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SYNTHESIS OF NATA DE COCO FIBER COMPOSITE WITH CONDUCTIVE FILLER AS AN ECO-FRIENDLY SEMICONDUCTOR MATERIAL

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ABSTRACT

The electronic industry's need for semiconductor material is increasing each year due to technology's rapid development. Semiconductor material has an electric conductivity of approximately 10^{-8}–10^{3} S/cm, and it is used as an important component in electronic devices. Semiconductor material is generally made of plastic modified with conductive filler. The problem with using semiconductor material is that the discarded components can be plastic waste that requires significant time to degrade; therefore, the synthesis of semiconductor material from natural substances must be observed. One of these natural substances is *nata de coco* fiber modified with a conductive filler. The impregnation method is used in the synthesis of the *nata de coco* fiber composite. The fillers used in this study are ZnO and silica, and the size of the filler particle and the concentration of the filler suspension are used as variations. From the SEM-EDX results, it can be seen that the filler is successfully deposited on the *nata de coco* fiber. Silica filler gives a higher conductivity than ZnO filler because of its lower energy band gap. The highest conductivity result is obtained from the composite impregnated in a 0.3-0.4 μm particle diameter of filler with 3% w/v suspension concentration for three days, producing the conductivity result of 6.95×10^{-6} S/cm for ZnO filler and 10.1×10^{-6} S/cm for silica filler, or about 16 times higher than the conductivity of *nata de coco* fiber.

Keywords: *nata de coco*; Natural fiber composite; Semiconductor material; Silica; ZnO

1. INTRODUCTION

The electronic industry's need for semiconductor material is increasing each year due to technology's rapid development. Most semiconductor material uses a composite made of plastic with silica filler. The plastic used for the composite accounts for 5% of the world's plastic use (Shen, 2010). However, the semiconductor materials that are no longer used can be wasted, and they require significant time to degrade. Therefore, it is critically important to develop an eco-friendly semiconductor material. The synthesis of *nata de coco* fiber composite with a conductive filler can be a solution that will provide economical and environmental benefits.

Previous research has been conducted on *nata de coco* as a fiber in composite materials. The tensile strength of *nata de coco* reaches 316 Mpa, which makes it strong enough to be compared with Kevlar and suitable to be used as a fiber in composite material (Mikrajudin, 2008). In addition, some research has provided insight into modifying a material's semiconductivity. Tjong and Liang (2005) stated that electrical characteristics of an isolator can be modified with the addition of a conductive particle, such as ZnO and silica. The insertion of a biocomposite

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the addition of a conductive particle, such as ZnO and silica. The insertion of a biocomposite from starch into the ZnO filler suspension for three days gives the composite a semiconductor characteristic (Nuryetti, 2012). Based on those previous researches, ZnO and silica are used in this research as the filler to modify the *nata de coco* fiber composite.

In this research, *nata de coco* fiber is impregnated in a filler suspension to obtain the composite material that has a semiconductor characteristic. A matrix will not be used in this research because it would degrade the tensile strength of the composite (Saputra & Darmansyah, 2010). The purpose of this research is to determine the optimum diameter of the filler particle and the concentration of the filler suspension that yields the best composite conductivity.

2. EXPERIMENTAL

2.1. Materials

The raw materials in this study are *Acetobacter xylinum* as the *nata de coco* starter from PT. Daya Agro Mitra Mandiri pure coconut water as the fermentation media, Gulaku brand sugar as the carbon source, Pupuk Indonesia brand urea as the nutrition source for the bacteria, and acetic acid glacial from Merck Millipore as the pH controller in the synthesis of the *nata de coco*. ZnO and silica quartz from Sigma Aldrich will be used as the composite filler.

2.2. Synthesis of *nata de coco*

The synthesis of *nata de coco* begins by filtering the coconut water. After the coconut water is filtered, 15 grams of sugar and 5 grams of urea are added into 1 liter of coconut water. This mixture is then stirred until boiled. Once the mixture is boiling, 3 ml of acetic acid is then added into the mixture. Acetic acid is needed to function as the pH controller, with the *Acetobacter xylinum* growing best in pH 3.5–7.5 (Iguchi & Yamanaka, 2000). This mixture containing coconut water, sugar, urea, and acetic acid becomes the fermentation media. This fermentation media is then poured into the sterilized plastic tray for one day until it reaches room temperature. After one day, 100 ml *nata de coco* starter is added to the media, and the mixture is closed tightly for 10 days until the *nata de coco* is formed.

