

UNIVERSITI TEKNOLOGI MARA

**FABRICATION AND CHARACTERISATION
OF GRADED INDEX NANOSTRUCTURED
TiO₂ COMPACT LAYERS FOR
DYE-SENSITIZED SOLAR CELL**

MOHD HANAPIAH ABDULLAH

Thesis submitted in fulfillment
of the requirements for the degree of
Doctor of Philosophy

Faculty of Electrical Engineering

February 2015

AUTHOR'S DECLARATION

I declare that the work in this thesis was carried out in accordance with the regulations of Universiti Teknologi MARA. It is original and is the result of my work, unless otherwise indicated or acknowledged as referenced work. This thesis has not been submitted to any other academic institution or non-academic institution for any other degree or qualification.

I, hereby, acknowledge that I have been supplied with the Academic Rules and Regulations for Post Graduate, University Teknologi MARA, regulating the conduct of my study and research.

Name of Student : Mohd Hanapiah Bin Abdullah
Student's ID No. : 2009320133
Programme : PhD in Electrical Engineering
Thesis Title : Fabrication and Characterisation of Graded
Index Nanostructured TiO₂ Compact Layers for
Dye-Sensitised Solar Cell

Signature of Student

Date : February 2015

ABSTRACT

A novel graded index nanostructured TiO₂ compact layer (arc-TiO₂) had been successfully deposited on Indium tin oxide (ITO) substrate by long-throw radio-frequency (RF) magnetron sputtering. The main aims of the arc-TiO₂ compact layer were to serve as an antireflective compact layer that could reduce reflection losses, improve the photo-response of dye molecules, and prevent electron recombination in a dye-sensitized solar cell (DSSC) application. The employment of the TiO₂ compact layer in the DSSC was carefully optimised in term of RF power and thickness of the arc-TiO₂ film. Meanwhile, the desired characteristics were systematically investigated by means of UV-absorption spectra, incident photon to current efficiency (IPCE), open-circuit voltage decay (OCVD) and electrochemical impedance spectroscopy (EIS). The average transmittance of the ITO/arc-TiO₂ conducting substrate in the region from 400 nm to 1000 nm was approximately 85%. Corresponding average reflectance difference that was recorded in comparison to the bare ITO was 2.5 %. The red-shift behaviour of the transmittance peak was actually due to the formation of a new hybrid band energy structure of 3.1 eV resulting from the tin (Sn) diffusion in the ITO film that shifted the absorption edge of the substrate. This had favoured the absorption characteristics and photo-response of N719 dye. Hence, the consistency of peaks between the transmittance spectra of the substrate with the corresponding IPCE spectra of the DSSC cell was improved. Additionally, the arc-TiO₂ compact layer preserved the higher conductivity of the ITO films from oxygen-related defects during the annealing process. The resistivity of the ITO/arc-TiO₂ substrate was conserved at $2.05 \times 10^{-4} \Omega \text{ cm}$ even at this high temperature. The preserved conductivity had consequently decreased the charge interfacial resistance (R₁) in the EIS measurement and facilitated the charge transport from the nanocrystalline-TiO₂ to the ITO. In later investigations, it was revealed that the 100 nm thickness of arc-TiO₂ compact layer prepared using 100W RF power become the optimal deposition parameters for preparing the compact layer. At this stage, the reduced interfacial resistance R₁ observed under EIS measurement was 2.36 Ω and the highest IPCE peak of 58% was achieved at 550 nm wavelengths. The higher IPCE had contributed to 8.93 mA/cm² of the cell's short-circuit photocurrent, J_{SC} and 0.67 V to the associated open circuit voltage, V_{OC} . The photo-generated electron lifetime, τ_n of 1.1 orders of magnitude higher than bare ITO was achieved. As a result, the overall conversion efficiency of 3.45% was recorded for the optimized DSSC device. This record was actually 50% improved compared to that of bare ITO cell. Therefore, the combined effects (i.e., reduced reflection losses and interfacial resistance with better photo-response) owed to the arc-TiO₂ compact layer prepared at their optimal condition was found to be important as it had originated the remarkable improvement in this arc-TiO₂ based DSSC.

ACKNOWLEDGEMENT

Bismillahi wal hamdalillah.....

In the name of Allah, Most Gracious, Most Merciful, All praise is due to Allah, the Cherisher and Sustainer of the worlds.

First and foremost, I would like to express my sincere gratitude to my Supervisor, Professor Dr. Mohamad Rusop bin Mahmood for his valuable guidance and relentless support throughout the course of this study. Much of this work would have been virtually impossible without his charismatic supervision.

My heartfelt gratitude also goes to my family members: to my dearest wife, Pn Syarifah Adilah Mohamed Yusof, and to all my beloved kids, Mohd Faqih Hanif, Mohd Fahim, Nurul Fathiyyah and Nur Fasiha Amni, for their encouragement, patience and deep understanding. Words cannot sufficiently express how grateful having all of you as accompanies, and it becomes unforgettable moments in this journey of study.

My deepest loved goes to my adored father (Abdullah Idris) and lovely mother (Bidah Mat Kassim), for their continuous prayers and endless support is the essence of this successful story. My Allah blessed both of you here and the day after.

Many thanks to all who supported me in any respect during the completion of this study, especially to the group of fellow buddies at NANO-ElecTronic Centre (NET), Dr. Mohamad Hafiz Mamat, Miss Lyly Nyl Ismail, Mrs. Asiah Mohd Nor, Dr. Puteri Sarah Mohd Saad, Dr. Shafinaz Sobihana Shariffudin, Dr. Nor Diyana Md Sin, Dr. Nur Amalina, Mr. Musa Mohamed Zahidi, Mr. Mohd Azwan Roseley, Mr. Mohd Danial Mohd Johari, Mr. Mohd Suhaimi Ahmad, and many more. They have contributed many ideas in the research, and we have also shared a lot of wonderful moments during the study.

Mohd Hanapiah Bin Abdullah

Universiti Teknologi MARA

February 2015

TABLE OF CONTENTS

	Page
AUTHOR'S DECLARATION	i
ABSTRACT	ii
ACKNOWLEDGEMENTS	iii
TABLE OF CONTENTS	iv
LIST OF TABLES	viii
LIST OF FIGURES	ix
LIST OF ABBREVIATIONS	xiv
CHAPTER ONE: INTRODUCTION	
1.1 Renewable Energy	1
1.2 Dye-Sensitized Solar Cell- A Renewable Energy Candidate	2
1.3 Overview of Factors influencing DSSC Performance	2
1.4 Problem Statement	4
1.5 Research Objectives	5
1.6 Scope of Research	6
1.7 Research Contribution	6
1.8 Thesis Organisation	7
CHAPTER TWO: LITERATURE REVIEW	
2.1 Introduction	9
2.2 Dye-Sensitised Solar Cell (DSSC)	9
2.3 Semiconductor Photoelectrode	11
2.3.1 Semiconductor Oxide	12
2.3.2 Transparent Conducting Oxide (TCO)	14
2.3.3 Ruthenium (Ru) Complex Photosensitizer	15
2.3.4 Redox Electrolytes	18
2.3.5 Counter Electrode	19
2.3.6 Sealing	19