STRUCTURAL AND ELECTRICAL CONDUCTIVITY MEASUREMENT OF ZINC OXIDE THIN FILM PREPARED BY SOL-GEL DIP COATING METHOD

PEH LY TAT

A thesis submitted in fulfilment of the
requirements for the award of the degree of
Master of Science (Physics)

Faculty of Science
Universiti Teknologi Malaysia

NOVEMBER 2015

To my beloved family members and
especially Mrs. Sheley Wang who always offered me the mighty support and love
throughout this research study.

ACKNOWLEDGEMENTS

I would like to send my remarkable gratefulness to my supervisor, Assoc. Prof. Dr. Karim Deraman and co-supervisor, Dr. Wan Nurulhuda Wan Shamsuri for the faithfulness guidance, support and the patience that had offered to me during the entire research project. I feel thankful to them as always providing me a highly encouragement environment during the research work until the thesis have been finally written up.

I wish to deliver my appreciation to my father, Mr. Peh Lay Huat; mother, Mdm. Lee Sooi Cheng; family members, and colleagues for their support either in mentally or physically. Furthermore, I would like to express my indebted to my wife, Mrs Sheley Wang for the understanding and patience that given to me throughout this research project. The sincere thanks are expressed to both of my lab mates, Mr Ong Wai Kit and Mr. Jackie Chen for the knowledge sharing among us during the conflict time while carrying the research work.

Sincere gratefulness is expressed to all the assistant science officers and lab assistants especially Mr. Mohd Nazari Kamarudin and Mr Keliwon for the technical and experience sharing to help me complete this research project in more fluent way. Moreover, I wish to thanks to Mr. Zailan and Mr Nazrul for performing the XRD and AFM measurement respectively in Universiti Kebangsaan Malaysia and Mdm Faezahana for the kindly assistance in the usage of surface profilometer in Universiti Tun Hussien Onn Malaysia.

Last but not least, thanks to Minister of Higher Education and Universiti Teknologi Malaysia for the financial support through the Research University Grant (RUG) Scheme.

ABSTRACT

Zinc oxide (ZnO) exhibits a hexagonal wurtzitic structure with a wide direct energy band gap of 3.37 eV. Furthermore, the ZnO thin films have low resistivity and high transparency in visible region and thus can be used as the fundamental layer in the fabrication of optoelectronic devices and as the transparent electrodes in solar cell panel. Undoped ZnO thin films were deposited onto corning glass substrates via the sol-gel dip coating method with annealing temperature and solution concentration being varied. The films were prepared under the annealing temperatures and solution concentrations ranging from 350 to 550 ^{o}C and 0.10 M to 0.30 M respectively. The effects of lowering the annealing temperature and solution concentration onto the characteristics of ZnO films were studied. The prepared sol-gel ZnO films can be used as a part of fundamental platform for the synthesis of novel ZnO nanostructures such as nanorods (1D) and nanowires (2D). The starting material in the form of zinc acetate dehydrates was dissolved into a mixing solution consisting of isopropanol and diethanolamine which acts as the solvent and stabilizer respectively. The thickness of the films has been measured by a surface profilometer. The structural properties of sol-gel ZnO films were characterized by X-Ray diffractometer (XRD) and atomic force microscope (AFM). Meanwhile, the film conductivity level was determined by Van der Pauw resistivity measurement method. The XRD spectra revealed that the prepared sol-gel ZnO films were polycrystalline with the hexagonal wurtzite structure and had the preferential growth along (002) c-axis orientation. The ZnO films prepared at 0.30 M concentration and annealed at 450 ℃ showed the highest crystallinity among all the samples. The grain size and the root mean square roughness of the ZnO films increased with the increase of annealing temperature and solution concentration. Furthermore, the biggest grain size of 66.60 nm was observed for ZnO films prepared with 0.30 M concentration and annealed at 550 ℃. The thickness of ZnO films annealed at 450 ℃ increases from 63 to 260 nm as the solution concentration increases from 0.10 to 0.30 M. The ZnO films prepared with 0.30 M concentration and annealed at 350 ℃ showed the lowest conductivity value of 2.49×10^{-4} S/cm with the activation energy E_a = 27 meV. Meanwhile, the highest film conductivity level was observed for the ZnO film prepared at 0.30 M concentration and annealed at 450 ℃ with value of 1.29×10^{-2} S/cm with E_a = 95 meV.

