

**FORMATION AND CHARACTERIZATION OF
Pb_xCd_{1-x}S INTERLAYER FOR PbS/CdS/ZnS
QUANTUM DOT SENSITIZED SOLAR CELLS**

by

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DECLARATION

I would like to declare that the contents presented in this thesis entitled “Formation and Characterization of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ Interlayer for PbS/CdS/ZnS Quantum Dot Sensitized Solar Cells” are my own work which was done at Universiti Sains Malaysia unless stated otherwise. The results obtained did not contain materials that were submitted without acknowledgement to any universities or materials that were previously published, written or produced by another person except due reference was made in the text.

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LIST OF ABBREVIATIONS

CB	Conduction Band
CdS	Cadmium Sulphide
DSSCs	Dye Sensitized Solar Cells
EDX	Energy Dispersive X-Ray
FESEM	Field Emission Scanning Electron Microscopy
FF	Fill Factor
FTO	Fluorine Doped Tin Oxide
ICDD	International Centre Diffraction Data
J-V	Current density-Voltage
nm	Nanometer
PbS	Lead Sulphide
Pb _x Cd _{1-x} S	Lead Cadmium Sulphide
QDs	Quantum Dots
QDSSCs	Quantum Dot Sensitized Solar Cells
SILAR	Successive Ionic Layer Adsorption and Reaction
TEM	Transmission Electron Microscopy
TiO ₂	Titanium Oxide
UV	Ultraviolet

V	Voltage
Vis	Visible
VB	Valence Band
XRD	X-ray Diffraction
ZnS	Zinc Sulphide

LIST OF SYMBOLS

E_g	Band Gap
M	Molar Concentration
M_w	Molar Mass
N	Number of Moles
J_{mp}	Current Density at Maximum Point
J_{sc}	Short-Circuit Current
P	Incident Solar Power Density
V	Volume
V_{mp}	Voltage at Maximum Point
V_{oc}	Open Circuit Voltage
η	Efficiency
x	Molar Fraction

**PEMBETUKAN DAN PENCIRIAN ANTARA LAPISAN $Pb_xCd_{1-x}S$
BAGI $PbS/CdS/ZnS$ SEL SURIA SENSITIF TITIK KUANTUM**

ABSTRAK

Sel suria sensitif titik kuantum (QDSSCs) mempunyai kecekapan yang rendah disebabkan oleh penggabungan semula antara muka elektrolit-elektrod. Titik kuantum plumbum sulfida (PbS), titik kuantum plumbum cadmium sulfida ($Pb_xCd_{1-x}S$), titik kuantum kadmium sulfida (CdS) dan diikuti oleh salutan zink sulfida (ZnS) telah berjaya dimendapkan ke atas elektrod TiO_2 melalui kaedah penjerapan dan tindakbalas lapisan berturut-ion (SILAR) sebagai fotoanod bagi QDSSCs. Pemendapan $Pb_xCd_{1-x}S$ di antara lapisan teras PbS dan lapisan luar CdS akan mengurangkan penggabungan semula dan meningkatkan kecekapan bagi QDSSCs. Elektrod TiO_2 akan dibentuk dengan memendapkan TiO_2 berliang meso di atas kaca oksida timah berdopkan florin (FTO) setelah pengkalsinan pada suhu 450 °C. Sel suria disediakan dengan mengapitkan fotoanod TiO_2 berliang meso dengan fotokatod Cu_2S . Enam kitaran SILAR PbS, CdS, ZnS dan $Pb_xCd_{1-x}S$ serta lapisan pelbagai $PbS/Pb_xCd_{1-x}S/CdS/ZnS$ sampel, kesan kitaran SILAR bagi $Pb_xCd_{1-x}S$ dikaji dengan empat jenis pecahan molar, x iaitu 0.05, 0.1, 0.15 and 0.2. Pengukuran ketumpatan arus-voltan ($J-V$) mengesahkan kecekapan sel suria untuk empat kitaran SILAR lapisan $Pb_xCd_{1-x}S$ dengan pecahan molar, x dalam 0.05 bagi lapisan pelbagai $PbS/Pb_xCd_{1-x}S/CdS/ZnS$ sampel akan meningkatkan sebanyak 38.2 % apabila berbanding dengan lapisan pelbagai $PbS/CdS/ZnS$ sampel. In adalah kerana jurang jalur diperolehi bagi empat kitaran SILAR lapisan $Pb_xCd_{1-x}S$ dengan pecahan molar, x dalam 0.05 adalah antara jurang jalur diperolehi bagi lapisan teras PbS dan lapisan luar CdS dengan pengukuran UV-Vis spektra penyerapan. Selain itu, perangkap keadaan di antara lapisan teras PbS dan lapisan luar CdS dapat diturunkan

