

OPTIMIZATION OF INKJET PRINTING FOR COPPER INDIUM GALLIUM
SELENIDE SOLAR ABSORBER THIN FILM

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Master of Philosophy

Faculty of Science
Universiti Teknologi Malaysia

SEPTEMBER 2018

I don't know what my destiny will be, but one thing I know, I am really happy spending my time towards it.

ACKNOWLEDGEMENT

It has been almost two years to conduct experiment and finalize my thesis, including the designing, construction, execution, performance, optimization and interpretation. Time flies by, the world keeps changing all the time. Thinking back to two years ago, the first I landed at this country, no friends, no relatives, no one to show me where UTM is, transformed from Chinese style to Malaysian atmosphere suddenly. I thought I could survive on my own, because I traveled to many places. But, these places are in China, not another language-spoken country. It was first time to come abroad, not well-planned. But, I am proud of my two main characteristics: persistent and rebellious. The former character in my blood makes me survive here, overcomes plenty of inconveniences, difficulties and live like a native. The other one makes me think differently, that gives me a lot of crazy and innovative ideas.

I really appreciate my research group. Prof. Jalil Ali group really support me a lot not only in academic, also in carrier aspect. They guide me and show me the way to be a researcher step by step. They open my mind and teach me to think in many aspects. Many thanks to my senior Elham Mazalan. The programme and electronic parts of my project cannot be finished without his help. I am grateful to my supervisor Dr. Kashif, because I really give him lots of trouble, refuse him many times of writing papers and conducting projects, hence that I can manage my time to focus more on my interests.

I would be most grateful to my parents. I've never worried about my living cost and tuition fee issues, so that I can devote all my time and energy to research. I love them.

ABSTRACT

The present research is focused on development and optimization of inkjet printing technique to print Copper Indium Gallium Selenide (CIGS) solar cell absorber thin film on rigid substrate. An office inkjet printer is modified to print specific CIG ink on soda lime glass substrate. Metal nitrates are used as precursors and dissolved in a solvent mixture of methanol and ethylene glycol. The ink viscosity, printing process, printing layer and substrate temperature are optimized to print uniform and compact CIGS thin film. The CIG ink viscosity of 3.7 cP and one-time drying approach are chosen to avoid coffee ring, achieve good adhesion, compact and uniform thin film formation. After optimizing of CIG ink viscosity and printing process, the effects of printing layer, and substrate temperatures in the range of 50 °C to 80 °C are investigated. Thin films are printed with similar crystallinity, compound structures, and elemental composition. The electron scanning micrographs have shown that the surface coverages for 3-layer printed thin films are not as good as 4-layer printed thin films due to deficiency of raw material. The CIGS grains grow freely in 3 dimensions and result in relatively large grain size. Among all 3-layer printed films, the substrate temperature of 70 °C has shown thickness of 0.9 μm , better light absorption, relatively large grain size of 797 nm with good surface coverage up to 92%, and surface roughness of 112.4 nm. On increasing the number of printing layer to 4, thin film printed at 70 °C shows thickness of 0.8 μm , grain size of 679 nm, good light absorption, 98% surface coverage, and surface roughness of 77.5 nm. Printed CIGS thin films are compared with film deposited by spray coating. The CIGS film deposited by spray coating has shown thickness of 1.5 μm , grain size of 802 nm, poor surface coverage of 78%, worse light absorption, and rough surface condition of 138.1 nm. The comparison between spray and inkjet printing reveals the probability of inkjet printing technique has great potential for deposition of solar thin films.

ABSTRAK

Projek penyelidikan ini memberi fokus terhadap pembangunan dan pengoptimuman teknik percetakan sembur dakwat untuk mencetak penyerap saput tipis bagi Tembaga Indium Galium Selenida (CIGS) pada substrat tegar. Pencetak sembur dakwat pejabat diubahsuai untuk mencetak dakwat CIG yang tertentu pada substrat kaca. Logam nitrat digunakan sebagai pelopor dan dilarutkan dalam campuran pelarut metanol dan etilena glikol. Kelikatan dakwat CIG, proses percetakan, lapisan percetakan dan suhu substrat dioptimumkan untuk mencetak saput tipis yang seragam dan padat. Kelikatan dakwat CIG 3.7 cP dan pendekatan pengeringan satu kali dipilih untuk mengelakkan daripada kesan gelang kopi, mencapai lekatan yang baik dan pembentukan saput yang padat dan seragam. Selepas mengoptimumkan kelikatan dakwat CIG dan proses percetakan, kesan lapisan percetakan, dan suhu substrat dalam julat 50 °C hingga 80 °C dikaji. Saput yang dicetak mempunyai kehabluran struktur sebatian dan komposisi unsur yang serupa. Mikrograf imbasan elektron menunjukkan saputan 3-lapisan mempunyai kekurangan bahan mentah, liputan permukaan tidak sebaik saputan 4-lapisan. Tetapi butiran boleh tumbuh dengan bebas dalam 3 dimensi dan menghasilkan saiz butiran secara relatif yang besar. Antara semua saputan bercetak 3-lapisan, suhu substrat pada 70 °C telah menunjukkan ketebalan 0.9 μm , penyerapan cahaya yang lebih baik, saiz butiran secara relatif sebesar 797 nm, liputan permukaan yang baik sehingga 92%, dan kekasaran permukaan sebesar 112.4 nm. Sebaik sahaja bilangan lapisan percetakan meningkat kepada 4, saput yang dicetak pada 70 °C menunjukkan ketebalan 0.8 μm , saiz butiran 679 nm, penyerapan cahaya yang baik, liputan permukaan yang lebih baik sebanyak 98%, dan kekasaran permukaan 77.5 nm. Saput nipis CIGS yang dicetak dibandingkan dengan saput yang termendap menggunakan penyalutan semburan. Saput CIGS yang dimendapkan oleh penyalutan semburan menunjukkan ketebalan 1.5 μm , saiz butiran 802 nm, liputan permukaan lemah 78%, penyerapan cahaya yang lebih lemah, dan keadaan permukaan kasar sebesar 138.1 nm. Hasil perbandingan antara percetakan semburan dan semburan dakwat menunjukkan kebarangkalian teknik percetakan sembur dakwat berpotensi besar untuk pemendapan saput tipis solar.