ABSTRAK

Zink oksida (ZnO) menyerupai struktur suatu wurtize heksagon berserta dengan sela jalur tenaga terus 3.37 eV. Selanjutnya, saput tipis ZnO mempunyai kerintangan eletrik yang rendah dan ketelusan lutsinar yang tinggi yang membolehkannya digunakan sebagai lapisan asas peranti optoelektronik dan elektrod lutsinar pada panel sel suria. Saput tipis ZnO yang belum didopkan dimendap pada substrat korning kaca menggunakan kaedah penyaduran secara pencelupan sol-gel. Saput tipis ZnO disepuh lindap pada suhu penyepuhlindapan dan kepekatan larutan berjulat masing-masing daripada 350 ℃ hingga 550 ℃ dan 0.10 M hingga 0.30 M. Kesan suhu penyepuhlindapan dan kepekatan larutan yang rendah terhadap sifat saput tips ZnO telah dikaji. Saput tipis ZnO yang telah dimendap melalui kaedah pencelupan sol-gel sesuai digunakan sebagai lapisan asas bagi sintesis struktur nano ZnO novel seperti nanorod (1D) dan nanowayar (2D). Serbuk kering zink acetate melarut dalam larutan yang mengandungi pelarut isopropanol dan diethanolamine. Ketebalan saput tipis ZnO diukur dengan menggunakan profilometer permukaan. Manakala, sifat struktur dikaji dengan menggunakan teknik analisis seperti difraktometer sinaran-X (XRD) dan mikroskop daya atom (AFM). Sifat kekonduksian elektrik diukur dengan menggunakan kaedah Van der Pauw. Spekrum XRD menunjukkan saput tipis ZnO mempunyai polihabluran yang berstruktur heksagonal berserta dengan (002) paksi-c sebagai arah pertumbuhan utama struktur hablur. Saput tipis ZnO yang disediakan pada kepekatan 0.30 M dan disepuh lindap pada suhu 450 ℃ menunjukan penghabluran terbaik di kalangan sampel. Saiz hablur dan morfologi saput tipis ZnO didapati meningkat dengan kenaikan suhu penyepuhlindapan dan kepekatan larutan. Saiz hablur yang terbesar ialah 66.60 nm diperolehi pada saput tipis ZnO yang disediakan dengan kepekatan 0.30 M dan suhu penyepuhlindapan 550 ℃. Ketebalan saput tipis ZnO yang disepuh lindap pada suhu 450 ℃ didapati meningkat daripada 63 hingga 260 nm apabila kepekatan larutan meningkat daripada 0.10 hingga 0.30 M. Saput tipis ZnO yang disediakan pada kepekatan 0.30 M dan disepuh lindap pada suhu 350 ° C menunjukan tahap kekonduksian yang terendah iaitu 2.49×10^{-4} S/cm dengan tenaga pengaktifan E_a= 27 meV. Manakala, saput tipis ZnO yang disediakan pada kepekatan 0.30 M dan disepuh lindap pada suhu 450 °C didapati mempunyai tahap kekonduksian elektrik yang tertinggi iaitu 1.29×10^{-2} S/cm dengan E_a = 95 meV.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF SYMBOLS AND ABREVIATIONS

ZnO	-	Zinc Oxide
ZnSe	-	Zinc Selenide
GaN	-	Gallium Nitrate
GaAs	-	Gallium Arsenide
InP	-	Indium Phosphide
CuO	-	Copper (II) Oxide
Zn	-	Zinc
Si	-	Silicon
Al	-	Aluminium
Ge	-	Germanium
P	-	Phosphorus
B	-	Boron
LED	-	Light Emitting Diode
XRD	-	X-ray Diffraction
AFM	-	Atomic Force Microscopy
EDX	-	Energy Dispersive Analysis of X-Ray
E_g	-	Energy Band Gap
E_a	-	Activation Energy
n_0	-	Concentration of Electron
p_0	-	Concentration of Holes
n_i	-	Equilibrium Concentration of electrons or holes in an intrinsic semiconductor
a, c	-	Lattice Constant
d_{hkl}	-	Lattice Interplanar Distance
Zn_i	-	Zinc Interstitials
O_i	-	Oxygen Interstitials
V_{zn}	-	Zinc Vacancies

