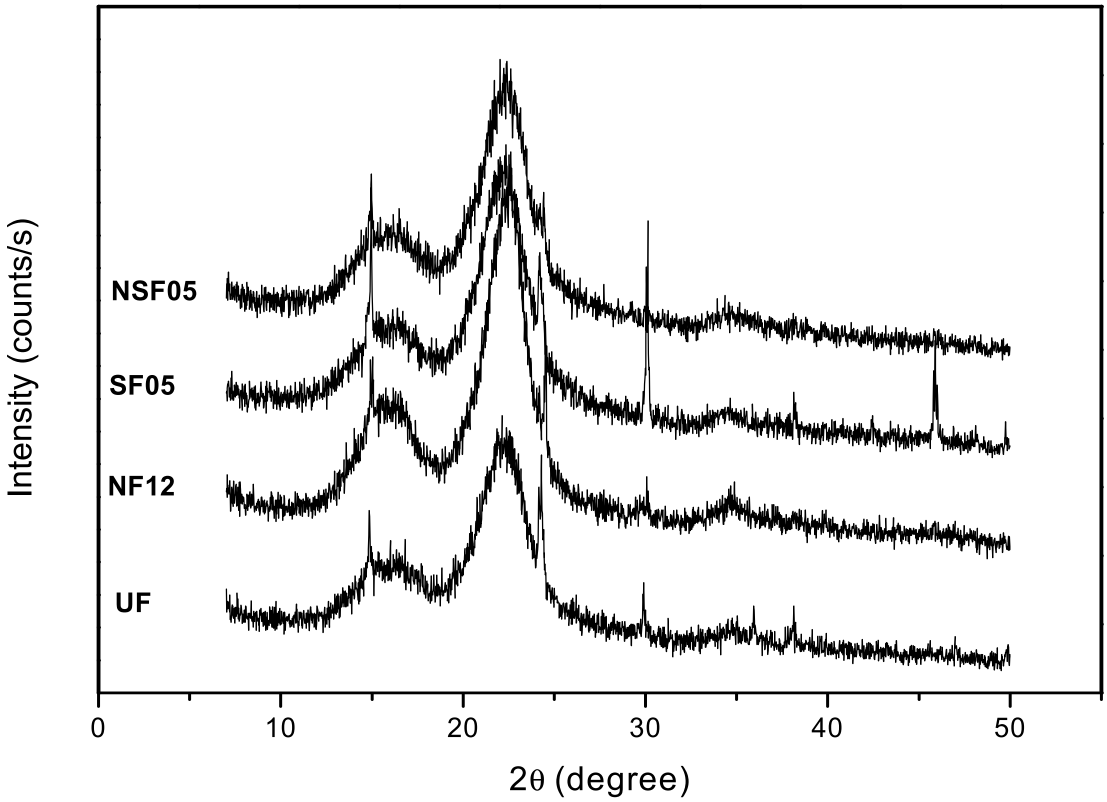

Figure 2. X-ray diffraction profiles of untreated and treated cantala fiber

The effect of chemical treatment on the crystallinity index of treated and untreated cantala fibers is presented in Table 2. The crystallinity index of cantala fibers increased in the following order SF05 (84.6%) < UF(85.1%) < NSF05 (86.7%) < NF12 (89.1%). After alkali treatment, the crystallinity index of NF12 and NSF05 was higher compared to UF. This can be attributed to the removal of the amorphous component from the fiber surface so it enabled the better rearrangement of cellulose chains. This is appropriate to previous work [13, 14].

On the contrary, the crystallinity index silane fiber (SF05) declined in comparison with untreated fiber (UF). The reduction in the crystallinity index after silane treatment could be attributed to the increasing of amorphous cellulose content. The establishment of new amorphous cellulose as a result of a reaction between silane molecules with cellulose in the amorphous cellulose regions encouraged the formation of a more amorphous cellulose. Similar behavior has been informed by [14, 15].

Table 2. Crystallinity index of untreated and treated cantala fiber

Material Code	**I_{002}** Peak Position (2θ) (°)	**I_{002}** Intensity Counts/s	**I_{am}** Peak Position (2θ) (°)	**I_{am}** Intensity Counts/s	**Crystallinity index (CrI) (%)**
UF	18.71	38.53	22.25	259.09	85.1
NF12	18.48	48.88	22.36	448.28	89.1
SF05	18.87	51.11	22.15	332.75	84.6
NSF05	18.82	41.07	22.26	308.57	86.7

3.3. Fiber Tensile Strength

The tensile properties of untreated and treated cantala fiber are displayed in Figure 3. The tensile strength of NF12 increased slightly, compared to UF. The improvement was contributed by the enhanced of crystalline constituents in the fiber as reported in XRD analysis. Besides, the tensile modulus of NF12 was also seen higher than UF. This matter, due to alkali treatment dissolves the amorphous material as supported by SEM. The loss of amorphous material caused the compaction of fiber cell walls so that the tensile modulus of NF12 was increased.

