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REVIEW OF RECENT RESEARCH ON PENTERNARY NANOSTRUCTURED THIN FILMS

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ABSTRACT

Nowadays, thin film technology has received an enormous interest among the scientists and was used in solar cell application. Solar energy is free, renewable energy that has been employed in the world. In the past, a variety of thin films (binary, ternary and quaternary films) have been produced in order to search cheaper absorbing materials. The obtained thin films are more flexible, high conversion efficiency and less expensive if compared to silicon based solar cells. The aim of this work is to study the synthesis of penternary nanostructured thin films by using several methods as reported by different researchers from around corner of the world (Physical and chemical technique). The obtained samples such as $Cu(In,Ga)(S,Se)_2$ and $Cu_2ZnSn(SSe)_4$ films have been characterized by using various tools. The solar cell devices show power conversion efficiency of 10 % and 12.4 % for $Cu(In,Ga)(S,Se)_2$ and $Cu_2ZnSn(SSe)_4$ thin films, respectively.

Keywords: thin films, pen ternary compound, solar cells, power conversion efficiency.

INTRODUCTION

The strong interest in new forms of transparent materials deposited at low temperature on conducting and non-conducting substrates is named as thin film technology [Khan *et al*, 2011; Guziewicz *et al*, 2004; Siang *et al*, 2011; Anuar *et al,* 2011; Sinha *et al,* 2018]. Thin film layer is a material ranging from the fractions of nanometer to micrometer in different thicknesses [Ho, 2015; Asenjo *et al,* 2010; Gopakumar *et al*,2010;Raniero *et al,* 2010].Nowadays, metal telluride, metal sulphide and metal selenide are important semiconductor materials (metal=Cu, Ni, Sn, Zn, Sb, Mg).These semiconductor materials have been observed to be potential candidates for electronic, photonics, optoelectronic devices, solar cells [Ullah *et al*, 2018; Kelvin *et al*, 2011; Khallaf *et al*, 2008; Ubale, 2010; Song and Lee, 2009; Monohorn *et al*, 2010], solar selective coatings, resistive switches, filament memories, and infrared detectors. Power conversion efficiency of solar cells was conducted and reported by many researchers (Table-1). Many different synthesis methods (chemical vapor deposition, ion beam deposition, solid gas reactions, electrodeposition, chemical bath deposition, molecular beam epitaxy, thermal evaporation, vacuum evaporation, spray pyrolysis, photochemical deposition, spin coating, atomic layer deposition, and successive ionic layer adsorption and reaction method) have been developed to prepare these materials [Liu *et al,* 2018; Shanthi *et al,* 2010; Bhalerao *et al*, 2016; Noraini *et al*, 2010; Gwee *et al*, 2010].

In this work, preparation and characterization of penternary thin films using various deposition techniques were reported. The main results of works will be highlighted and discussed.

Table-1. Power conversion efficiency of different types of thin films.

Researcher(s)/year	Thin films	Power conversion efficacy (%)
Green and co-workers, 2016	CdTe	22.1
Barote and co-workers, 2011	PbS	0.04
Ahire and co-workers, 2006	Bi_2S_3	0.056
Shinde and co-workers, 2014	$CdSe_{0.6}Te_{0.4}$	0.43
Li and co-workers, 2016	Cu_2ZnSnS_4	3.8
Lugo and co-workers, 2015	$CuIn(S,Se)_2$	2.3
Subramaniam and co-workers, 2014	Cu_2ZnSnS_4	1.1
Sarah and co-workers, 2009	Sb_2S_3	0.7
Santanu and co-workers, 2012	copper zinc tin selenide	9.1
Jaramillo and co-workers, 2015	SnS	4.36
Goto and co-workers, 2004	$CuGaS_2$	13
Chen and co-workers, 2014	$CuInS_2$	5.3
Ho, 2017	$Ni_3Pb_2S_2$	2.7

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LITERATURE SURVEY

Nowadays there is an extended interest on the research of different synthesis methodologies for thin films. These quaternary (Table-2), ternary (Table-3) and binary (Table-4) materials are preferred for their excellent structural, compositional, optical, morphological, electrical, and photovoltaic properties.