DC	-	Direct Current
PLD	-	Pulse Laser Deposition
DEA	-	Diethanolamine
λ	-	Wavelength of Monochromatic X-ray Beam
θ	-	Diffraction Bragg Angle
Å	-	Angstrom = 10^{-10} m
D	-	Estimated Crystallite Size Derived From XRD
β	-	FWHM of 2θ
FWHM	-	Full Width at Half Maximum
2D	-	2 Dimensional
3D	-	3 Dimensional
ρ	-	Electrical Resistivity
σ	-	Electrical Conductivity
d	-	Thickness
$f(R)$	-	Correction Factors
K_B	-	Boltzmann Constant = $8.6173\text{x}10^{-5}$ eV K^{-1}
T_A	-	Annealing Temperature

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Background of Research

Thin film is defined as a coated layer of materials consisted of metal, semiconductor, and insulator with the thickness ranging from nanometer up to several micrometers in scale. In 1857, Michael Faraday invented the first metallic thin film via electrolysis process. Hence, the studied of the thin films has been attracted much more attention due to it's potentially for application in nano scale devices. The rapid growth in thin film technology leads to the new discoveries in science and technology and as the result, it was widely applied in engineering process system in most recent years. Moreover, thin films have the low power consumption, small occupying spaces and high speed performance.

Among the materials, zinc oxide compound semiconductor contributing a significant development in thin films technology. The hexagonal wurtzitic zinc oxide (ZnO) has a wide direct band gap of 3.37 eV and exciton binding energy of 60 meV at room temperature which three times larger than that of GaN (20 meV) [1-4]. ZnO is an intentional n-type semiconductor which resulted from active native stoichiometric points defects such as vacancies, interstitials and antisites [4]. Furthermore, ZnO have the excellent properties of non-toxicity, hydrogen and plasma stability and high transparency in visible region [5]. Hence, ZnO thin films becoming the promising material in fabricating various electronic devices such as gas sensor [6], optoelectronics [7], transparent electrode in solar cell panel [8] and light emitting diode (LED) [9].

1.2 Problem Statement

Previously, there are a numbers of researchers reported on the sol-gel ZnO thin films prepared under the dip coating and spin coating deposition technique. As the example, Yi Chen *et al.* [10] prepared the ZnO thin films via the sol-gel spin coating technique onto the silicon substrate and studied the effect of annealing temperature on the ZnO films characteristics with the temperature range from 550 to 950 ℃. Meanwhile, Sivakumar K. *et al.* [11] and Kyu Seog Hwang *et al.* [12] also reported the influence of annealing temperature prior to the nanocrystallinity of ZnO thin films fabricated via the sol-gel dip coating and spin coating method onto the glass substrate respectively. In Sivakumar K. *et al.* [11] work, they varied the annealing temperatures only as 300, 400, and 500 ℃ and Kyu Seog Hwang *et al.* [12] varied the annealing temperatures from 100 to 500 ℃ with every 100 ℃ increment step. Moreover, Jianguo *et al.* [13] studied the effect of precursor solution concentration which varied from 0.02 to 0.08 M on the structural and photoluminescence properties of sol-gel spin coating derived ZnO thin films. In our research work, the ZnO thin films were also prepared by the low cost sol-gel dip coating method with the varied parameters of annealing temperature and the level of precursor solution concentration. The sol-gel ZnO thin films were only prepared under the conditions of low annealing temperature together with low Zn^{2+} precursor solution concentration. Therefore, the influence of these deposition parameters and the relationship within structural and electrical conductivity measurement of sol-gel ZnO films have been studied in detail.

1.3 Research Objectives

The objectives of this research are:

i) To synthesize the ZnO thin films via the sol-gel dip coating method.

ii) To determine the effect of annealing temperature and precursor solution concentration onto the structural properties and electrical conductivity of ZnO films.

iii) To develop the Van der Pauw electrical resistivity measurement system.

iv) To determine the relationship within structural and electrical conductivity of sol-gel ZnO films.

1.4 Scope of Research

The scopes of this research included the preparation of ZnO thin films via the sol-gel dip coating deposition method with varied the annealing temperature and the level of precursor solution concentration. In the of sol-gel dip coated ZnO films process, the withdrawal speed and preheating temperature were fixed at 10 mm/min and 200 ℃ respectively. Meanwhile, the coating cycle from withdrawal to preheating step was fixed at nine cycles in the entire ZnO films preparation work. The annealing temperatures varied from 350 to 550 ℃ with the increment step of 50 ℃ which considered as low annealing temperature range. The Zn^{2+} solution concentration varied from 0.10 to 0.30 M with 0.05 M increment step. The structural properties of the sol-gel ZnO films were studied by using X-ray Diffractometer (XRD), atomic force microscope (AFM) and the Energy Dispersive Analysis of X-Ray (EDX). The thickness of the prepared sol-gel ZnO films was determined by the surface profilometer. Meanwhile, the Van der Pauw resistivity measurement method was used to determine the electrical conductivity of sol-gel ZnO films. Thus, the possible relationship of the structural and electrical conductivity of sol-gel prepared ZnO films been reported.

