**UNIVERSITI PUTRA MALAYSIA**

# *SIMULTANEOUS SYNTHESIS AND INCORPORATION OF ZINC OXIDE PARTICLES IN BAMBOO PULP THROUGH CHEMICAL AND BIOLOGICAL METHODS FOR ANTIMICROBIAL PAPER*

**ZAKIAH BINTI SOBRI**

**IPTPH 2021 1**

# SIMULTANEOUS SYNTHESIS AND INCORPORATION OF ZINC OXIDE PARTICLES IN BAMBOO PULP THROUGH CHEMICAL AND BIOLOGICAL METHODS FOR ANTIMICROBIAL PAPER

By

**ZAKIAH BINTI SOBRI**



**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science**

**November 2019**

# DEDICATION

***Bismillahirrahmanirrahim***

***To my beloved parent, family, teachers and friends***

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

**SIMULTANEOUS SYNTHESIS AND INCORPORATION OF ZINC OXIDE PARTICLES IN BAMBOO PULP THROUGH CHEMICAL AND BIOLOGICAL METHODS FOR ANTIMICROBIAL PAPER**

By

**ZAKIAH BINTI SOBRI**

**November 2019**

**Chairman : Associate Professor Edi Syams bin Zainudin, PhD**
**Institute : Tropical Forestry and Forest Products**

Metal oxide nanoparticles such as zinc oxide have been recognized for its potential use in health-related and packaging application. This inorganic compound can be applied for antimicrobial paper. The objectives of this study were to synthesized and incorporate zinc oxide particles simultaneously in bamboo pulp and subsequently used for antimicrobial paper. The effect of pulp type and synthesis methods on the formation of zinc oxide-pulp are evaluated and characterized by Field Emission Scanning Electron Microscope and Scanning Electron Microscope with Energy Dispersive X-Ray Analysis (FESEM-EDX/SEM-EDX), Fourier Transform Infrared Spectroscopy (FTIR) and X-Ray Diffraction (XRD). The morphology of chemical synthesized zinc oxide-pulp appeared in the range of 64.1 nm to 87.5 nm while biosynthesized zinc oxide-pulp size range from 0.33 µm to 2.00 µm with spherical, rod and hexagonal shapes. Biosynthesized zinc oxide-pulp showed higher intensity and a broader band of FTIR spectrum at 400 $cm^{-1}$ to 600 $cm^{-1}$ indicated less impurity content compare to chemical synthesized zinc oxide-pulp. Narrow and sharp peaks at 20° of 2$\boldsymbol{\theta}$ of XRD suggested that more crystalline zinc oxide-pulp is formed by biological method. The antimicrobial property of zinc oxide-pulp has demonstrated stronger antimicrobial activities against *Staphylococcus aureus* (ATCC 43300), *Salmonella choleraesuis* (ATCC 10708) and *Escherichia coli* (ATCC 25922) on biosynthesized zinc oxide-pulp samples compared to chemical synthesized zinc oxide-pulp. Unbleached zinc oxide-pulp has stronger antimicrobial activity than bleached zinc oxide-pulp and significant relationships among the parameters on antimicrobial properties are presented by ANOVA analysis suggested that optimum temperature for biosynthesis zinc oxide-pulp is 70℃ with 0.5 M and 0.7 M of zinc chloride with 100 ml of algae extract. Reduction of mechanical property of zinc oxide-paper developed via chemical method is because of the interruption made by zinc oxide particles at bonded area of fibre.
Keywords: zinc oxide, pulp, bamboo, biosynthesis, chemical

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

**SINTESIS SERENTAK DAN PENGGABUNGAN ZARAH ZINK OKSIDA DALAM PULPA BULUH MELALUI KAEDAH KIMIA DAN BIOLOGI UNTUK KERTAS ANTIMIKROB**

