UNIVERSITI PUTRA MALAYSIA

SYNTHESIS AND CHARACTERISATION OF COPPER OXIDE DOPED ZINC OXIDE NANOPHOTOCATALYST AND ITS PHOTOCATALYTIC ABILITY IN METHYL ORANGE DYE DEGRADATION

NORZAFIRAH BINTI RAZALI

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NORZAFIRAH BINTI RAZALI

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By

NORZAFIRAH BT RAZALI

August 2011

Chairman : Associate Prof. Abdul Halim Bin Abdullah, PhD
Faculty : Science

CuO-doped ZnO (CuO-ZnO) nanophotocatalyst was synthesized by stirring CuO and ZnO in absolute methanol solution overnight, at ambient pressure and temperature. CuO was initially produced from four different precursors namely copper acetate, copper sulphate, copper chloride, and copper nitrate while the precursor of ZnO was produced from zinc acetate. Effects of different precursors to the properties of CuO (CA, CS, CCl, CN) were investigated. Next, the produced CuOs were doped onto ZnO (CZA, CZS, CZCl, CZN) and the properties were studied. Thermogravimetry analysis (TGA), X-ray Diffraction (XRD), Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), X-Ray Fluorescence (XRF), Atomic Absorption Spectroscopy (AAS), surface area and pore distribution...
(BET and BJH), and Zeta potential were used to characterize the synthesized ZnO, CuO and CuO-ZnO nanophotocatalysts. In photocatalysis, the ability of ZnO, CuO and CuO-ZnO nanophotocatalysts in decolorizing methyl orange (MO) dye were evaluated using UV-Vis spectroscopy while the degradation process was determined through Total Organic Carbon (TOC) analysis.

Interaction between intermediates and by-products during the synthesis process of CuO affected the crystallinity, morphology and particle size of synthesized CuOs. Although no significant changes in most of the physical properties of ZnO was observed, the introduction of CuO had improved the photoactivity of ZnO. This attributed to the enhancement in surface area of the CuO-ZnO photocatalyst and the ability of CuO to act as electron acceptor that prolongs charge separation which resulted in a better photodegradation process. Among the prepared CuO-ZnO nanophotocatalysts, CZN (CuO produced from copper nitrate) exhibited the highest rate of methyl orange photodegradation.

In order to determine the optimum % of CuO to be doped on ZnO, 0.5 – 2.0 % CuO-ZnO photocatalyst were synthesized using CZN. No significant changes in the properties of ZnO as the % of CuO doped were varied. In photodegrading MO, 1.0% of CuO-ZnO nanophotocatalyst showed the highest degradation rate. The optimum condition in degradation of MO was obtained at dye concentration of 30 ppm and pH of 7 by using 1.5g of 1.0% of CuO-ZnO photocatalyst. The photoactivity of CuO-ZnO was maintained until fifth cycle in the study of reusability. 1.0% of CuO had significantly improved the photoactivity of bare ZnO under visible-light irradiation. The potential of 1.0% CuO-ZnO nanophotocatalyst in degrading real dye-containing industrial
wastewater from one of textile factories at Melaka was evaluated. From the TOC analysis, 22% of total organic carbon in the real wastewater was degraded within 60 min.
Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Master Sains

SINTESIS DAN PENCIRIAN NANO-FOTOMANGKIN ZINK OKSIDA DIDOP DENGAN KUPRUM OKSIDA DAN KEMAMPUAN FOTOPEMANGKINANNYA DALAM MENDEGRADASI PEWARNA METIL OREN

