Evaluation of Physical and Mechanical Properties Composite of Nata de coco Fibers/Resin Filled SiO$_2$, and Al$_2$O$_3$

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Abstract

Nata de coco is a result of fermentation of coconut water by using bacteria Acetobacter xylinum. Fibers contained in the nata de coco is a cellulose, which currently has been applied to various other purposes, such as for the diaphragm transducer, artificial leather, paper mixing materials, carbon films electro-conductive and etc. To obtain a strong fiber material, it is required special treatment, namely by adding other material such as nanoparticles of SiO$_2$, and Al$_2$O$_3$, then combined with various types of resin, so that the composite fiber materials have new properties that are stronger than some metal alloy.

This study has been carried out by making nata de coco fiber and composite fiber-resin-filler, in which the nutrients and pH were varied by selecting the best concentration variation of sugar 2.0% w/v; urea 0.5% w/v and acetic acid 0.3% v/v (pH 3.8). This variation produces a thick fiber of nata de coco about 14.57 mm and wet mass fiber of approximately 595 grams for 700 ml medium of coconut water. From the XRD pattern, the structure of pure nata de coco fiber is cellulose fiber material. The main peak intensity located 2$\theta$ positions around 26° – 26.5°. While for the examination by using SEM-EDX, the filler material has been distributed uniformly in the fiber. The mechanical tests using the Ultimate Tensile Strength is shown that pure nata de coco fiber has tensile strength of 390.39 MPa and young modulus around 11,198 GPa.

1. Introduction

Nata de coco is one of several coconut water potential that most developed in Indonesia. Nata de coco is a result of coconut water fermentation by using the Acetobacter xylinum bacteria. Chemically, the fiber contained in nata de coco is a cellulose fiber, known as bacterial cellulose [Piluharto, B., 2003]. Bacterial celluloses have some advantages such as having a high purity without lignin, pectin, and hemicellulose, which are commonly found in plant cellulose [S, Makoto et al., 2005].

Besides that cellulose fibers or nata de coco fiber produced by Acetobacter xylinum has certain physical properties which different from plant cellulose [Yano, S. et al., 2008]. The unique physical properties of cellulose derived from this bacteria is having a high purity, crystallinity, mechanical strength, and porosity, and also having quite enough capacity to absorb water and easy to get break down [S. Makoto et al., 2005, L.L. Zhou et al., 2007 and Brigid A., D. et al., 2009]. This makes nata de coco fiber potentially to be developed further not only as ingredients of processed foods or beverages, but also can be used for important industries such as manufacturing of the transducer diaphragm for speakers and headphones [K. Watanabe et al., 1995, Iguchi, M et al., 2000, H. Ichikawa et al., 2005 and A.N. Nakagaito et al., 2005], artificial skin to replace skin damaged by fire [Fontana, J.D et al., 1990], membrane separation [M. Takai et al., 1994, J. George, K.V. Ramana, S.N. Sabapathy and A.S. Bring., 2005], mixing materials in paper industry [Shibazaki, H et al., 1994, R. Mormino and H. Bungay., 2003], producing carbon films electro-conductive [Yoshino, K et al., 1991], and materials for biomedical purposes [G. Serafica et al., 2002 and D.A. Schumann et al., 2009].
Based on the physical and mechanical properties of nata de coco fibers, it required a study about the use of cellulose fibers to be used as the basic material of natural fibers which can be used as new composite materials for the manufacture of other products such as prepack board motor or body of car, bullet resistant panels, roof frame, helicopter seat, and etc. In the application for the basic ingredients of such products, nata de coco fiber should have the advantage, both physical properties such as high fiber thickness and density and also mechanical properties such as high young modulus and tensile strength [A.N. Nakagaito et al., 2005].

