



UNIVERSITI PUTRA MALAYSIA

***STUDY OF POLY(3,4-ETHYLENEDIOXYTHIOPHENE) BASED COUNTER
ELECTRODES FOR EFFICIENT DYE-SENSITIZED SOLAR CELL***

MUHAMMAD NORHAFFIS MUSTAFA

FS 2018 20



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ELECTRODES FOR EFFICIENT DYE-SENSITIZED SOLAR CELL**

By

MUHAMMAD NORHAFFIS BIN MUSTAFA

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

November 2017

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

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MUHAMMAD NORHAFFIS BIN MUSTAFA

November 2017

Chairman: Associate Professor Yusran Sulaiman, PhD
Faculty: Science

In this work the enhancement of dye-sensitized solar cell (DSSC) performance was studied by developments of new counter electrode (CE) using a combination of three different materials namely poly(3,4-ethylenedioxythiophene) (PEDOT), carbon-based material (CBM; graphene oxide (GO), reduced graphene oxide (rGO), nanocrystalline cellulose (NCC) and multi-walled carbon nanotube (MWCNT)) and titanium dioxide (TiO₂). The counter electrode was prepared by coating indium tin oxide (ITO) glass with TiO₂ followed by deposition of PEDOT incorporated with different CBMs to produce novel CEs with high performance. Among the CEs PEDOT-NCC/TiO₂ exhibited the highest PCE of 2.10 % compared to PEDOT-MWCNT/TiO₂ (1.29 %), PEDOT-rGO/TiO₂ (1.10 %) and PEDOT-GO (1.17 %). PEDOT-NCC/TiO₂ also displayed a lower charge transfer resistance ($R_{ct} = 2.4 \Omega$) and higher cathodic peak current density ($I_{cp} = -2.60 \text{ mA.cm}^{-2}$) compared to other CEs due to the synergistic effect of high conductivity of PEDOT, high surface area and high optical transparency of NCC and porous structure of TiO₂ that provide large surface area. The impact of this study in photovoltaic technology is to produce an efficient and low cost CE that capable to substitute a typical CE in DSSC which is platinum that is expensive and rare metal. The future plan can be pursued by producing a flexible CE to increase its application in various fields.

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

**PENYELIDIKAN KEATAS POLI(3,4-ETILENADIOKSITIOFENA) SEBAGAI
ELEKTROD BERLAWANAN YANG BERKESAN UNTUK PEWARNA SEL
SOLAR BERKEPEKAAN**

Oleh

MUHAMMAD NORHAFFIS MUSTAFA

November 2017

Pengerusi: Profesor Madya Yusran Sulaiman, PhD
Fakulti: Sains

Dalam penyelidikan ini, peningkatan prestasi pewarna sel solar berkepekaan (DSSC) telah dikaji oleh elektrod berlawanan (CE) baru yang telah dicipta menggunakan gabungan tiga bahan yang berbeza iaitu poli (3,4-etilenadioksitiofena) (PEDOT), bahan berasaskan karbon (CBM; grafin oksida (GO), grafin oksida terturun (rGO), selulosa nanokristal (NCC) dan nanotub karbon berbilang dinding (MWCNT)) dan titanium dioksida (TiO_2). CE telah disediakan dengan menyaluti kaca indium timah oksida (ITO) dengan TiO_2 , diikuti oleh pemendapan PEDOT digabungkan dengan CBM yang berbeza untuk menghasilkan CE novel yang berprestasi tinggi. Di antara CEs, PEDOT-NCC/ TiO_2 memaparkan kecekapan penukaran kuasa (PCE) yang paling tinggi iaitu 2.10 % berbanding PEDOT-MWCNT/ TiO_2 (1.29 %), PEDOT-rGO/ TiO_2 (1.10 %) dan PEDOT-GO (1.17 %). PEDOT-NCC/ TiO_2 juga menunjukkan rintangan pemindahan caj yang lebih rendah ($R_{CT} = 2.4 \Omega$) dan ketumpatan arus puncak katod yang lebih tinggi ($I_{CP} = -2,60 \text{ mA.cm}^{-2}$) berbanding CE yang lain disebabkan oleh kesan sinergi konduksian PEDOT yang tinggi, luas permukaan dan transparansi optik NCC yang tinggi dan struktur berliang TiO_2 yang memberikan luas permukaan yang luas. Kesan kajian ini dalam teknologi photovoltaik adalah untuk menghasilkan CE yang cekap dan berkos rendah yang mampu menggantikan CE yang biasa digunakan didalam DSSC iaitu platinum yang berkos tinggi dan bahan yang jarang ditemui. Pelan pada masa hadapan boleh dilakukan dengan menghasilkan CE yang fleksibel untuk meningkatkan aplikasi DSSC dalam pelbagai bidang.