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LIST OF ABBREVIATIONS

TWh	-	Terawatt hours
AM	-	Air Mass
AR	-	Anti-reflective
AZO	-	Aluminum-doped zinc oxide
i-ZnO	-	Intrinsic zinc oxide
CBD	-	Chemical bath deposition
CGI	-	Cu/(In+Ga) ratio
GGI	-	Ga/(In+Ga) ratio
c-Si	-	Crystalline silicon
PVD	-	Physical vapor deposition
DC	-	Direct current
RF	-	Radio frequency
mm	-	Millimeter
cm	-	Centimeter
μm	-	Micrometer
nm	-	Nanometer
JV	-	Current density-voltage
J_{sc}	-	Short circuit current density
V_{oc}	-	Open circuit voltage
η	-	Efficiency of solar device
FF	-	Fill factor
PCE	-	Power conversion efficiency
PID	-	Proportional integral derivative
PL	-	Photoluminescence

PV	-	Photovoltaics
EQE	-	External quantum efficiency
R2R	-	Roll to roll
SEM	-	Scanning electron microscopy
SLG	-	Soda lime glass
TCO	-	Transparent conducting oxide
XRD	-	X-ray diffraction
FTIR	-	Fourier-transform infrared spectroscopy
FESEM	-	Field emission scanning electron microscopy
EDS	-	Electron dispersive X-ray spectroscopy
MER	-	Methanol/Ethylene glycol ratio
cP	-	Centipoise
TCL	-	Three phase contact line
V_{Cu}	-	Copper vacancy
OVC	-	Ordered vacancy compound
JCPDS	-	Joint committee on powder diffraction standards

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Due to the fast development, the demand of the energy is rising dramatically. Nowadays, 20×10^3 TWh electric energy is required worldwide every year. Electrical energy is mainly produced from fossil fuel (approximate 2/3 of total) [1]. It is predicted that if the current global energy consumption pattern continues, the world energy consumption will be increased by 50% at the end of 2030 [2]. Currently, the main source of energy is fossil fuel which is depleting at an alarming rate. These fossil fuel resources will not only be completely depleted in the near future, but also posing a serious threat to environment due to emission of pollutants [3]. The growing demand for energy and the increase in environmental pollution requires not only the optimal use of conventional sources, but also replacement with renewable energy resources gradually. The renewable energy has the capability of meeting energy demands as well as mitigating issues related to the global warming. Among renewable energy resources, solar energy is one of the most abundant, inexhaustible and clean sources of energy. Unlike nuclear power, there is no radiation threat, and as in case of hydropower or wind energy, a huge budget is not necessary to build solar panels and to choose the certain geographical area. The solar cell technology is capable to bring cleaner energy generation right to the source of demand. For solar power, in one day, the irradiation from the sun on the earth gives about 10,000 times more energy than

the daily use of us [4]. However, how to collect solar energy at reasonable cost is the biggest challenge.

Photovoltaics (PV) solar cells are capable to convert solar energy directly into electricity without any pollutant emission during operation [5]. Silicon is the most commonly used material for PV solar panels [4]. Monocrystalline silicon solar panel is the oldest, more efficient but expensive than the polycrystalline silicon and thin film PV panel techniques. The silicon module record efficiency of 24.1% is kept by SunPower (USA) company. Monocrystalline PV is made from single cell silicon crystal, and the Czochralski process of making single cell crystal is one of the most complex, time consuming and costly part to fabricate whole PV devices. The high cost and fragility of monocrystalline and polycrystalline silicon limit the deployment of PV technology on large-scale and far from affordable range for common people.