1.5 Significant of the Research

In this research work, the undoped ZnO films prepared by sol-gel dip coating deposition method due to its simplicity in controlling the films compositional, ability to fabricate a large scale area of thin film, low costing consumption and high films homogeneity. The study with respect to the effect of annealing temperature and precursor solution concentration on the structural properties and electrical conductivity level of ZnO films has been conducted. This research work outcomes can be a part of fundamental platform for the synthesis of novel ZnO nanostructured materials such as nanorods (1D), nanowires (1D) and nanoflower (3D) in future.

REFERENCES

[1] Tarwal N.L., Shinde V.V., Kamble A.L., Jadhav P.R.,Patil D.S., Patil V.B., and Patil P.S. Photoluminescence and photoelectrochemical properties of nanocrystalline ZnO thin films synthesized by spray pyrolysis technique. *Applied Surface Science*. 2011. 257:10789-10794.

[2] Shuo-Yi Kuo, Wei-Chun Chen, and Chin-Pao Cheng. Investigation of annealing-treatment on the optical and electrical properties of sol-gel derived zinc oxide thin films. *Supperlattices and Microstructures*. 2006. 39: 162-170.

[3] Seung Hwangbo, Yun-Ji Lee, and Kyu-Seog Hwang. Photoluminescence of ZnO layer on commercial glass substrate prepared by sol-gel process. *Ceramics International*. 2008. 34: 1237-1239.

[4] Patcharee Jongnavakit, Pongsaton Amornpitoksuk, Sumetha Suwanboon and Tanakorn Ratana. Surface and photocatalytic properties of ZnO thin film prepared by sol-gel method. *Thin Solid Films*. 2012. 520: 5561-5567.

[5] Kao M.C., Chen H.Z., and Young S.L. Effects of preannealing temperature of ZnO thin films on the performance of dye-sensitized solar cells. *Applied Physics A*. 2010. 98: 95-599.

[6] Fawzy A.Mahmoud and Kiriakidis G. Nanocrystalline ZnO thin film for gas sensor application. *Journal of Ovonic Research*. 2009. 5: 15-20.

[7] Chen K.J., Hung F.Y., Chang S.J., and Young S.J. Optoelectronic characteristics of UV photodetector based on ZnO nanowires thin films. *Journal of Alloys and Compounds*. 2009. 479: 674-677.

[8] Rajesh Kumar, Neeraj Khare, Vijay Kumar, and Bhalla G.L. Effect of intrinsic stress on the optical properties of nanostructured ZnO thin films grown by rf magnetron sputtering. *Applied Surface Science*. 2008. 254: 6509-6513.

[9] Hosseinmardi A., Shojaee N., Keyanpour-Rad M., and Ebadzadeh T. A study on the photoluminescence properties of electrospray deposited amorphous and crystalline nanostructured ZnO thin films. *Ceramics International*. 2012. 38: 1957-1980.

[10] Yi Chen, Jyoti Nayak, Hyun-U Ko, and Jaehwan Kim. Effect of annealing temperature on the characteristics of ZnO thin films. *Journal of Physics and Chemistry of Solids*. 2012. 73: 1259-1263.

[11] K. Sivakumar, V. Senthil Kumar, M. Thambidurai, and T.S. Senthil. Influence of annealing temperature on nanocrystalline ZnO thin films prepared by sol-gel dip coating method. *Advanced Materials Research*. 2013. 678: 91-102.

[12] Kyu-Seog Hwang, Yun-Ji Lee, and Seung Hwangbo. Growth, structure and optical properties of amorphous or nano-crystalline ZnO thin films prepared by prefiring-final annealing. *Journal of Ceramic Processing Research*. 2007. 8(5): 305-311.

