Fabrication of zinc oxide-embedded kapok (Ceiba pentandra) paper

E S Mones¹, M D L Balela¹, C C M Futalan², R D Manalo³, and M U Herrera⁴

¹Department of Mining, Metallurgical, and Materials Engineering, College of Engineering, University of the Philippines Diliman, Quezon City, Philippines 1101
²AECOM Philippines Consultants Corporation, Fort Legend Towers, Bonifacio Global City, Taguig, Metro Manila, Philippines 1634
³Department of Forest Products and Paper Science, College of Forestry, University of the Philippines Los Baños, Laguna, Philippines 4031
⁴Institute of Mathematical Sciences and Physics, College of Arts and Sciences, University of the Philippines Los Baños, Laguna, Philippines 4031

Email: muharrera@up.edu.ph

Abstract. Zinc oxide-embedded kapok (Ceiba pentandra) papers with antimicrobial property were fabricated. Fabrication of papers from kapok fibers was done using chloroform and sodium chlorite treatments. Meanwhile, embedding of zinc oxide particles on the fabricated kapok papers was done using an in situ method. The said method involved soaking the paper in zinc acetate and sodium hydroxide solutions. The method also involved heat treatment of the sample to fasten the formation of zinc oxide particles. X-ray diffraction (XRD) measurement shows the presence of synthesized ZnO particles on the cellulose fibers while scanning electron microscopy (SEM) revealed the flake-like morphology of the embedded ZnO particles. Agar diffusion test shows that the samples have higher antimicrobial effect against Staphylococcus aureus than Escherichia coli.

1. Introduction

Metal oxides [1-8] have attracted attention in the inhibition of bacterial growth. Studies were done to modify and improve the properties of different materials by inclusion of metal oxides [5,9,10]. Embedding of metal oxides on templates or substrates (e.g., metal oxide-glass, metal oxide-paper) can lead to the enhancement of the properties.

Zinc oxide (ZnO) [6-9,11-15] is an n-type oxide semiconductor that has an energy gap of 3.37 eV. Its physical and chemical properties make it useful in diodes, UV sensors, thin films, antimicrobial applications, and photocatalysis.

Paper [5,9], a regularly-used material, is flexible and portable. Different metal oxides were embedded on paper for various applications. Papers are commonly made of fibers from woods. In the view of conserving woods for environmental protection, finding alternative sources of fibers can be explored. Kapok fibers [16] were studied for its potential in papermaking. Kapok fibers are from the fruits of kapok trees (Ceiba pentandra). They have cotton-like appearance and have light-yellowish color. They are fluffy, less dense, and have microtubular structures. Due to its hydrophobicity, kapok fibers are used in oil removal applications. This study aims to modify the properties of handmade...
kapok paper by embedding ZnO particles. Papers have different applications such as template for printing and decorative items, food and material packaging. Embedding of ZnO in paper opens many possible applications wherein the desirable properties of both ZnO and paper are taken advantage.

2. Methodology

2.1. Fabrication of Kapok (Ceiba pentandra) papers

2.1.1. Chloroform treatment. Twelve grams (12g) of Kapok fibers from San Jose del Monte, Bulacan, Philippines and 1.2 L chloroform (CHCl$_3$, J.T. Baker Chemical Co.) were mixed and stirred constantly for 4 h. The fibers were filtered using Buchner funnel while being supported by a rotary vacuum pump (ROTOR VACUUM PUMP SA18 032616 SHIMADZU Corporation) to quicken the filtration. The fibers were washed with methanol (CH$_3$OH, Fisher ChemAlert) and then air dried for at least 12 h.

2.1.2 Sodium chlorite treatment. Thirty millilitres (30mL) of stock solution consisting of 60 mL acetic acid (CH$_3$COOH, MACRON Fine Chemicals), sodium hydroxide solution (1.3 g NaOH per 1 water) and 2 g of dry weight chloroform-treated kapok were mixed in 250 mL Erlenmeyer flask. The Erlenmeyer flasks containing the mixture were placed in hot water bath at 75°C. Three milliliters (3 mL) of 20% sodium chlorite (NaClO$_2$, SIGMA-ALDRICH) solution was added to the mixture and the 250 mL Erlenmeyer flask was swirled afterwards. Another 3 mL of sodium chlorite solution was added after 0.5, 1.0, 1.75, and 2.5 h. After 2.5 h, the mixture was maintained in hot bath for 45 min. The mixture was removed from hot bath and then filtered with the aid of rotary vacuum pump. The sodium chlorite-treated kapok was cleaned using 60 mL chilled distilled water with simultaneous filtration of filtrate. It was further cleaned using four 75 mL 1% acetic acid solution and two 30 mL methanol and then air dried for at least 36 h.