This study aims to obtain nata de coco fiber using a variety of process conditions such as composition of acetic acid (pH), sugars (sucrose source), and urea (nitrogen source). After that, nata de coco fiber treated with the addition of filler particles such as SiO$_2$, and Al$_2$O$_3$, then nata de coco fiber that has been filled by granules filler, composited by using various types of resins such as epoxy, polyester, and vinyl ester and then testing the other physical and mechanical properties using test equipment such as tensile test, and then analyze with SEM, SEM-EDX, XRD. From these results, the best candidate of composites can be selected.

According to [Tajima et al., 1995], nata de coco composite/SiO$_2$ filler with a specific concentration can increase the elastic modulus three times greater than pure nata de coco fiber (fiber nata de coco pure around 10 GPa), but according to [Yano et al., 2008] nanocomposite with high elastic modulus contain SiO$_2$ nanoparticles (Snowtex 20) below 4% wt in the nata de coco hydrogel. This happens because when the SiO$_2$ filler in fiber composites is greater than that amount, it will cause the reduction of the fiber strength and elasticity as a result of the damage of microfibril ribbons form that had changed in bonding orientation direction (tear) due to an excess of SiO$_2$ filler.

With the addition of filler particles such as SiO$_2$ and Al$_2$O$_3$ in this study, it is expected can increase the strength of nata de coco fiber, therefore it can provide a new contribution of properties to the development of composite materials and also can increase the added value of nata de coco as more useful material.

2. Experiment
2.1 Equipment and Material
The materials used are coconut milk, sugar, urea, acetic acid glacial, Acetobacter xylinum (nata de coco seeds) from Institute Technology of Indonesia, Serpong, Indonesia. SiO$_2$ and Al$_2$O$_3$ filler. Epoxy resins, vinyl ester, and polyester from PT. Justus Kimia Raya, Jakarta, Indonesia. The instrument used are plastic tray size 20x15cm, analytical balance sheet, stainless steel pan, brush, press tools, papers, rubber, and some glassware.

2.2 Preparation of Nata de coco
Coconut water is filtered to get clean from ash and other solid impurities. In one liter of coconut water, add 20 grams of sugar, 3 ml of acetic acid, and 5 grams of urea. Combined these ingredients and then boiled for 5 minutes. Poured the mixture of materials that have been boiled into a plastic tray with a thickness of 1.5 to 2.0 cm, cover tightly with paper, and let them until completely cool.

Filled nata de coco seed as much as 70 ml/700 ml of coconut water medium into a plastic tray containing it. Keep for nine days in a cool and safe (not disturbed and rocked). Coconut water is washed and cleaned with running water until acid was disappeared and the color becomes clear white. The cleaning process is done to prevent the nata become smelly and the fungi growth on it.

2.3 Preparation of Nata de coco Fiber Sheets
Nata de coco slabs dimensions around 20 x 15 cm. There are two stages to get a sheet of nata de coco fiber, cold press at a pressure of 10 tons and then hot press at a pressure of 5 tons with a temperature of 110°C for 30 minutes.

2.4 Nata de coco Fiber Composite
The selected method in making nata de coco fiber composite is hand lay-up method. The advantage of this process is inexpensive and simple process. In addition, this process is also fully compatible with the form of reinforcement (nata de coco fiber) that are already flat.
2.5 Testing and Analizing of Nata de coco Fiber Composite

Physical properties test is done by measuring the thickness, mass, and density of fibers and also percentage of nata de coco wet swelling. Morphological analysis of nata de coco fiber slabs carried out using SEM. SEM-EDX is used to determine the distribution of the filler, as well as to know the percentage of composition and the type of filler material contained in the fiber. XRD analysis is used to identify the type and characteristic of crystal material of nata de coco fiber. Sample of Nata de coco fiber sheets are tested according to ASTM-D638. Tensile test carried out on all samples.