[13] Jianguo Lv, Changlong Liu, Wanbing Gong, Zhenfa Zi, Xiaoshuang Chen, Kai Huang, Tao Wang, Gang He, Xueping Song, and Zhaoqi Sun. Effect of solution concentrations on crystal structure, surface topographies and photoluminescence properties of ZnO thin films. *Superlattices and Microstructures*. 2012. 51: 886-892.

[14] Kyu-Seog Hwang, Bo-An Kang, Ju-Hyun Jeong, Young-Sun Jeon, Byung-Hoon Kim. Spin coating-pyrolysis derived highly *c*-axis-oriented ZnO layers pre-fired at various temperatures. *Current Applied Physics*. 2007. 7: 421-425.

[15] Betty Lise Anderson and Richard Lise Anderson. *Fundamental of Semiconductor Devices*. New York: McGraw-Hill. 2005.

[16] Rolf Enderlein and Norman J.M. Horing. *Fundamentals of Semiconductor Physics and Devices*. Singapore: World Scientific Publishing Co. Pte. Ltd. 1997.

[17] Peter Y. Yu and Manuel Cardona. *Fundamentals of Semiconductors: Physics and Materials Properties*. 3rd. ed. New York: Springer Berlin Heidelberg. 2006.

[18] John Orton. *The Story of Semiconductor*. United States: Oxford University Press. 2004.

[19] Ghosh R.,Paul G.K. and Basak D. Effect of thermal annealing treatment on structural, electrical and optical properties of transparent sol-gel ZnO thin films. *Materials Research Bulletin*. 2005. 40:1905-1914.

[20] Linhua Xu, Caige Zheng, Juhong Miao, and Fenglin Xian. Dependence of structural and optical properties of sol-gel derived ZnO thin films on sol concentration. *Applied Surface Science*. 2012. 258:7760-7765.

[21] Tan Hang Khume. *Synthesis and Characterization of Aluminium Doped Zinc Oxide Thin Films For Solar Cell*. MSc. Thesis. Universiti Teknologi Malaysia; 2007.

[22] Collins T.C. and Hauenstein R.J. Fundamental Properties of ZnO. In: Cole W. Litton. *Zinc Oxide Materials for Electronic Device Applications*. United Kingdom: John Wiley & Sons Ltd. 1-28; 2011.

[23] Davood Raoufi and Taha Raoufi. The effect of heat treatment on the physical properties of sol-gel derived ZnO thin films. *Applied Surface Science*. 2009. 255: 5812-5817.

[24] Jimenez Gonzalez A.E. and Soto Urueta J.A. Optical transmittance and photoconductivity studies on ZnO:Al thin films prepared by the sol-gel technique. *Solar Energy Materials and Solar Cells*. 1998. 52: 345-353.

[25] Musat V., Rego A.M., Monteiro R., and Fortunato E. Microstructure and gas-sensing properties of sol-gel ZnO thin films. *Thin Solid Films*. 2008. 516: 1512-1515.

[26] Jianguo Lv, Wanbing Gong, Kai Huang, Jianbo Zhu, Fanming Meng, Xueping Song, and Zhaoqi Sun. Effect of annealing temperature on photocatalytic activity of ZnO thin films prepared by sol-gel method. *Superlattices and Microstructures*. 2011. 50: 98-106.

[27] Jianguo Lv, Fengjiao Shang, Guangcai Pan, Feng Wang, Zhitao Zhuo, Changlong Liu, Wanbing Gong, Zhenfa Xi, Xiaoshuang Chen, Gang He, Miao Zhang, Xueping Song, Zhaoqi Sun, and Feng Liu. Effect of solution concentration on surface morphology and photocatalytic activity of ZnO thin films synthesized by hydrothermal methods. *Jounal of Materials Science: Mater Electron*. 2014. 25: 882-887.

[28] Ait Ahmed N., Fortas G., Hammache H., Sam S., Keffous A., Manseri A., Guerbous L., and Gabouze N. Structural and morphological study of ZnO thin films electrodeposited on n-type silicon. *Applied Surface Science*. 2010. 256: 7442-7445.

[29] Seval Aksoy, Yasemin Caglar, Saliha Ilican, and Mudjat Caglar. Sol-gel derived zinc oxide films: effect of deposition parameters on structure, microstructure and photoluminescence properties. *Superlattices and Microstructures*. 2011. 50: 470-479.

[30] Zaier A., Oum El az, Lakfif, A. Kabir, Boudjadar S., and Aida M.S. Effects of substrate temperature and solution molarity on the structural opto-electric properties of ZnO thin films deposited by spray pyrolysis. *Materials Science in Semiconductor Processing*. 2009. 12: 207-211.