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-David Petraeus-



I certify that a Thesis Examination Committee has met on 16 November 2017 to conduct the final examination of Muhammad Norhaffis Bin Mustafa on his thesis entitled “Study Of Poly(3,4-Ethylenedioxythiophene Based Counter Electrodes For Efficient Dye-Sensitized Solar Cell” 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.

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LIST OF SYMBOLS

Symbol	Meaning	Usual unit
OCP	Open circuit voltage	V
I_{cp}	Cathodic peak current density	$\text{mA}\cdot\text{cm}^{-2}$
E_{pp}	Peak to peak separation	V
P_{in}	Power input	$\text{mW}\cdot\text{cm}^{-2}$
P_{max}	Maximum power	mW
J_{sc}	Short circuit current	mA
V_{oc}	Open circuit voltage	V
R_{ct}	Charge transfer resistance	Ω
R_s	Solution resistance	Ω
CPE	Constant phase element	F
Z'	Real impedance	Ω
Z''	Imaginary impedance	Ω
J_o	Exchange current density	$\text{mW}\cdot\text{cm}^{-2}$
J_{lim}	Limiting diffusion exchange current density	$\text{mW}\cdot\text{cm}^{-2}$

LIST OF ABBREVIATIONS

ATR	attenuated total reflection
CA	chronoamperometry
CE	counter electrode
CPs	conducting polymers
CPE	constant phase element
CV	cyclic voltammetry
EDOT	3, 4-ethylenedioxythiophene
EIS	electrochemical impedance spectroscopy
FTIR	fourier transforms infrared spectroscopy
ITO	indium tin oxide coated glass
kHz	kilohertz
LSV	linear sweep voltammetry
mA	milli ampere
mM	millimolar
OCP	open circuit potential
PEDOT	poly(3,4 ethylenedioxythiophene)
PEDOT-GO	poly(3,4 ethylenedioxythiophene)-graphene oxide
PEDOT-rGO	poly(3,4-ethylenedioxythiophene)-reduced graphene oxide
PEDOT-NCC	poly(3,4-ethylenedioxythiophene)-nanocrystalline cellulose
PEDOT-MWCNT	poly(3,4-ethylenedioxythiophene)-multiwalled carbon nanotubes
PEDOT-GO/TiO ₂	poly(3,4 ethylenedioxythiophene)-graphene oxide/titanium dioxide
PEDOT-rGO/TiO ₂	poly(3,4 ethylenedioxythiophene)-reduced graphene oxide/titanium dioxide
PEDOT-NCC/TiO ₂	poly(3,4 ethylenedioxythiophene)-nanocrystalline cellulose/titanium dioxide
PEDOT-MWCNT/TiO ₂	poly(3,4 ethylenedioxythiophene)-multiwalled carbon nanotubes/titanium dioxide counter electrode
FESEM	field emission scanning electron microscopy
RE	reference electrode
V	volts
WE	working electrode
PEDOT-GO CE	poly(3,4 ethylenedioxythiophene)-graphene oxide counter electrode
PEDOT CE	poly(3,4 ethylenedioxythiophene) counter electrode
GO CE	graphene oxide counter electrode
Pt CE	platinum counter electrode
DSSCs	dye-sensitized solar cell
R_{ct1}	charge transfer resistance at high frequency
R_s	series resistance
LiClO ₄	lithium perchlorate
R_{ct2}	charge transfer resistance at low frequency
TTIP	titanium isopropoxide
KCl	potassium chloride
N719	ruthenizer 535-bis TBA
Ag/AgCl	silver/silver chloride