Table-2. Structure, morphology and optical properties of quaternary thin films.

Quaternary thin films	Structure property	Morphology property	Optical property	Researcher(s)/year
CZTS (Cu_2ZnSnS_4)	Kesterite phase. Minor impurity phases (SnS, CuS, SnS_2) were detected.	Grain size was about 1 µm. SEM images show darker (rich in copper) and brighter (rich in zinc) area.	Band gap was 1.52 eV	Tara and co-workers, 2014
CZTS	Kesterite (copper molar ratio=1.6) and secondary phase such as CuS (higher copper molar ratio) were prepared using different conditions.	A similar morphology was detected in all samples	The band gap was estimated 1.5 eV.	Lin and co-workers, 2016
CZTS (Cu_2ZnSnS_4)	Kesterite crystal structure without the formation of secondary phase.	Surface was denser and surface was coarser were observed when the oxygen content in the film was higher and lower, respectively	Higher oxygen content (10.3 %) in the thin films resulted in a higher band gap (2.84 eV) as compared to films with oxygen content of 8.89 % (2.75 eV).	Yu and co-workers, 2016
Cu_2FeSnS_4	Intensity of the (112) plane was increased as the substrate temperature was increased	The films show dense and uniform morphology	Direct band gap value was estimated to be 1.5 eV	Adelifard, 2016
Cu_2NiSnS_4	The obtained films show single phase	Uniform morphology with grain size (1 micrometer)	Band gap was 1.2 eV	Chen and co-workers, 2016
Cu_2CoSnS_4	Tetragonal phase (without impurities)	Uniform flower-like spherical and grain size about 1.5 micrometer	--	Zhong and co-workers, 2015

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Table-3. Structure, morphology and optical properties of ternary thin films.

Ternary thin films	Structure property	Morphology property	Optical property	Researcher(s)
$CuInTe_2$	Tetragonal structure with preferred orientation along (112) plane for the films prepared for 5, 10 minutes	--	Band gap value reduced from 1.06 to 0.99 eV when the deposition time was increased (5 to 20 minutes)	Meglali and co-workers, 2018
$CuInSe_2$	Sphalerite structure was observed. The most intensive peak corresponding to (112) plane	Uniform surface morphology (films prepared at -7 and -8 V) and irregular shape of grains (when the deposition potential is -6 V) could be seen.	The band gap obtained in experiment was 0.96 (-6 V), 1.09 (-7 V) and 1.03 eV (-8 V).	Bouraiou and co-workers, 2011
$CuInS_2$	Tetragonal chalcopyrite with the crystallize size (12.8-61.2 nm) were measured for the films prepared under 15-24 hours.	Thicker films (348 nm) produced at longer time (24 hours) if compared to thinner films (267 nm) at 15 hours.	Band gap values are 1.46 (15 hours) and 1.4 eV (24 hours)	Lugo and co-workers, 2014
$FeCdS_3$	The films prepared using tartaric acid showed amorphous structure. The films deposited using EDTA and acetic acid indicated polycrystalline.	Morphology properties strongly depend on the presence of complexing agent such as EDTA (web-like architecture), tartaric acid (homogenous dense with needle like grains) and acetic acid (vertical rod-like).	The highest (2.63 eV) and the lowest band gap (2.45 eV) were found in the presence of EDTA and acetic acid, respectively	Ubale and Ibrahim, 2012
Cu_4SnS_4	Orthorhombic structure with the predominant peak at $2\theta=30.1°$	Small grain size and uniform morphology for the films prepared at 25 °C and 45 minutes.	Direct band gap (1.68 eV) was observed.	Anuar and co-workers, 2009
$Cd_{1-x}Zn_xS$	Cubic (when x more than 0.85) and hexagonal structure (x<0.85) was detected	--	Band gap value varies from 2.4 to 3.48 eV depending on the mole ratio used	Lee and co-workers, 2003
$AgSbS_2$	As-deposited films (300 K) indicate amorphous. However, better crystallinity in higher temperature (373 K). Binary phase such as AgS_2 appeared at 500 K	--	The direct band gap decreases (2 to 1.77 eV) as the temperature is increasing	Ibrahim, 1995
Cu_2SnS_3	Monoclinic phase was obtained	Homogeneous and bigger grains (2-6 μm)	Band gap of 0.93 eV was measured	Berg and co-workers, 2012

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Table-4. Structure, morphology and optical properties of binary thin films.