Oleh

NORZAFIRAH BT RAZALI

Ogos 2011

Pengerusi : Prof. Madya Abdul Halim Bin Abdullah, PhD

Fakulti : Sains

Nanofotomangkin ZnO yang didop dengan CuO doped ZnO (CuO-ZnO) disintesis dengan mengacau serbuk CuO dan ZnO di dalam larutan metanol mutlak semalaman pada suhu dan tekanan bilik. CuO yang digunakan disintesis daripada prekursor berbeza iaitu kuprum sulfat, kuprum nitrat, kuprum klorida dan kuprum asetat manakala ZnO dihasilkan daripada zink asetat. Pengaruh pelbagai prekursor terhadap sifat CuO telah dikaji. Selanjutnya, CuO yang dihasilkan didopkan pada ZnO dan sifatnya diselidiki. ZnO, CuO dan CuO-ZnO nano-fotomangkin yang terhasil dikaji dengan menggunakan analisis termogravimetri (TGA), pembelauan sinar X (XRD), mikroskop pengimbas electron (SEM), mikroskop pembiasan elektron (TEM), sinar-X
berpendarfluor (XRF), analisa luas permukaan dan taburan liang pori, jerapan Atom Spektrofotometer (AAS), pengubah inframerah fourier (FTIR) dan analisa potensi zeta. Dalam kajian fotopemangkinan, kemampuan ZnO, CuO dan CuO-ZnO dalam menyahwarna pewarna metal jingga dimulai dengan menggunakan UV-vis spektroskopi dan proses degradasi ditentukan melalui analisa jumlah karbon organic (TOC). Interaksi antara bahan pengantara dengan hasil sampingan telah mempengaruhi kehabluran, morfologi dan saiz partikel CuO yang disintesis. Walaupun tiada perubahan yang ketara pada hampir kesemua sifat fizikal ZnO, kehadiran CuO telah meningkatkan fotoaktiviti ZnO. Ini adalah disebabkan oleh penambahan luas permukaan dan kemampuan fotomangkin CuO-ZnO. CuO berperanan sebagai penerima elektron yang memanjangkan masa pemisahan cas yang menghasilkan fotodegradasi yang lebih baik. Antara CuO-ZnO fotomangkin yang disediakan, CZN (CuO yang terhasil daripada kuprum nitrat) menunjukkan fotodegradasi metal jingga yang paling tinggi.

Dalam menentukan % CuO yang optimum untuk didop ke atas ZnO, 0.5 – 2.0 % CuO-ZnO fotomangkin telah disintesis menggunakan CZN. Tiada perubahan yang ketara dikesan melalui analisa pada sifat-sifat asal ZnO setelah didop CuO pada peratusan berbeza. Dalam fotodegradasi MO, 1.0% CuO-ZnO fotomangkin menunjukkan tahap degradasi tertinggi. Keadaan optimum dalam degradasi MO diperolehi pada 30 ppm kepekatan pewarna dan pH 7 dengan menggunakan 1.5g 1.0% CuO-ZnO fotomangkin. Dalam kajian penggunaan semula mangkin, fotoaktiviti CuO-ZnO adalah kekal sehingga kitaran ke-5. 1.0% CuO-ZnO juga mempamerkan peningkatan yang mendadak dalam peratusan degradasi dibawah sinaran cahaya nampak berbanding dengan ZnO. Potensi 1.0% CuO-ZnO fotokatalis dalam mendegradasi air buangan kilang dari salah sebuah
kilang di Melaka telah dinilai. Daripada analisa TOC, 22% dari jumlah karbon organic dalam air buangan tersebut telah didegradasikan dalam tempoh 60 minit.
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I certify that a Thesis Examination Committee has met on 18th August 2011 to conduct the final examination of Norzafirah binti Razali on her Master of Science thesis entitled “Synthesis and characterisation of Copper oxide doped Zinc oxide nanophotocatalyst and its photocatalytic ability in methyl orange dye degradation” in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The committee recommends that the student be awarded the Master of Science.

Members of the Examination Committee were as follows:

Mohamed Ibrahim bin Mohamed Tahir, PhD
Doctor
Department of Chemistry
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Mohd Zobir bin Hussien, PhD
Professor
Department of Chemistry
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Tinia Idaty binti Mohd Ghazi, PhD
Doctor
Department of Chemical & Environmental Engineering
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Zainab binti Ramli, PhD
Associate Professor
Department of Chemistry
Faculty of Science
Universiti Teknologi Malaysia
(External Examiner)