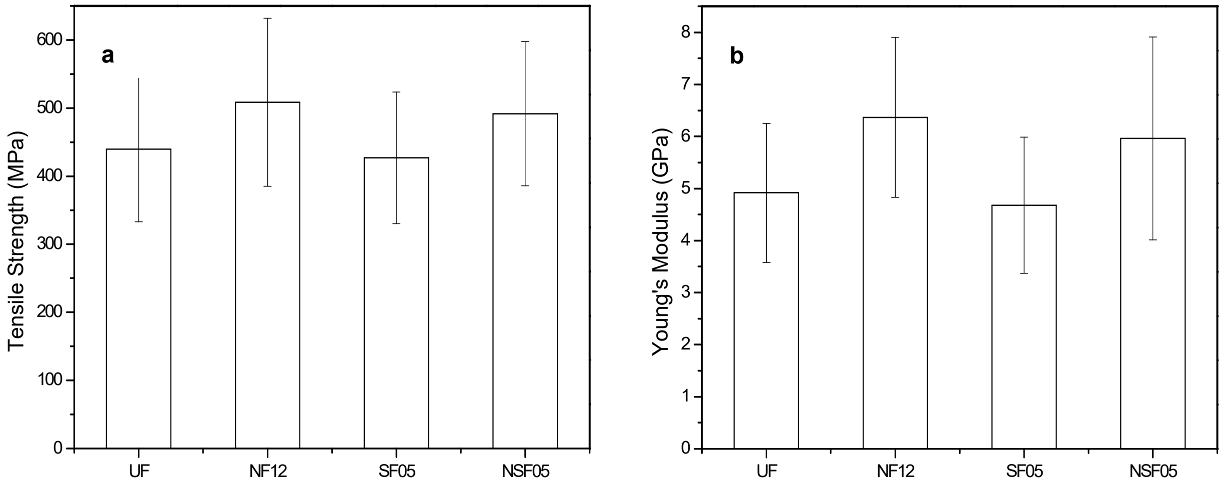

Figure 3. Tensile properties of untreated and treated cantala fibers : a). strength; b). young's modulus

The tensile strength of SF05 and NSF05 were lower than UF. It was caused by the addition of amorphous material obtained by the link formation between silane molecules and fiber constituent. Other than that, the increasing of amorphous materials lead to a decrease in the tensile modulus of SF05 and NSF05. A similar tendency is observed by [11, 16].

3.4. Flexural Properties of Composites

The effect of chemical treatment on flexural strength of cantala fiber/rHDPE composites is shown in Table 3 and Figure 4. The mean and standard deviation values of at least 5 samples for each variation of the treated cantala fiber reinforced rHDPE composites are presented. The flexural strength of composites reinforced with alkali treated fiber (NF12/rHDPE) was increased by 16% compared to UF/rHDPE composites. This improvement was due to the formation of mechanical interlocking between the NF12 and rHDPE. This observation is in line with the previous research (17).

Table 3. Flexural strength of composites

Material Code	**Flexural Strength (MPa)**	
	Mean Value	**Standard Deviation**
UF/rHDPE	30.88	4.14
NF12/rHDPE	35.82	3.29
SF05/rHDPE	36.90	3.37
NSF05/rHDPE	38.88	3.55

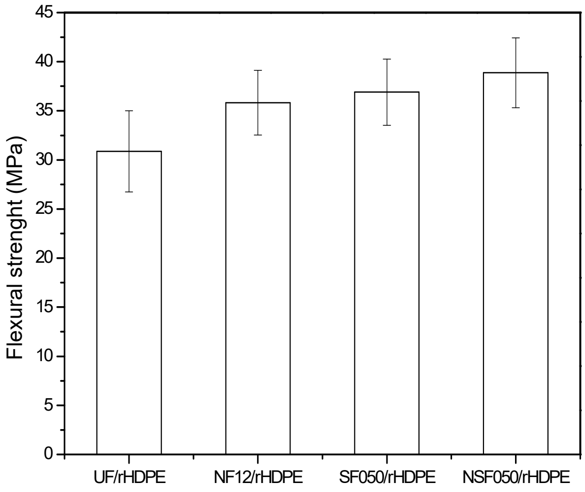

Figure 4. Flexural properties of composites

On the silane treatment, the increase in flexural strength of SF05/rHDPE composites was 19.5%, which was contributed by chemical interaction. Furthermore, a 25.9% increase in flexural strength looked at the NSF05/rHDPE composite which was attributed to the formation of mechanical interlocking and chemical interaction in the fiber-matrix interface zone.

4. Conclusions

Cantala fiber had been modified by alkali, silane, and the combination of both treatments. The treatment effect on fiber properties as well as the composite character had been investigated. On the alkali treatment, the removal of amorphous component caused the enhancement of the cellulose crystallinity. This matter encouraged an increase in tensile strength and tensile modulus of NF12. In contrast, the tensile strength and tensile modulus of SF05 and NSF05 decreased. It was caused by the addition of amorphous material obtained by the link formation between silane molecules and fiber constituent.

In the case of NF12/rHDPE composites, it was found that flexural strength was increased by 16 %. It was predicted due to the formation of mechanical interlocking between the NF12 and rHDPE. The increase in flexural strength of SF05/rHDPE composites of 19.5 % was contributed by chemical interaction. Finally, the 25.9% increment in flexural strength of NSF05/rHDPE composites was attributed to the formation of mechanical interlocking and chemical interaction on the fiber-matrix interface.

Acknowledgement

The research was supported by The Ministry of Research, Technology and Higher Education of the Republic of Indonesia (098/SP2H/LT/DRPM/IV/2017).

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