2.3. Filler Milling

The size of the gap between *nata de coco* fiber is approximately 1.4–1.6 μm (Yano & Nogi, 2005). On the other side, ZnO and silica that are purchased for this study had a diameter of approximately 40 mesh. The size of filler particles need to be reduced by using high energy ball mill in order to achieve the desired diameter. Three different particle diameters are obtained from the milling process and are useful to study the effect of particle diameter to the composite's electric conductivity.

2.4. Filler Particle Diameter

The ZnO and silica particles that went through the milling process are then characterized by using a Particle Size Analyzer (PSA) and the results will be compared to the conductivity results in order to determine the effect of particle diameter on the composite's conductivity.

2.5. Preparation of Filler Suspension

The filler suspension is prepared by mixing the filler with aquadest. This suspension is then stirred with a magnetic stirrer for five minutes. The suspension concentration will influence the filler that can be attached to the composite (Saputra & Darmansyah, 2010). In this study, 1%, 3%, and 6% w/v variations of filler suspension were prepared to determine the effect of suspension concentration on the composite's conductivity.

2.6. Fiber Modification Process

Generally, composite material has an isolator characteristic; therefore, the conductive filler must be used to modify the composite fiber to increase the composite's conductivity until it reaches the range needed for a semiconductor material. This study uses the impregnation method, and the *nata de coco* is impregnated in the filler suspension for three days (Nuryetti, 2012). By this method, the silica particle will enter the gap between the *nata de coco* fibers and will become bonded because of the gravitational force and the formation of a hydrogen bond between the hydroxyl group in the cellulose and the oxygen atom in the filler particle (Jaisai et al., 2012; Soichiro, 2007).

2.7. Synthesis of Composite

The *nata de coco* that has been impregnated in the suspension for three days is then sent to the water removal step, which is performed to produce a composite that only consists of *nata de coco* fiber and filler. The water removal was performed by using a cold press machine with 5 tonnes of pressure and a hot press machine with 5 tonnes pressure in 100^oC for 30 seconds (Saputra & Darmansyah, 2010).

2.8. Composite Morphology and Content

Composite morphology and contents were analyzed by using SEM-EDX which was performed in SEM Laboratory, Department of Metallurgy and Material Engineering, Universitas Indonesia. The SEM machine was a Leo 420i, while the EDX was made by Oxford. SEM results show the morphology of the composite, which is needed to discover the ability of filler particle to attach on the composite. EDX result shows the weight percentage of the filler in the composite. This result will then be compared to the conductivity result.

2.9. Composite Electric Conductivity

A semiconductor is a material that has a conductivity of 10^{-8}–10^{3} S/cm. Semiconductor material is used in electronic components as diodes, transducers, sensors, transistors, and detectors (Kwok, 1995). Electric conductivity is characterized by using an LCR-Meter, and the measurement result is the conductance in 1 volt and 1–100000 Hz frequency. The conductivity is then calculated by using Equation 1:

$$\sigma = G \times \frac{l}{A} \tag{1}$$

where σ is electric conductivity (S/cm); l is the gap between the electrodes (cm), G is conductance (S), and A is the sample area (cm^2). Conductivity analysis for a solid material can be done by using the equation model developed by Jonscher (1983) and Lee et al. (1991), as written in Equations 2 and 3:

$$\sigma = \sigma_0 x\, f^s \tag{2}$$

$$\log \sigma = \log\sigma_0 + s \log f \tag{3}$$

where σ is sample conductivity dependent to the frequency (S/cm), σ_0 is sample conductivity independent to the frequency (S/cm), f is frequency (Hz), and s is the constant (0<s<1). By plotting the log σ in the y axis and log f in the x axis, log σ_0 can be obtained from the intercept of the line to calculate the conductivity independent to the frequency. Composite electric conductivity was measured by an LCR-Meter (Hioki 3532-50) in the Indonesian National Nuclear Energy Agency (BATAN).