SEOW HENG FONG, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

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

**Profesor Madya Abdul Halim bin Abdullah**  
Faculty of Science  
Universiti Putra Malaysia  
(Chairman)

**Profesor Md Jelas bin Haron**  
Faculty of Science  
Universiti Putra Malaysia  
(Member)

---

**BUJANG BIN KIM HUAT, PhD**  
Professor and Dean  
School of Graduate Studies  
Universiti Putra Malaysia

Date:
DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

NORZAFIRAH BINTI RAZALI

Date : 18th August 2011
TABLE OF CONTENTS

ABSTRACT ii
ABSTRAK v
ACKNOWLEDGEMENT viii
APPROVAL ix
DECLARATION xi
LIST OF EQUATIONS xv
LIST OF FIGURES xvi
LIST OF TABLES xx
LIST OF ABBREVIATIONS xxii

CHAPTER

1 INTRODUCTION
1.1 Background 1
1.2 Problem of statement 7
1.3 Objectives of study 9

2 LITERATURE REVIEWS
2.1 Advanced Oxidation Processes (AOPs) 10
2.2 Photocatalysis 11
2.3 Semiconductor photocatalyst 14
2.4 ZnO photocatalyst 15
2.5 Photocatalysis efficiency 29
2.6 CuO-doped ZnO photocatalyst 40

3 METHODOLOGY AND MATERIALS
3.1 Materials 44
3.2 Methodology
3.2.1 Preparation of ZnO and CuO nanoparticles 44
3.2.2 Preparation of CuO-ZnO nanophotocatalyst 45
3.3 Characterization
3.3.1 Thermogravimetry analysis (TGA) 46
3.3.2 X-Ray Diffraction (XRD) 47
3.3.3 Fourier Transform Infrared Spectroscopy (FTIR) 47
3.3.4 X-Ray Fluorescence (XRF) 47
3.3.5 Scanning Electron Microscopy (SEM) 48
3.3.6 Transmission Electron Microscopy (TEM) 48
3.3.7 Surface Area Measurement and Porosity 48
3.3.8 Band gap determination 49
3.3.9 Zeta potential analysis 49
3.4 Photocatalysis 49
3.4.1 Effect of various percentage of CuO 51
3.4.2 Effect of operating parameter
3.4.2.1 Effect of photocatalyst loading 51
3.4.2.2 Effect of MO initial concentration 51
3.4.2.3 Effect of pH 52
3.4.2.4 Effect of light 52
3.4.3 Reusability test 53
3.4.4 Test on real wastewater 53
3.5 Analysis 53
3.5.1 UV-Vis spectroscopy 53
3.5.2 Total Organic Carbon (TOC) analysis 54

4 RESULTS AND DISCUSSION
4.1 Characterization of CuO 56
4.1.1 Thermogravimetric analysis 56
4.1.2 X-Ray Diffractometry (XRD) 57
4.1.3 Fourier Transform Infrared Spectroscopy (FTIR) 61
4.1.4 Scanning Electron Microscopy (SEM) 64
4.1.5 Transmission electron microscopy (TEM) 68
4.1.6 Pores and Surface area analysis 70
4.1.7 Photocatalysis 73
4.2 Characterization of 0.5% CuO-ZnO photocatalyst 75
4.2.1 X-Ray Diffraction (XRD) 75
4.2.2 X-Ray Fluorescence and BET Surface Area 77
4.2.3 Fourier Transform Infrared Spectroscopy (FTIR) 80
4.2.4 Band gap measurement 81
4.2.5 Scanning Electron Microscopy 83
4.2.6 Transmission Electron Microscopy 86
4.2.7 Photocatalytic activity 88
4.3 Characterization of 0.5 – 2.0% CuO-ZnO photocatalysts 93
4.3.1 Crystallinity, crystallite size, elemental analysis and BET surface area 93
4.3.2 Band gap determination 97
4.3.3 Morphology and particle size distribution 98
4.4 Photocatalysis 99
4.4.1 Effects of varying percentages of CuO on the photoactivity of nanophotocatalyst 103
4.4.2 Effects of mass loading of nanophotocatalyst 108
4.4.3 Effects of methyl orange dye initial concentration 110
4.4.4 Effect of pH on the photoactivity of nanophotocatalyst 113
4.4.5 Removal of methyl orange dye under visible light illumination 116
4.4.6 Mineralization of methyl orange dye 119
4.4.7 Photocatalysis on real industrial wastewater 121
4.4.8 Reusability test of 1.0% CuO-ZnO nanophotocatalyst 122

5 CONCLUSION 125
RECOMMENDATION AND SUGGESTION 127