[31] Mudjat Caglar, Saliha Ilican, Yasemin Caglar, and Fahrettin Yakuphanoglu. Electrical conductivity and optical properties of ZnO nanostructured thin film. *Applied Surface Science*. 2009. 255: 4491-4496.

[32] Mohd Firdaus Malek, Mohamad hafiz Mamat, Mohd Zainizan Sahdan, Musa Mohamed Zahidi, Zuraida Khusaimi, and Mohamad Rusop Mahmood. Influence of various sol concentration on stress/strain and properties of ZnO thin films synthesized by sol-gel technique. *Thin Solid Films*. 2013. 527: 102-109.

[33] Wei X.Q, Zhang Z.G., Liu M., Chen C.S., Sun G., Xue C.S., Zhuang H.Z., and Man B.Y.. Annealing effect on the microstructure and photoluminescence of ZnO thin films. *Materials Chemistry and Physics*. 2007. 101: 285-290.

[34] Asghari Maqsood, Islam M., Ikram M., Salam S., and Ameer S.. Synthesis, characterization and hall effect measurements of nano-crystalline ZnO thin films. *Key Engineering Materials*. 2012. 510-511: 186-193.

[35] Jin B.J., Woo H.S., Im S., Bae S.H., and Lee S.Y. Relationship between photoluminescence and electrical properties of ZnO thin films grown by pulsed laser deposition. *Applied Surface Science*. 2001. 169-170: 521-524.

[36] Xuemei Teng, Hongtao Fan, Shusheng Pan, Cong Ye, and Guanghai Li. Abnormal photoluminescence of ZnO thin film on ITO glass. *Materials Letters*. 2007. 61: 201-204.

[37] Zendehnam A., Mirzaee M., and Miri S. Effect of annealing temperature on PL spectrum and surface morphology of zinc oxide thin films. *Applied Surface Science*. 2013. 270: 163-168.

[38] Liu Y.C., Tung S.K., and Hsieh J.H.. Influence of annealing on optical properties and surface structure of ZnO thin films. *Journal of Crystal Growth*. 2006. 287: 105-111.

[39] Linhua Xu, Linxing Shi, and Xiangyin Li. Preparation of nanocone ZnO thin film and its aging effect of photoluminescence. *Applied Surface Science*. 2009. 255: 5957-5960.

[40] Michel A. Aegerter and Martin Menning. *Sol-Gel Technologies for Producers and Users*. United States: Kluwer Academic Publisher. 2004.

[41] Masoud Eslami, Zohreh Hamnabard, and Ali Nemati. Synthesis and spectral properties of Nd-doped glass-ceramics in SiO_2-CaO-MgO system prepared by sol-gel method. *Journal of Rare Earths*. 2013. 31(6): 595.

[42] Quan Chen and Andrew M. Soutar. Progress on nanoceramics by sol-gel process. *Key Engineering Materials*. 2009, 391: 79-95.

[43] Casanova J.R., Heredia E.A., Bojorge C.D., Canepa H.R., Kellermann G., and Craievich A.F. Structural characterization of supported nanocrystalline ZnO thin films prepared by dip-coating. *Applied Surface Science*. 2011. 257: 10045-10051.

[44] Alon McCormick. Recent Progress In The Study Of The Kinetics Of Sol-Gel SiO2 Synthesis Reactions. In: Yosry A. Attia. *Sol-Gel Processing and Applications*. New York: Plenum Press. 3; 1994.

[45] Mutia Suhaibah Binti Abdullah. *The Sol-Gel Synthesis and Characterization of Strontium Titanate Doped With Pr and Al Ions*. MSc Thesis. Universiti Teknologi Malaysia; 2010.

[46] Ivanova T., Harizanova A., Koutzarova T., and Vertruyen B. Study of ZnO sol-gel films: Effect of annealing. *Materials Letters*. 2010. 64: 1147-1149.

[47] Sivakumar K., Senthil Kumar V., Muthukumarasamy N., Thambidurai M., and Senthil T.S. Influence of pH on ZnO nanocrystalline thin films prepared by sol-gel dip coating method. *Bulletin Materials Science*. 2012. 35(3): 327-331.

[48] Joydip Sengupta, Sahoo R.K.,. Bardhan K.K, and Mukherjee C.D. Influence of annealing temperature on the structural, topographical, and optical properties of sol-gel derived ZnO thin films. *Materials Letters*. 2011. 65: 2572-2574.