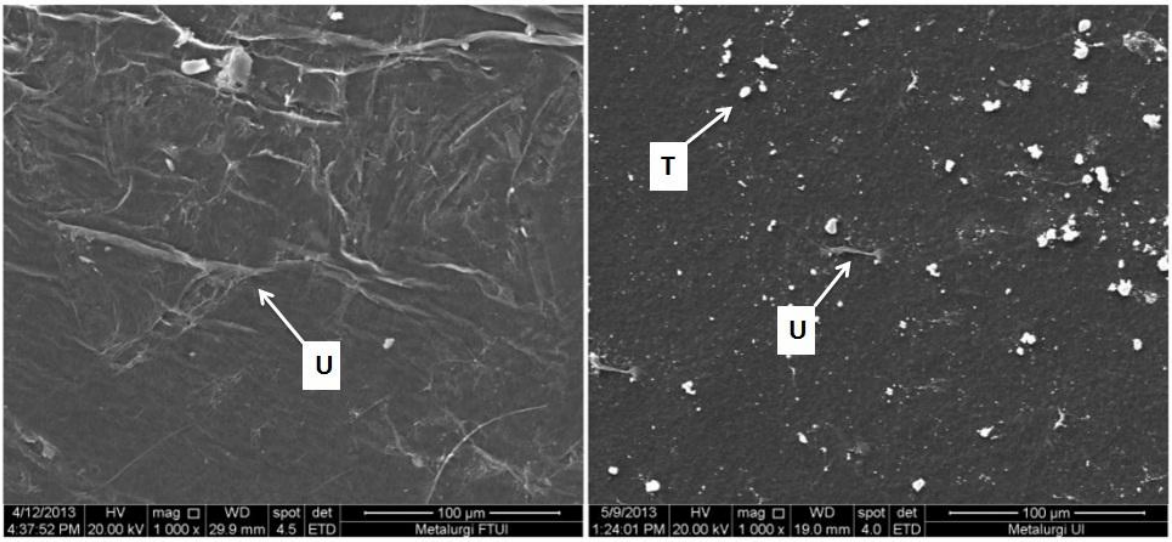
3. RESULTS AND DISCUSSION

3.1. PSA

Fillers that have been through the milling process are characterized using PSA in order to determine the the filler's particle diameter. From the PSA results, three variations of particle diameters were obtained: 0.3–0.4 μm; 0.6–0.7 μm; and 1.4–1.5 μm. A smaller silica particle size cannot be obtained because the milling process is only able to reduce the particle diameter near the nano order (Smallman & Bishop, 1999).

3.2. Morphology

SEM characterization is used to discover the composite's morphology. The SEM composites' results with 1000 times magnification are shown in Figure 1. The part indicated by the U arrow is the *nata de coco* fiber, and the T arrow indicates the filler particle.



(a) (b)

Figure 1 SEM result of : (a) unmodified *nata de coco* fiber; (b) *nata de coco*/ZnO composite