2.1.3 Kapok paper production. One gram (1g) of sodium chlorite-treated kapok was broken down into small pieces and soaked in 299 mL distilled water for 24 h. The slurry was osterized using a commercial blender (Waring Commercial) for 5 min. The slurry underwent filtration in with the aid of rotary vacuum pump. After filtration, the sodium chlorite-treated kapok was slightly air dried for 5 to 10 min. The sample was flattened into sheets using rolling pin and then air dried for at least 24 h.

2.2 Embedding of zinc oxide

Zinc oxide particles were embedded in kapok paper via in situ method. Kapok papers with 1 mm diameter were immersed in 0.75M zinc acetate ((CH$_3$·COO)$_2$Zn·H$_2$O, TECHNO PHARMCHEM HARYANA, 99.5%) solution for 1 hr. The papers were then immersed in 0.5M sodium hydroxide (NaOH, MACRON Fine Chemicals, AR) solution. Afterwards, the papers were heated for 1 min at 50°C on a hot plate (CORNING PC-420D).

2.3 Characterizations

X-ray diffraction measurement was done to determine the composition of the deposited material on the paper. The said measurement was done using XRD SHIMADZU MAXima XRD-7000. Meanwhile, the surface morphology of the sample was observed using scanning electron microscope (SEM, JEOL Ltd., JSM-5310).

Agar disc diffusion was employed to determine the antimicrobial indices of the samples against Escherichia coli and Staphylococcus aureus. The samples have diameter of 10 mm. Microbial suspensions were prepared from 24-hour old culture of the bacteria Escherichia coli (UPCC 1195) and Staphylococcus aureus (UPCC 1143). Pre-poured Nutrient Agar (NA), about 3 mm thick, was inoculated with the respective microbial suspension by swabbing the agar surface. The NA plates were incubated at 35°C and observed after 24 h.
3. Results and Discussion

Zinc oxide (ZnO) was successfully embedded on the kapok paper. Figure 1 shows the XRD pattern of ZnO-embedded kapok paper. The diffraction peaks at 15.8° and 23.1° correspond to (101) and (002) planes of the cellulose fibers (Kapok paper). On the other hand, the diffraction peaks at 31.1°, 32.9°, 36°, and 47.8° correspond to (100), (002), (101), and (102) planes, respectively, of hexagonal wurtzite ZnO.

![XRD pattern of ZnO-embedded kapok paper synthesized via in situ method](image)

Figure 1. XRD pattern of ZnO-embedded kapok paper synthesized via in situ method

Figures 2a and 2b show the SEM images of kapok paper while Figures 2c and 2d show the SEM images of ZnO on kapok paper synthesized using in situ method. As can be observed in Figure 2c and 2d, ZnO with flake-like morphology were distributed on the surface of the paper. In the in-situ method, zinc acetate salts were dissolved in aqueous solutions into ions. The smallness of ions enabled them to penetrate into the pores of the paper. Soaking the treated paper in sodium hydroxide solution enabled the zinc ions to transform to zinc hydroxide. With heating, zinc hydroxide transformed into zinc oxide particles.

Figure 3a and 3b show the ZnO-embedded kapok paper subjected to antimicrobial test against *Escherichia coli* (gram-negative) and *Staphylococcus aureus* (gram-positive), respectively. The clearing zones were measured in millimeters and the average diameters of the clearing zones were calculated. The antimicrobial indexes (AI) were computed using the following formula:

\[
AI = \frac{\text{Diameter of clearing zone}}{\text{Diameter of sample}}
\]

(1)

Table 1 shows the clearing zones and the AI of ZnO-embedded kapok papers for each test organisms. For zinc-based materials, one possible antibacterial action is the generation of reactive oxygen species (ROS). It is possible that the antimicrobial action is more evident in *S. aureus* because this type of organisms has thin cell wall.
Figure 2. SEM images of kapok papers at (a) x1000 and (b) x5000 magnifications and flake-like ZnO at (c) x1000 and (d) x5000 magnifications

Table 1. Average clearing zones and antimicrobial indices of ZnO-embedded kapok papers tested against *E. coli* and *S. aureus*

| Test organism                  | Average clearing zones (mm) | Antimicrobial index (AI) |
|--------------------------------|-----------------------------|--------------------------|
| *Escherichia coli* (gram negative) | 10.67                       | 0.1                      |
| *Staphylococcus aureus* (gram positive) | 15.33                       | 0.5                      |

Figure 3. ZnO-embedded kapok papers in agar disc diffusion test against (a) *E. coli* and (b) *S. aureus*

4. Summary and Conclusion

ZnO-embedded kapok papers were successfully fabricated. Kapok papers were fabricated from Kapok fibers using chloroform and sodium chlorite treatments. Meanwhile, zinc oxide particles were embedded in the fabricated kapok paper using *in situ* technique. XRD measurements show presence of
ZnO particles on kapok paper. Moreover, SEM shows the flake-like morphology of the embedded ZnO. In addition, the antimicrobial activity of the ZnO-embedded paper showed higher antimicrobial index against gram-positive \textit{S. aureus} than gram-negative \textit{E. coli}.

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