XRD	x-ray diffraction
JCPDS	joint committee on powder diffraction standards
AC	alternating current
A.M.	air mass
I_{cp}	cathodic peak current density
I_3^-	tri-iodide ion
I^-	iodide ion
I-V curves	current-voltage curves
V_{oc}	open circuit voltage
J_{sc}	short circuit current
P_{max}	maximum power
FF	fill factor
η	overall power conversion efficiency
J_{max}	maximum current
J_o	exchange current density
J_{lim}	limiting diffusion exchange current density
ECA	electrocatalytic activity
PCE	power conversion efficiency

CHAPTER 1

INTRODUCTION

1.1 Background study

Nowadays, fossil fuel energy undergoes depletion due to the increasing demand for energy supply. Li *et al.* (2006) reported that the fossil fuels resource reserves throughout the world in 2002 can only last 40 years for oil, 200 years for coal and 60 years for natural gas. Thus, renewable energy is needed to overcome this problem. Solar energy is one of the renewable energies that grab the attention of researchers due to the supply of energy from the sun to the earth is gigantic about 3×10^{24} Joules a year that is 10,000 times more than energy consumption of population around the globe. Thus, global energy demand will be satisfied by covering 0.1% of the earth's surface with minimum power conversion efficiency (PCE) of 10% solar cell (Grätzel, 2001).

Dye-sensitized solar cell (DSSC) is a modern type of solar cell that involves four important components which are photoanodes, counter electrode (CE), electrolyte and dye. A typical DSSC uses titanium dioxide (TiO_2), platinum (Pt), tri-iodide/iodide and ruthenizer 535-bis TBA N719 as a source of photoanodes, CE, electrolyte and dye, respectively. O'Regan and Grätzel (1991) reported that 7.1-7.9 % PCE was achieved in simulated solar light for a low cost and high-efficiency DSSC based on colloidal TiO_2 as photoanode. Even though DSSCs have reached PCE as high as 10 % under AM 1.5 (100 mW/cm^2) (Nazeeruddin *et al.*, 1993), the applications of DSSC are still limited due to the expensive and rare material of Pt as a CE. Thus, this study will focus on producing a synergistic effect by fabricating novel CEs containing conducting polymer, carbon-based material and metal oxide to enhance the PCE of DSSC.

1.2 Problem statements

Pt as a CE in DSSC faced a lot of drawbacks such as rare, required high-temperature treatment, undergo corrosion with the electrolyte and high cost. Thus, a novel approach was made to produce new CEs that have promising properties such as high conductivity, high surface area and high availability that can improve the electrocatalytic activity and PCE of DSSC. This research will focus on producing novel PEDOT-GO/ TiO_2 CE for efficient DSSC performance. Besides, various carbon-based materials such as reduced graphene oxide (rGO), nanocrystalline cellulose (NCC) and multiwalled carbon nanotube (MWCNT) will be studied throughout this research to produce novel CEs with enhanced DSSC performance which are PEDOT-rGO/ TiO_2 , PEDOT-MWCNT/ TiO_2 and PEDOT-NCC/ TiO_2 . PEDOT possesses high conductivity, rGO and MWCNT contribute to the high surface area and high conductivity, NCC provides high surface area and high optical transparency while TiO_2 provides high

surface area. Thus, the incorporation of PEDOT and various carbon-based material with and without TiO_2 is believed to produce a synergistic effect that can enhance the PCE of DSSC.

1.3 Objectives of research

The objectives of this research are:

1. To fabricate PEDOT-GO/ TiO_2 as a counter electrode for DSSC through chronoamperometry technique.
2. To study the effect of PEDOT incorporated with various carbon-based materials (rGO, NCC and MWCNT) and TiO_2 as counter electrodes on the DSSC performance.
3. To characterize the counter electrode using FTIR, XRD, FESEM, CV, EIS, J-V and Tafel polarization analysis.

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LIST OF PUBLICATIONS

Mustafa, M., Shafie, S., Zainal, Z. and Sulaiman, Y. (2017a) *A Novel Poly(3,4-ethylenedioxythiophene)-graphene Oxide/Titanium Dioxide Composites Counter Electrode for Dye-Sensitized Solar Cell.*

Mustafa, M., Shafie, S., Zainal, Z. and Sulaiman, Y. (2017b) *Poly(3,4-ethylenedioxythiophene) doped with various carbon-based materials as counter electrodes for dye sensitized solar cells.*