Thin film solar cell is second generation solar cell consists of very thin absorber layer, from a few nanometers to tens of micrometers, much thinner than silicon solar cell which is about 200 μm . This allows solar cell to be flexible, lower in weight and results in a low level of material and energy consumption during production [6]. Among all thin film solar cells, Cu (In, Ga)Se₂ (CIGS) thin film solar cell technology has a great potential for low-cost fabrication and high-performance, recently achieved about 22.6% at cell level on a rigid glass substrate, while 20.4% has obtained on flexible polyimide substrate [7,8]. CIGS compound is a direct bandgap material with a light absorption coefficient higher than 10^5 cm^{-1} which can be controlled by varying the In/Ga and S/Se ratios [9]. These inherent material characteristics such as tunable band-gap, lower energy payback time as compared to silicon solar cell, thermal and chemical stability compared to organic solar cells or organic/inorganic hybrid solar cells, CIGS is considered as promising candidate in thin film solar cells technologies.

Until now, all the record efficiencies of CIGS are all produced by vacuum deposition techniques [7,8,10,11]. Vacuum technique has high degree of accuracy and can deposit good quality of thin film. But the equipment is expensive and process is

complex. In contrast, the non-vacuum technique is low cost and capable for large-scale, roll-to-roll manufacturing [12–14].

1.2 Problem Statement

Despite of significant advancement in the field of solar cell industry, PV solar modules are still not widely deployed due to two major factors: 1. current cost of the solar system as compared to power generation systems derived from fossil fuel and 2. conversion efficiency of solar cell [6]. These challenges can be addressed by enhancing the conversion efficiency and reducing the fabrication cost of the solar cell. Among different types of solar cells, thin film solar cells have high efficiency with thickness of only few micrometers. Among different types thin film solar cells such as CIGS, CdTe and amorphous silicon solar cell, CIGS shows potential for higher efficiency and less impact to the environment.

Usually, the record performance of the CIGS solar cell is demonstrated on small-area (less than 1 cm²) fabricated by vacuum-based growth techniques such as sputtering and evaporation [11,15]. To produce high efficiency solar cell on large-area at low cost is crucial for the future development of the CIGS technology [16]. Number of non-vacuum alternative deposition techniques such as spray coating, dip coating, spin coating, electrodeposition etc. have been developed to deposit CIGS solar thin film, which require non-expensive vacuum equipment, less energy intensive deposition and much better material utilization [14,17–19]. However, each of the existing non-vacuum technologies suffers from one or more major deficiencies, from utilization of toxic materials, insufficient cell efficiency, lack of stability or durability and effective optimized deposition [20,21].

Therefore, this research focuses on the modification and optimization of the inkjet printing technique to deposit CIGS solar cell absorber layer. Inkjet printing

process is maskless, more than 99% materials utilization, non-contact deposition technique and capable for printing on a pre-existing pattern on a substrate. To the best of our knowledge, there is no report describing printing CIGS thin film by an office inkjet printer. In this study, an office inkjet printer is modified to optimize the thin film deposition in context of thin film structure, morphology, light absorption and thin film roughness.

1.3 Objectives

The main objective of this research work is to modify and optimize inkjet printing technique to grow CIGS solar thin films and compare with spray coating method. The specific objectives of the study are:

1. To modify and optimize an inkjet printing system for the deposition of CIGS solar thin films on glass substrate.
2. To optimize inkjet printing processes in context of ink viscosity, printing process, thin film thickness and substrate temperature to deposit thin film with good crystallinity, surface coverage and light absorption performance.
3. To compare thin films deposited by inkjet printing and spray coating.

1.4 Scope of Study

This research project is focusing on modification of inkjet printing technique to deposit CIGS absorber layer. To optimize inkjet printing technique, different CIG ink viscosities (3.2, 3.7, 4.7 and 6.5 cP), substrate temperatures (40, 50, 60, 70 and 80 °C), printing process (separate drying and one-time drying process) and printing layers (2, 3, 4, 5 and 6 layers) towards high performance are investigated and optimized. The film thickness is characterized by surface profiler. FESEM with EDS analysis is

conducted to study the thickness, composition and morphology of the thin film surface. AFM is used to study the surface roughness. XRD and FTIR are used to investigate the crystallinity and compound composition, respectively. UV-Vis-NIR is used to characterize the optical absorption of thin films.

1.5 Significances and Original Contributions of This Study

This research work contributes to knowledge particularly at designing and development of cost-effective inkjet printing technology which could lead towards the development of affordable CIGS cells and even modules. The increase in solar technology installation reduces fuel poverty and inherently improve the security of energy supply which is pollution free and eco-friendly technology. Nowadays, due to shifting of media from papers to electronic means, the existed printing industry can be utilized to print solar cell which can lower the production cost of PV solar modules significantly and lead to large scale development of solar technology using existing machineries.

1.6 Thesis Structure and Organization

The research background, problem statement, objectives, scope, significant of this research are presented in Chapter 1. The overviews of solar cell, deposition techniques, introduction of inkjet printing and spray coating are discussed in Chapter 2. The processes of performing experiment and details of experiment are written in Chapter 3. The obtained results and discussion are presented in Chapter 4. And the conclusion and future recommendation are given in Chapter 5.

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