Oleh

**ZAKIAH BINTI SOBRI**

**November 2019**

**Pengerusi : Profesor Madya Edi Syams bin Zainudin, PhD**
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Logam oksida partikel nano seperti zink oksida dikenali dengan potensinya dalam penggunaan pembungkusan dan aplikasi dalam bidang perubatan. Kompoun inorganic ini dapat diaplikasi sebagai kertas antimikrob. Kajian ini bertujuan sintesis dan menggabungkan partikel zink oksida secara serentak dalam pulpa daripada buluh dan seterusnya digunakan sebagai kertas antimikrob. Kesan jenis pulpa dan kaedah-kaedah sintesis ke atas pembentukkan pulpa-zink oksida telah dinilai dan dicirikan dengan Mikroskop Elektron Pengimbasan Pelepasan Medan/Mikroskop Elektron Pengimbasan dengan Sinaran Dispersif Tenaga (FESEM-EDX/SEM-EDX), Fourier Ubah Inframerah (FTIR) dan Difraksi Sinar-X (XRD). Morfologi pulpa-zink oksida disintesis kimia bersaiz 64.1 nm ke 87.5 nm manakala pulpa-zink oksida biosintesis bersaiz 0.33 μm ke 2.00 μm dengan bentuk-bentuk sfera, rod dan hexagon. Pulpa-zink oksida biosintesis menunjukkan intensity yang lebih tinggi dan spectrum FTIR yang lebih lebar pada 400 $cm^{-1}$ ke 600 $cm^{-1}$ menunjukkan kurang campuran berbanding pulp-zink oksida sintesis kimia. Puncak spektra XRD yang kecil dan tinggi pada 20° of 2**θ** telah membuktikan lebih banyak zink oksida kristal yang disintesis melalui kaedah biologi daripada kaedah kimia. Ciri-ciri antimikrob pulpa-zink oksida telah menunjukkan aktiviti antikrob yang lebih kuat dihasilkan daripada pulpa-zink oksida biosintesis berbanding pulpa-zink oksida sintesis kimia untuk melawan bakteria *Staphylococcus aureus* (ATCC 43300), *Salmonella choleraesuis* (ATCC 10708) dan *Escherichia coli* (ATCC 25922). Pulp-zink oksida *unbleached* mempunyai aktiviti antimikrob yang lebih kuat berbanding pulpa-zink oksida *bleached* dan melalui hubungan yang ketara di antara parameter pada sifat antimikrobial yang ditunjukkan oleh analisis ANOVA dapat disimpulkan suhu optimum bagi pulpa-zink oksida biosintesis adalah 70℃ dengan kepekatan zink klorida 0.5 M dan 0.7 M, serta 100 ml ekstrak alga. Penurunan ciri-ciri mekanikal kertas-zink oksida yang dihasilkan daripada kaedah kimia adalah disebabkan oleh kehadiran partikel zink oksida yang mengganggu kawasan pembentukan ikatan serat.


Kata kunci: zink oksida, pulpa, buluh, biosintesis, hidroterma

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This thesis was submitted to Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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I hereby confirm that:

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## LIST OF ABBREVIATIONS

| | |
|---|---|
| AOX | Absorbable Organic Halogens |
| ASAE | Alkaline Sulphite with Anthraquinone and Ethanol |
| ASAM | Alkaline Sulphite with Anthraquinone and Methanol |
| AS-AQ | Alkaline Sulphite with Anthraquinone |
| BOD | Biochemical Oxygen Demand |
| DTMS | Dodecyltrimethoxysilane |
| ECF | Elemental Chlorine-free |
| EDX | Energy Dispersive X-Ray |
| FESEM | Field Emission Scanning Electron Microscopy |
| FTIR | Fourier Transform Infrared Spectroscopy |
| PLA | Polyactic acid |
| ROS | Reactive Oxygen Species |
| SEM | Scanning Electron Microscopy |
| TAPPI | Technical Association for The Pulp, Paper and Converting Industry |
| TCF | Total Chlorine-free |
| TSA | Titanium dioxide/Sodium alginate |
| XRD | X-Ray Diffraction Analysis |

# CHAPTER 1

# INTRODUCTION

## 1.1 Background

Chemicals have been used as antimicrobial agents in textiles, art preservation, and food packaging plastics or paper. An antimicrobial property is important for paper-based products such as books or documents that are prone to microbe problems due to the uncontrolled environment. Titanium dioxide-sodium alginate nanocomposite modified paper has antimicrobial activity and is suitable for hygienic applications and food packaging (Abdel *et al.*, 2016). Paper also can be coated with biosynthesized antimicrobial agents like titanium dioxide, zinc oxide and silver. Gogoi *et al.*, (2006) and Xu *et al.*, (2004) studied the size of particles of such agents induced the antimicrobial activities.