[49] Kevin Alvin Eswar, Husairi F.S., Azlinda Ab Aziz, Mohamad Rusop, and Saifollah Abdullah. Effect of post annealing temperature on surface morphology and photoluminescence properties of ZnO thin film. *Advanced Materials Research*. 2014. 832: 654-658.

[50] Minrui Wang, Jing Wang, Wen Chen, Yan Cui, and Liding Wang. Effect of preheating and annealing temperatures on quality characteristics of ZnO thin film prepared by sol-gel method. *Materials Chemistry and Physics*. 2006. 97: 219-225.

[51] Bandyopadhyay S., Paul G.K., Roy R., Sen S.K. and Sen S.. Study of structural and electrical properties of grain-boundary modified ZnO films prepared by sol-gel technique. *Materials Chemistry and Physics*. 2002. 74: 83-91.

[52] Rene Guinebretiere. *X-Ray Diffraction by Polycrystalline Materials*. Great Britain and United States: ISTE Ltd. 2007.

[53] Cullity B.D. *Elements of X-Ray Diffraction*. 2nd. ed. Philippines: Addison-Wesley Publishing Company Inc. 1978.

[54] Van der Pauw L.J. A method of measuring the resistivity and hall coefficient on lamellae of arbitrary shape. *Philips Technical Review*. 1958. 20: 221.

[55] Yung-Shou Ho and Kuan-Yi Lee. Fabrication of highly oriented (002) ZnO film on glass by sol-gel method. *Thin Solid Films*. 2010. 519: 1431-1434.

[56] Lin Cui, Gui-GenWang, Hua-Yu Zhang, Rui Sun, Xu-Ping Kuang, and Jie-Cai Han. Effect of film thickness and annealing temperature on the structural and optical properties of ZnO thin films deposited on sapphire (0001) substrates by sol-gel. *Ceramics International*. 2013. 39: 3261-3268.

[57] Trilochan Sahoo, Myoung Kim, Mi-Hee Lee, Lee-Woon Jang, Ju-Won Jeon, Joon Seop Kwak, In-Yong Ko, and In-Hwang Lee. Nanocrystalline ZnO thin films by spin coating-pyrolysis method. *Journal of Alloys and Compounds*. 2010. 491: 308-313.

[58] Dutta M., Mridha S., and Basek D.. Effect of sol concentration on the properties of ZnO thin films prepared by sol-gel technique. *Applied Surface Science*. 2008. 254: 2743-2747.

[59] Shane O'Brien, Koh L.H.K., and Gabriel M. Crean. ZnO thin films prepared by a single step sol-gel process. *Thin Solid Films*. 2008. 516: 1391-1395.

[60] Peh Ly Tat, Karim Deraman, Rosli Hussin, Wan Nurulhuda Wan Shamsuri, and Zuhairi Ibrahim. The structural and surface morphology of annealed ZnO films. *Advanced Materials Research*. 2014. 903: 73-77.

[61] Peh Ly Tat, Karim Deraman, Wan Nurulhuda Wan Shamsuri, Rosli Hussin, and Zuhairi Ibrahim. Microstructural and electrical conductivity of annealed ZnO thin films. *Advanced Materials Research*. 2014. 970: 120-123.

[62] Vinod Kumar, Neetu Singh, R.M. Mehra, Avinashi Kapoor, L.P. Purohit, and H.C. Swart. Role of film thickness on the properties of ZnO thin films grown by sol-gel method. *Thin Solid Films*. 2013. 539: 161-165.

[63] Mazabalo Baneto, Alexandru Enesca, Yendoube Lare, Koffi Jondo, Kossi Napo, and Anca Duta. Effect of precursor concentration on structural, morphological and opto-electric properties of ZnO thin films prepared by spray pyrolysis. *Ceramics International*. 2014. 40: 8397-8404.

[64] Young-Sung Kim, Weon-Pil Tai, and Su-Jeong Shu. Effect of preheating temperature on structural and optical properties of ZnO thin films by sol-gel process. *Thin Solid Films*. 2005. 491: 153-160.

[65] Shafinaz Sobihana Shariffudin, Sukreen Hana Herman and Mohamad Rusop. Layer-by layer nanoparticles ZnO thin films prepared by sol-gel method. *Advanced Materials Research*. 2012. 403: 1178-1182.