# HIGH TRANSMITTANCE-n-$TiO_2$/ZnO BILAYER DEPOSITED BY SOL-GEL SPIN COATING METHOD FOR ELECTRODEPOSITED-p-$Cu_2O$ BASED HETEROJUNCTION THIN FILM SOLAR CELL

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## DEDICATION

*Every challenging work need self-efforts as well as guidance of elders especially those who very close to our hearts. My humble effort I dedicate to*

*My dearest husband,*
*Muhammad Sallehudin Bin Khalid,*
*Thank you for allowing me to pursue my dream.*

*My sweetheart daughter,*
*Nur Aisyah Wafiy Binti Muhammad Sallehudin,*
*Thank you for being the strength of Ummi's heart*

*My beloved mother and father,*
*Rahamah Binti Sumani and Mohamad Arifin Bin Sirat*
*For affection, love, encouragement and prays of days and nights,*
*Always there when thick and thin,*
*And this is for you, Mak and Abah.*

*Alhamdulillah. Thank you.*

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# ABSTRACT

Metal oxide semiconductor heterojunction are gaining interest in current fundamental photovoltaic research. Copper oxide ($Cu_2O$) based heterojunction which consists of p-$Cu_2O$ and n-Titanium dioxide ($TiO_2$) have been spotted as a potential window and absorbing layer, respectively. However, n-$TiO_2$ suffers low utilization in solar spectrum and high recombination rate of electron and holes. By coupling $TiO_2$ and Zinc oxide (ZnO) thin film, the layers are known as n-$TiO_2$/ZnO bilayer thin film which can enhance utilization of solar spectrum as a window layer. Due to low lattice mismatch between ZnO and $Cu_2O$, high crystal structure with (002)ZnO/(111)$Cu_2O$ preferred orientation plane is essential to improve the heterointerface layer. Both combination layers exhibited a similar atomic arrangement at the interface due to the crystal structure of n-ZnO and p-$Cu_2O$ are hexagonal wurtzite and cubic, respectively. Herein, n-$TiO_2$/ZnO/p-$Cu_2O$ heterojunction thin film was successfully fabricated onto FTO substrate by using sol-gel spin coating and electrodeposition method. Annealing treatment affected process in fabricating n-$TiO_2$/ZnO bilayer thin film with different annealing temperature and time. Meanwhile, cyclic voltammetry was executed to obtain the most optimized parameter before p-$Cu_2O$ stacking onto n-$TiO_2$/ZnO bilayer thin film with different bath temperature and deposition time. Based on the findings, the preferred (002)-ZnO orientation plane of thin film appeared at 34.28º in n-$TiO_2$/ZnO bilayer thin film when annealed for 2 hours at 500 ℃. High transmittance of n-$TiO_2$/ZnO bilayer thin film was achieved up to 80% at the edge of the visible light spectrum in the range of 450-300 nm. High crystal structure and absorbance spectrum of p-$Cu_2O$ as absorbing layer was observed at bath temperature 40 ℃ for 1.5 hours. P-n junction was successfully formed as indicated by significant electrical rectification properties with conversion efficiency of 0.0615%. The results prove the homogeneity, high transmittance and crystallinity of n-$TiO_2$/ZnO/p-$Cu_2O$ heterojunction thin film aside from enhancing the surface structure and atomic arrangement at its heterointerface which will be beneficial for solar cell application.