3. Result and Discussion

3.1 Process of Fiber Sheets Formation

Nata de coco production is began with the mixing of various concentrations of sugar as a carbon source, urea as a nitrogen source, and glacial acetic acid as a pH regulator in coconut water medium, then the nourished medium of these nutrients is heated for 5 minutes. This heating serves as a sterilizer so that the medium does not contaminated by undesired bacteria. After it do cooling process at room temperature for 24 hours, then add starter (Acetobacter xylinum) into the coconut water medium. After nine days of fermentation process, it will form a white gel on the surface of the medium.

In the early stages of nata de coco making process, Acetobacter xylinum that have been incorporated into the coconut water medium will get a rapid increase in the number of colonies, then the bacteria that exist in this medium produce large amounts of cellulose fibers with the help of isomerase and polymerase enzymes which are also produced by this bacteria, so that the surface of coconut water medium will look turbid or gel is formed with a higher viscosity than the fluid beneath. As longer gel layer as thicker it and very clearly visible, whereas the amount of fluid in the medium is getting decrease.

According to research done by [Bambang Piluharto et al., 2003 and Kongruang, S et al., 2008], glucose that play a role in the formation of cellulose is glucose in β form so that all glucose in α form will be changed into β form through isomerase enzyme in Acetobacter xylinum. The next stage is glucose binding to other glucose through 1,4 β-glycoside bond. The last stage is polymerization, the formation of cellulose. Polymerization occurs through polymerization enzymes in Acetobacter xylinum.

The reason why some microorganisms (Acetobacter xylinum) make cellulose in a large amount biologically is to maintain its existence in the surface of growth medium, so that the bacteria are still able to obtain sufficient oxygen to do activity and produce cellulose, and also defend themself from unwanted substances such as: dirt, other bacteria, and ultraviolet light [Brigid, A et al., 2009].

3.4 Filler Distribution in Nata de coco Fibers

From the results of SEM-EDX in Figure 2, the SiO₂ filler has been distributed uniformly in nata de coco fiber, with the concentration about 16.06% (Table 1).

Figure 2: Results of fiber morphology by SEM -EDX.
Table 1: Distribution and Composition of Fibers by SEM-EDX.

<table>
<thead>
<tr>
<th>No.</th>
<th>Element</th>
<th>Mass (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Carbon (C)</td>
<td>44.55</td>
</tr>
<tr>
<td>2</td>
<td>Oxygen (O)</td>
<td>35.00</td>
</tr>
<tr>
<td>3</td>
<td>Magnesium (Mg)</td>
<td>0.23</td>
</tr>
<tr>
<td>4</td>
<td>Aluminium (Al)</td>
<td>0.90</td>
</tr>
<tr>
<td>5</td>
<td>Silicate (Si)</td>
<td>16.06</td>
</tr>
<tr>
<td>6</td>
<td>Phosphorus (P)</td>
<td>0.39</td>
</tr>
<tr>
<td>7</td>
<td>Chlor (Cl)</td>
<td>1.13</td>
</tr>
<tr>
<td>8</td>
<td>Kalium (K)</td>
<td>1.71</td>
</tr>
</tbody>
</table>

3.5 XRD Analysis of Nata de coco Fiber

From the XRD results, the nata de coco fibers are crystalline with the highest peak intensity in 2θ position at point between 26 to 26.5°. The intensity position is similar to XRD patterns of cellulose fibers. This shows that nata de coco fiber is crystalline of cellulose fiber.

![Figure 3: The XRD Pattern of Pure Nata de coco Fiber and Nanocomposite Filler Fiber](image)

3.6 Tensile Test Results

- Pure Fiber Tensile Test
  In this observation the fibers that have the highest tensile strength is sampled at various concentration of acetic acid 0.3% v/v; sugar 2.0% w/v; and urea 0.5 % w/v with the value of 390.392 MPa. It proves that the more the number of nata de coco fiber formed, the greater the tensile strength of these fibers. While for the other concentrations has a value of tensile strength around 226.904 - 210.813 MPa respectively, then this sample was selected as a formula for the next process of fiber-resin composite.