Zinc oxide is a type of metal oxide that has been used in wide applications from rubber industries to electronic technology. Zinc oxide is usually used as filler and activators in the rubber compounds to improve thermal conductivity and retain high electrical resistance of silicon rubber. Zinc oxide is applied in the paper to enable optoelectronic paper for printing or light operating source of the electronic systems. For instance, tetrapodal zinc oxide microstructures have been used to produce appropriate photosensor to measure the short light exposures (Sandberg *et al.*, 2016). In dentistry, dental paste used for temporary filling is made up of zinc oxide. On the other hand, zinc oxide is a good component to be used as ultraviolet (UV)-blocking textile due to its good absorption property that promote self-cleaning.

Zinc oxides also possesses antimicrobial properties as it has been used in pharmaceutical and cosmetic industries for disinfecting and antibacterial properties in medicine and any anti-inflammation products such as cream and sunscreen. Vijayakumar *et al.*, (2018) demonstrated the antimicrobial activity of zinc oxide particles against bacteria and fungus. Furthermore, Jaisai *et al.*, (2012) also studied the modified paper with zinc oxide particles produced through deep coating technique intended for the use of wallpaper, cleaning tissue, facemask and writing papers also possess antimicrobial properties.

## 1.2 Problem statement

Coating technique is mostly used to protect the paper from microorganisms or water with a layer of antimicrobial and hydrophobic materials. The other solution is by growing metal oxide on the paper during the synthesis process. Metal oxide can be synthesized via chemical or bio-technique; using natural resources such as plants, algae and animals (Kim & Venkatesan, 2013).

Liu, *et al*., (2016), reported that 30 min of immersing paper sheet in grafted dialdehyde starch with a modifier, guanidine hydrochloride can improve paper strength and display strong absorption antimicrobial activity against *E. coli* and *Staphylococcus aureus*. Similar results were also obtained by Kamel, (2012) that used immersion and drying techniques with grafted paper sheet to deposit silver nanoparticles via *in-situ* reduction of silver nitrate. As a result, silver nanoparticles had been shown to successfully deposited on the uneven surface of cellulose. On the other hand, Abdel *et al*., (2016), studied the optimum ratio of titanium dioxide/sodium alginate nanocomposite (TSA) to be added on paper and pressed with a hydraulic press to deposit the TSA before drying with a rotating cylinder. The deposition is successfully obtained at 7% and 20% of TSA with the diameter of $TiO_2$ nanospheres formed at more than 73 nm.

However, regarding the use of medical appliances and food packaging that may be exposed to water, oil, blood or any medicinal liquids, there is a possibility of zinc oxide removal from the surface of the paper. Hence, growing metal oxide in pulp could reduce the potential coating to be rubbed off or destroyed as the wanted materials are placed as finishing in the papermaking process. Okyay *et al*., (2015) has encouraged sonochemical growth since the method showed positive outcomes of growing zinc oxide on pulp as demonstrated on glass slides when viewed by scanning electron microscope (SEM) and energy dispersive x-ray (EDX). Furthermore, the biological method sounds more environmentally friendly and cost-efficient, particularly in the present era where knowledge of environmental care is important. Therefore, this study was carried out to prepare and characterize the zinc oxide particles using both chemical and biosynthesis methods.

## 1.3 Research Hypothesis

Bamboo pulp incorporated with zinc oxide conducted via in situ approaches were chosen to save time by using a single-step for zinc oxide production. Two synthesis methods, namely chemical and biological methods were compared to evaluate the properties of zinc oxide particles on bamboo pulp and antimicrobial properties of zinc oxide-pulp. A preliminary study of the biological method was introduced with few parameters such as amount of algae extract, temperature and concentration of precursor. The biological method was expected to produce good properties of zinc oxide particles and good antimicrobial properties similar to or better than chemical methods. Hence, characterization of zinc oxide-pulp was determined in terms of morphology, chemical properties and antimicrobial properties.

## 1.4 Research Aim and Objectives

General objective: To synthesize and incorporate zinc oxide particles simultaneously in bamboo pulp and subsequently used for antimicrobial paper.

Specific objectives:

a) To evaluate the effect of pulp type and synthesis methods on the formation and characteristics of zinc oxide-pulp.
b) To determine the antimicrobial and mechanical properties of zinc oxide paper developed via chemical and biological methods.

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