# ABSTRAK

Semikonduktor oksida logam hetero-simpang kini menjadi minat dalam asas penyelidikan fotovolta. Kuprum oksida ($Cu_2O$) hetero-simpang terdiri dari p-$Cu_2O$ dan n-Titanium dioksida ($TiO_2$) berpotensi menjadi lapisan tingkap and penyerap. Walaupun begitu, n-$TiO_2$ mengalami kekurangan penggunaan spektrum suria dan tinggi kadar penggabungan semula elektron dan lubang. Penggabungan $TiO_2$ dan Zink oksida (ZnO) saput nipis, lapisan dikenali sebagai n-$TiO_2$/ZnO dwilapisan saput nipis mampu meningkatkan penggunaan spektrum suria sebagai lapisan tingkap. Disebabkan tiada kesepadanan kekisi antara ZnO dan $Cu_2O$ rendah, struktur hablur yang tinggi dengan satah orientasi pilihan (002)ZnO/(111)$Cu_2O$ adalah penting untuk meningkatkan hetero-antara muka. Gabungan dua lapisan itu mempamerkan susunan atom yang serupa kerana struktur hablur n-ZnO adalah wurtzit heksagon dan p-$Cu_2O$ adalah kubus. Di sini, n-$TiO_2$/ZnO/p-$Cu_2O$ hetero-simpang saput nipis telah berjaya difabrik pada substrat FTO dengan menggunakan kaedah salutan mejam sol-gel dan elektromendapan. Penyepuhlindapan berperanan menghasilkan n-$TiO_2$/ZnO dwilapisan saput nipis dengan suhu dan masa penyepuhlindapan yang berbeza. Sementara itu, voltammetri berkitar dilaksanakan untuk mendapatkan parameter yang paling sesuai sebelum p-$Cu_2O$ difabrik pada n-$TiO_2$/ZnO dwilapisan saput nipis dengan suhu dan masa pemendapan berbeza. Berdasarkan penemuan, satah orientasi (002)-ZnO muncul pada 34.28º dalam n-$TiO_2$/ZnO dwilapisan saput nipis apabila disepuhlindapan selama 2 jam pada 500 ℃. Kepancaran tinggi n-$TiO_2$/ZnO dwilapisan saput nipis dicapai sehingga 80% pada pinggir spektrum cahaya dalam julat 450-300 nm. Struktur hablur tinggi dan spektrum penyerapan p-$Cu_2O$ sebagai lapisan penyerap diperhatikan pada suhu 40 ℃ selama 1.5 jam. Simpang p-n berjaya dibentuk melalui sifat elektrik yang bererti dengan kecekapan penukaran sebanyak 0.0615%. Hasil menunjukkan homogen, pancaran yang tinggi dan kehabluran n-$TiO_2$/ZnO/p-$Cu_2O$ hetero-simpang saput nipis selain dapat mempertingkatkan struktur permukaan dan susunan atom serta bermanfaat untuk aplikasi sel suria.

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# LIST OF SYMBOLS AND ABBREVIATIONS

| | | |
|---|---|---|
| 3D | - | Three dimensional |
| $l_{Cu2O}$ | - | Lattice parameter of $Cu_2O$ |
| $l_{ZnO}$ | - | Lattice parameter of ZnO |
| A | - | Surface area |
| AFM | - | Atomic Force Microscopy |
| Ag/AgCl | - | Silver/silver chloride |
| AZO | - | Zinc/Aluminium doped |
| bcc | - | Body-centered cubic |
| BiVO | - | Bismuth vanadate |
| Cds | - | Cadmium sulphide |
| CdTe | - | Cadmium telluride |
| CE | - | Counter electrode |
| CIGS | - | Copper Indium Gallium Selenide |
| cm | - | centimetre |
| $CO_2$ | - | Carbon dioxide |
| Cu | - | Copper |
| $Cu_2O$ | - | Copper oxide |
| CV | - | Cyclic voltammetry |
| CZTS | - | Copper zinc tin sulphide |
| $D_2$ | - | Deuterium |
| DC | - | Direct current |
| DEA | - | Diethanolamine |
| E | - | Irradiance |
| $E_g$ | - | Band gap |
| eV | - | Electron volt |
| fcc | - | Face-centered cubic |

| | | |
|---|---|---|
| $Fe_2O_3$ | - | Hematite |
| FE-SEM | - | Field Emission Scanning Electron Microscopy |
| FF | - | Fill factor |
| FTO | - | Fluorine doped tin oxide |
| FWHM | - | Half-width at full maximum |
| h | - | Planck constant |
| $H_2O$ | - | Hydrogen |
| HCP | - | Close-pack hexagonal |
| IEA | - | International Energy Agency |
| $I_{max}$ | - | Maximum current |
| $I_{sc}$ | - | Short circuit current |
| ITO | - | Indium tin oxide |
| I-V | - | Current-voltage |
| LSV | - | Linear Sweep Voltammetry |
| M | - | mol |
| mm | - | millimetre |
| $MoO_3$ | - | molybdenum oxide |
| mV/s | | Millivolt/seconds |
| nm | - | nanometre |
| pH | - | Potential of hydrogen |
| PLD | - | Pulsed laser deposition |
| $P_{max}$ | - | Maximum power point |
| Pt | - | Platinum |
| PV | - | Photovoltaic |
| RE | - | Reference electrode |
| RF | - | Radio Frequency |
| rpm | - | Revolutions per minutes |
| SiO | - | Silicon monoxide |
| $SnO_2$ | - | Tin oxide |
| sso | - | Sample surface offset |
| TCO | - | Transparent Conducting Oxide |
| $TiO_2$ | - | Titanium dioxide |
| UV | - | Ultra-violet |