- Tensile Strength of Composite Test
  In Figure 4 (a), and (b) about the tensile test results of composite, the highest tensile strength of resin-filler fiber composite is vinyl ester-Al$_2$O$_3$ fiber composite with the tensile strength 21.02 MPa and tensile modulus 0.9 GPa Figure 4 (b). While the lowest of resin filler fiber composite is epoxy-SiO$_2$ fiber composite with tensile strength 4.21 MPa and tensile modulus 0.914 GPa Figure 4 (a).
It can be seen, that for a single sheet of fibers filled with SiO$_2$ and Al$_2$O$_3$ filler, has a certain tensile strength differences when composited with different types of resin, which filler fiber material with vinyl ester resin composite has the highest tensile strength around 19.81 to 21.02 MPa and the best tensile strength of vinyl ester resins of filler is Al$_2$O$_3$ filler 21.02 MPa. This suggests that the interaction between vinyl ester Al$_2$O$_3$ fiber composite molecules is better than the interaction between vinyl ester fiber composite molecule with SiO$_2$ filler. Besides that, mechanically neat vinyl ester resin has a tensile strength higher than other neat resin. It can be seen from Table 2. as the following:

### Table 2: Tensile Test Result of Resin Composite Fiber

<table>
<thead>
<tr>
<th>No</th>
<th>TYPES OF RESIN</th>
<th>Fillers</th>
<th>Value (MPa)</th>
<th>Fillers</th>
<th>Value (MPa)</th>
<th>Fillers</th>
<th>Value (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Epoxy</td>
<td>Neat Epoxy</td>
<td>21.00</td>
<td>Neat Vinyl Ester</td>
<td>29.35</td>
<td>Neat Polyester</td>
<td>25.79</td>
</tr>
<tr>
<td>1</td>
<td>SiO$_2$</td>
<td>4.21</td>
<td>SiO$_2$</td>
<td>19.81</td>
<td>SiO$_2$</td>
<td>16.27</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Al$_2$O$_3$</td>
<td>12.98</td>
<td>Al$_2$O$_3$</td>
<td>21.02</td>
<td>Al$_2$O$_3$</td>
<td>10.07</td>
<td></td>
</tr>
</tbody>
</table>

It can be seen that the neat vinyl ester has a highest tensile strength value among other neat resins, such as epoxy and polyester, which the value of tensile strength of neat vinyl ester resins at 29.35 MPa, while for neat epoxy and polyester resins have tensile strength at 21.00 and 25.79 MPa. This proves that the tensile strength of vinyl ester is higher than epoxy and polyester resin for a single sheet composited.

4. Conclusion

Based on the results the conclusions are as follow, Nata de coco fiber from coconut water with acetic acid 0.3% v/v; sugar 2.0% w/v, and urea 0.5% w/v, is the fiber with the best composition that produces wet fiber thickness 14.57 cm and mass of 595 grams for every 700 ml of coconut water medium. From the mechanical testing, this dry fibers has tensile strength 390.39 MPa.

Preparation of filler composite fibers has been successfully done with immersion method, by immersing the fiber into filler colloidal solution for 2 week. From the morphological analysis using SEM and SEM-EDX, it showed that the filler particles have been well spread on the surface and inside the nata de coco fibers. The XRD tests showed that nata de coco fiber materials are crystals with the highest peak is located at 2θ around the point 26° - 26.5° with the intensity of crystalline peak around 4998.52 au.

In one layer resin-filler fiber composite test, tensile strength values obtained around 4.21 to 21.02 MPa, which the best variation is Al$_2$O$_3$-vinyl ester fiber composite with value of 21.02 MPa and...
the lowest tensile strength is epoxy-SiO₂ fiber composite with value of 4.21 MPa. The tensile strength of composite materials were decreased when compared with the tensile strength of nata de coco pure fiber, because the existence of a liquid resin that contacted with fibers during the process of making fiber composite, therefore the crystallinity of composite fiber decreased as well.

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References