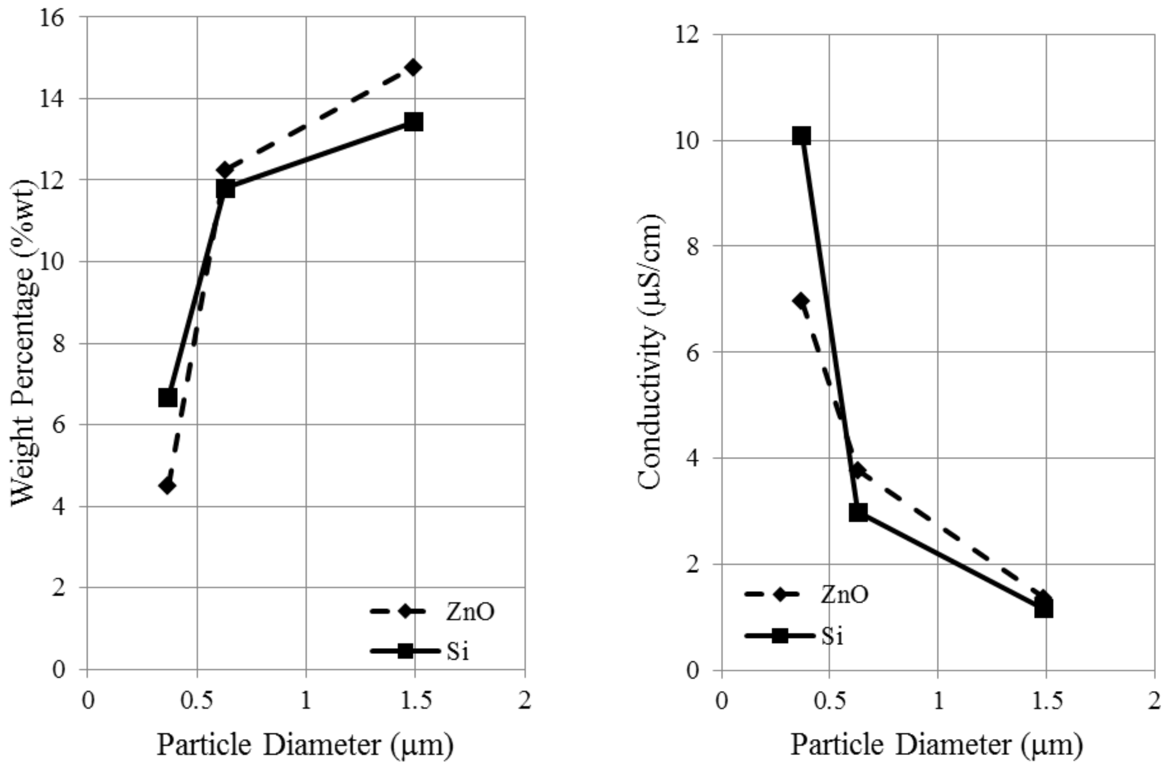
From Figure 1, it can be seen that the filler particles are succesfully attached to the composite; however, the particles are not uniformly attached because there are so many filler particles agglomerated in the composite synthesis process. The small particle has a significant possibility to agglomerate because it has a large surface area that increases the possibility to interact with the other particles (Stanilan, 2013). In addition, it can also be seen that the size of the *nata de coco* fiber may vary because the bacteria activity is difficult to be controlled although the media is the same (Saputra & Darmansyah, 2010; Soichiro, 2007).

3.3. Effect of Particle Diameter on Weight Percentage and Composite Conductivity

The conductivity result for the *nata de coco* fiber without filler is 4.33×10^{-7} S/cm; therefore, the *nata de coco* fiber itself also can be categorized as a semiconductor material (Kwok, 1995). The samples that are compared in order to discover the effect of particle diameter on weight percentage and composite conductivity are impregnated in 3% w/v suspension for three days. The effects of particle diameter on the filler weight percentage and composite conductivity are plotted in Figure 2.

From Figure 2, it can be seen that the bigger particle diameter results in a higher weight percentage of the composite filler. The reason of this result is that for the same surface area, the bigger particle diameter will give a heavier filler particle even though the number of particles attached on the composite is fewer than the composite with smaller particles. The composite with smaller-sized filler particles might have a greater number of particles attached, but since the particles are small, the weight percentage of the filler particle on the composite is less than the composite with the larger particle diameter.

On the other side, the bigger particle diameter resulting in the decrease of composite's conductivity. There are some factors that influence the conductivity of a polymer filled with a conductive filler such as the interparticle length of the filler and the contact area between the filler particles (Clingerman, 1998). Although the smaller diameters produced lower weight percentages in both fillers, the conductivity was higher because the interparticle length is smaller and the contact area is wider.



(a) (b)