dengan pemendapan lapisan $Pb_xCd_{1-x}S$ dalam sampel lapisan pelbagai $PbS/Pb_xCd_{1-x}S/CdS/ZnS$. Antara sampel lapisan pelbagai $PbS/Pb_xCd_{1-x}S/CdS/ZnS$, empat kitaran SILAR bagi lapisan $Pb_xCd_{1-x}S$ dengan pecahan molar, x dalam 0.05 menunjukkan kecekapan sel suria yang paling tinggi iaitu 0.34 % apabila berbanding dengan empat kitaran SILAR bagi lapisan $Pb_xCd_{1-x}S$ dengan pecahan molar, x dalam 0.1, 0.15 and 0.2. Ini adalah disebabkan oleh empat kitaran SILAR bagi lapisan $Pb_xCd_{1-x}S$ dengan x dalam 0.05 mempunyai jalur konduksi yang tinggi akan membawa kepada suntikan electron yang lebih cepat dari $PbS/Pb_xCd_{1-x}S$ jalur konduksi ke elektrod TiO_2 . Oleh itu, penggabungan semula yang rendah akan diperolehi dan kecekapan sel suria akan meningkatkan. Di samping itu, empat kitaran SILAR bagi lapisan $Pb_xCd_{1-x}S$ memberikan kecekapan sel suria yang tinggi daripada enam kitaran SILAR bagi lapisan $Pb_xCd_{1-x}S$ dalam lapisan pelbagai $PbS/Pb_xCd_{1-x}S/CdS/ZnS$ sampel. Ini adalah disebabkan oleh pemendapan titik kuantum yang tinggi bagi $Pb_xCd_{1-x}S$ dengan enam kitaran SILAR lapisan $Pb_xCd_{1-x}S$ mengelakkan penusukan elektrolit dan menurunkan kecekapan sel suria dalam pengukuran $J-V$.

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FOR PbS/CdS/ZnS QUANTUM DOT SENSITIZED SOLAR CELLS**

ABSTRACT

Quantum dot sensitized solar cells (QDSSCs) have low efficiency due to the recombinations at electrolyte-electrode interfaces. Lead sulphide (PbS) quantum dots (QDs), lead cadmium sulphide ($Pb_xCd_{1-x}S$) QDs, cadmium sulphide (CdS) QDs and followed by coating with zinc sulphide (ZnS) were deposited on TiO_2 electrode as TiO_2 mesoporous photoanode using successive ionic layer adsorption and reaction (SILAR) method for QDSSCs. $Pb_xCd_{1-x}S$ QDs deposited between PbS core and CdS shell layer could reduce the recombination and improve the efficiency. TiO_2 electrode was formed with the deposition of TiO_2 mesoporous film on fluorine doped tin oxide glass (FTO) after calcination at 450 °C. The PbS QDs, $Pb_xCd_{1-x}S$ QDs, CdS QDs and coating with ZnS were formed on TiO_2 electrode with SILAR method. Solar cells were prepared by sandwiching the TiO_2 mesoporous photoanode with Cu_2S counter electrode. Six SILAR cycles of PbS, CdS, ZnS and $Pb_xCd_{1-x}S$ as well as multilayer of PbS/CdS/ZnS and PbS/ $Pb_xCd_{1-x}S$ /CdS/ZnS were prepared for characterizations. In multilayer of PbS/ $Pb_xCd_{1-x}S$ /CdS/ZnS, the effects of number of SILAR cycles of $Pb_xCd_{1-x}S$ were studied with four different molar fraction, x of 0.05, 0.1, 0.15 and 0.2. From current-density voltage (J-V) measurement, four SILAR cycles of $Pb_xCd_{1-x}S$ interlayer with molar fraction, x of 0.05 in multilayer PbS/ $Pb_xCd_{1-x}S$ /CdS/ZnS samples showed 38.2 % improvement in the efficiency when compared to the multilayer PbS/CdS/ZnS sample. This was because the band gap value obtained for four SILAR cycles of $Pb_xCd_{1-x}S$ interlayer with molar fraction, x of 0.05 were between band gap value of PbS core and CdS shell layer during UV-Vis spectrometer analysis. Moreover, the traps states were reduced between the PbS