| | | |
|---|---|---|
| UV-Vis | - | Ultraviolet-Visible Absorption Spectroscopy |
| Vmax | - | Maximum voltage |
| $V_{oc}$ | - | Open circuit voltage |
| vs | - | versus |
| WD | - | Working distance |
| WE | - | Working electrode |
| $WO_3$ | - | Tungsten trioxide |
| XRD | - | X-Ray Diffraction |
| ZnO | - | Zinc oxide |
| $ZnTiO_3$ | | Zinc Titanate |
| α | - | Absorbance spectra |
| α-Si | - | Amorphous silicon |
| η | - | Conversion efficiency |
| μm | - | micrometre |

# LIST OF APPENDICES

# CHAPTER 1

# INTRODUCTION

This chapter deliberated on the introduction of the project. It consists of several subtopics including project overview, problem statement, research objectives, scope and limitations as well as the research contribution of the project.

## 1.1 Project overview

According to International Energy Agency (IEA), renewable energy technologies seem to have started growing in 2017 and have been paid attention by various industries. Several modern renewable energy technologies have been established as mainstream and cost-competitive sources of energy such as hydropower, bioenergy, geothermal power, fossil fuels and solar. However, fossil fuels are still at the highest percentage of consumption at 79.5% of the total final energy consumption [1]. This had been a challenge to researchers and industries since fossil fuels are some of the major sources that can cause pollution by carbon dioxide ($CO_2$) emission. $CO_2$ is one of the gases that can cause climate change and rise global temperature. Based on the growing rate of fossil fuel consumption, the concentration of $CO_2$ is foreseen to reach a dangerous level at 750 ppm by 2050. Since there is no natural decomposition of $CO_2$ in the atmosphere, the pollution effect can take 500 to 2000 years to reduce [2]. Thus, modern renewable technologies have received attention including solar photovoltaics (PV) which undergo natural processes by using sun as the main source which is much more environmentally friendly [2].

Based on a roadmap technologies statistics conducted by IEA, crystalline silicon technologies represent 85% to 90% of PV modules performance at world

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# APPENDIX A

## LIST OF PUBLICATIONS

| Year | Articles | Scopus/ISI |
|---|---|---|
| 2022 | **Arifin, N.M.,** Mohamad, F., Hussin, R., Ismail, A.Z.M., Ramli, S.A., Ahmad, N., Nor, N.H.M., Sahdan, M.Z., Zain, M.Z.M., Izaki, M., Annealing Treatment on Homogenous n-$TiO_2$/ZnO Bilayer Thin Film Deposition as Window Layer for p-$Cu_2O$ Based Heterostructure Thin Film. *Coatings. (Accepted on 07 November 2022)* | Q2 (IF 3.236) |
| 2021 | **Arifin, N.M.,** Mohamad, F., Hussin, R., Ismail, A.Z.M., Ramli, S.A., Ahmad, N., Nor, N.H.M., Sahdan, M.Z., Zain, M.Z.M., Izaki, M., Development of homogenous n-$TiO_2$/ZnO bilayer/p-$Cu_2O$ heterostructure thin film. *J Sol-Gel Sci Technol* 100, 224–231 (2021). *https://doi.org/10.1007/s10971-021-05650-7* | Q1 (IF 2.422) |
| 2020 | **N. Mohamad**, F. Mohamad, L. Sheng, A. Ismail, N. Ahmad, N. Nor, M. Izaki, "Construction of Nanorod-$TiO_2$/p-$Cu_2O$ Heterostructure Thin Films for Solar Cell Application," *Int. J. Adv. Trends Comput. Sci. Eng.*, vol. 9, no. 1, pp. 304–310, 2020. *https://doi.org/10.30534/ijatcse/2020/5391.12020* | Scopus |

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| 2019 | F. Mohamad, **N. M. Arifin**, A. Z. M. Ismail, N. Ahmad, M. N. N. Hisyamudin, and M. Izaki, "$Cu_2O$-based homostructure fabricated by electrodeposition method," *Acta Phys. Pol. A*, vol. 135, no. 5, pp. 911–914, 2019. *https://doi: 10.12693/APhysPolA.135.91* | Q4 (IF 0.536) |