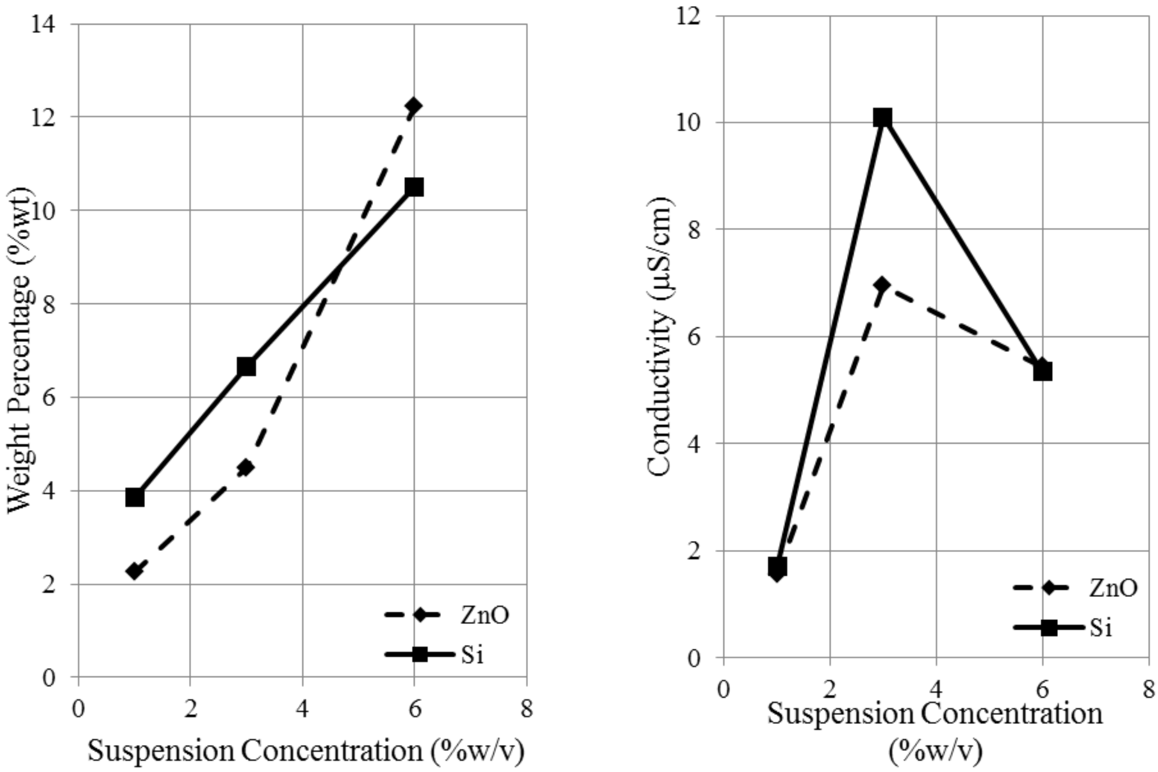
Figure 2 Effect of particle diameter to: (a) EDX result; (b) conductivity result

3.4. Effect of Suspension Concentration on Weight Percentage and Composite Conductivity

The samples compared to determine the effect of suspension concentration on weight percentage and composite conductivity are impregnated in a filler suspension with 0.3–0.4 µm particle diameter for three days. This comparison result can be seen in Figure 3, which shows that higher suspension concentrations result in a higher weight percentage of the composite filler. The reason for this result is that the more particles involved in the interaction with the *nata de coco* fiber, the greater the possibility for particles to be attached on the composite. This is confirmed by the result in Figure 3, where the increase in suspension concentration is followed by the increase of filler weight percentage in the composite, which also means an increase in the number of filler particles attached in the composite.

From Figure 3, it can be seen that the composite conductivity is increasing significantly in 1% and 3% w/v concentrations. This result shows that the higher suspension concentration results in a higher weight percentage of filler particles on the composite. The increase in the number of filler particles attached in the composite increases the conductivity result because the filler particles that can conduct electricity started to form a conductive path.

Meanwhile, in the 6% w/v concentration, the conductivity decreased because the filler had already formed a complete conductive path in the 3% w/v concentration. The addition of filler after the conductive path is completed will only produce a small increase in conductivity or even decrease the conductivity because the conductive path is blocked due to the excessive filler (Clingerman, 1998).



(a) (b)

Figure 3 Effect of suspension concentration to : a) EDX result and ; b) conductivity result

3.5. Comparison Between ZnO and Silica Filler

The comparison between the ZnO filler and the silica filler was performed in order to determine which filler gives the best composite conductivity. *Nata de coco* fiber composite with silica filler tends to produce a higher conductivity result compared to the composite that uses the ZnO filler because the energy band gap of silica is lower than the energy band gap of ZnO. The lower energy band gap, the more conductive the material will be. The energy band gap of silica 1.1–1.2 eV is lower than the energy band gap of ZnO 3.2–3.4 eV (Smallman & Bishop, 1999; Callister, 2007).

4. CONCLUSION

Nata de coco fiber composite modified with ZnO and silica filler were prepared, and the conductivity of the composites were measured. The composites obtained from this study can be categorized as semiconductor materials. The highest conductivity result was obtained from the composite impregnated in 0.3–0.4 μm particle diameter of filler with 3% w/v suspension concentration for three days, which produced conductivity results of 6.95×10^{-6} S/cm for ZnO filler and 10.1×10^{-6} S/cm for silica filler, or about 16 times higher than the conductivity of *nata de coco* fiber.

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