core and CdS shell layers with the deposition of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer in multilayer of $\text{PbS}/\text{Pb}_x\text{Cd}_{1-x}\text{S}/\text{CdS}/\text{ZnS}$ sample. Among the multilayer $\text{PbS}/\text{Pb}_x\text{Cd}_{1-x}\text{S}/\text{CdS}/\text{ZnS}$ samples, four SILAR cycles of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer with molar fraction, x of 0.05 provided the highest efficiency of 0.34 % when compared with four SILAR cycles of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer with molar fraction, x of 0.1, 0.15 and 0.2. This is due to the conduction band of four SILAR cycles of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer with molar fraction, x of 0.05 was higher and lead to faster electron injection from the conduction band of $\text{PbS}/\text{Pb}_x\text{Cd}_{1-x}\text{S}$ to the TiO_2 electrode. Thus, lower recombination was obtained and the efficiency was improved. Besides that, four SILAR cycles $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer showed higher efficiency than six SILAR cycles of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer in samples with multilayer $\text{PbS}/\text{Pb}_x\text{Cd}_{1-x}\text{S}/\text{CdS}/\text{ZnS}$. This was owing to the high loading of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ QDs with six SILAR cycles of $\text{Pb}_x\text{Cd}_{1-x}\text{S}$ interlayer would prevent the penetration of electrolyte and decreased the efficiency in J - V measurement.

CHAPTER 1

INTRODUCTION

1.1 Introduction

Quantum dot sensitized solar cells (QDSSCs) are drawing much attention as a third generation solar cells that is derived from dye sensitized solar cells (DSSSCs). The attractive properties of the QDs over conventional dye used in the DSSSCs include its tunable band gap (Xie *et al.*, 2012), large intrinsic dipole moments for rapid charge separation and large extinction coefficient to reduce dark current and increase overall efficiency of solar cell (Emin *et al.*, 2011). Furthermore, the ability of QDs to produce multiple exciton generation (MEG) where a single absorbed photon can generate more than one electron hole-pair (Nozik, 2008).

In QDSSCs, deposition of QDs on the wide band gap of semiconductor of titanium dioxide (TiO_2) as sensitizers. Mesoporous structure is made for TiO_2 wide band gap semiconductor to provide large surface area for absorption of more QDs in harvesting visible light effectively (Chen *et al.*, 2012). TiO_2 is an attractive material for the solar cells application because of surface photochemistry, physical and chemical stability of the semiconductor material. TiO_2 is an n-type semiconductor with wide band gap of 3.0 eV for rutile and 3.2 eV for anatase. The anatase is preferred to use in solar cells because of the anatase higher mobility and its catalytic properties (Scanlon *et al.*, 2013). The conduction band, E_{cb} of anatase TiO_2 is -4.21 with respect to absolute vacuum scale (AVS) or it is named as E_{cb} -4.21 eV versus vacuum (Xu and Schoonen, 2000). The wide band gap of bulk TiO_2 absorbs portion of solar spectrum from ultraviolet down to ~ 400 nm.