Binary thin films	Structure property	Morphology property	Optical property	Researcher(s)
MnS	Hexagonal phase with lattice parameters, a=b=3.9 Å and c=6.4 Å	Spherical grain particles with different sizes (ranging from 1.6 to 7 micrometer)	Direct (3.67 eV) and indirect band gap (2.7 eV) value are obtained	Sunil and co-workers, 2017
CuS	Hexagonal structure with the crystallite size (11-13 nm)	The films show smooth, free cracking and homogeneous	Direct (2.2 to 2.5 eV) and indirect band gap (1.8-1.9 eV) values were found	Chaki and co-workers, 2014
MgS	Mochonic phase with the preferential peak corresponds to (010) plane.	--	Direct band gap values (2.25 to 2.75 eV) strong depend on the concentration of solution.	Ezenwa and co-workers, 2015
CoSe	Cubic structure and crystallite size was 30.6 nm	The obtained films exhibit well define boundaries and homogeneity morphology	Direct band gap value increases as the pH increasing from pH 8.4 (2.8 eV), 8.7 (2.96 eV) to 9.6 (3.1 eV).	Agbo and co-worker, 2016
NiSe	The number of NiSe peak (rhombohedral) was increased with the increase in pH (up to pH 11)	Irregular shaped with diameter (1-10 micrometer) was observed form the films prepared at pH 11	The obtained films exhibit band gap of 1.8 eV	Mohd and co-workers, 2010
CdS	Single phase of hexagonal structure was detected	Uniform morphology with grain sizes (8.4 to 57.2 nm)	Direct band gap value (2.42 to 2. 36 eV) based on optical absorption data	Meshram and co-workers, 2012
PbS	Cubic structure and the most prominent peak corresponding to (200) plane.	uniform morphology consists of several different grain sizes (208 to 320 Å)	--	Seghaier and co-workers, 2006
ZnSe	Cubic structure and crystalline size was 44 nm	Uniform spherical grain was highlighted	Direct band gap (2.78 eV) was measured	Kashif and co-workers, 2014

$Cu(In,Ga)(S,Se)_2$ or CIGSS thin films have a great potential for reaching high efficiency at low production cost, so that are used in solar cell applications. The study of internal wave propagation and photon absorption rate was done [Richter *et al*, 2013] by using technology computer aided design simulator. During the experiment, molybdenum films are used as metallic back electrode. The reflectance of the $MoSe_2$/air interface with mean square error equals to 2.42, to support that these absorbing materials could be used for the solar cell application.

The $Cu(In,Ga)(S,Se)_2$ thin films have been prepared using physical vapor deposition method. It is fundamentally a vapourization coating technique, involving transfer of material on an atomic level. This process is carried out under vacuum conditions, in which material vapourized from a source is transported in the form of vapour through a vacuum or low pressure gas environment to the substrate where it condenses. Generally, researchers found that this technique indicates more environmentally friendly if compared to other deposition techniques, however, it operates at very high temperatures and vacuum conditions. Research results show the band gap energy was about 1.5 eV for the films prepared under different Sulphur and gallium contents [Mario and William, 2005]. They reported that power conversion efficacy of 10 % could be reached.

The co-evaporation technique has been used to synthesize $Cu(In,Ga)(S,Se)_2$ films [Keller *et al*, 2013]. The atom probe tomography (APT) was used to study the chemical composition of obtained films in nanometer scale resolution and 3-dimensional visualization. Researchers highlighted that the selenium concentration (45-47 %) was underestimated (using APT technique) than energy dispersive analysis X-ray (EDX) results.The fabrication of glass/Mo/CIGSSe/CdS/i-ZnO/ZnO:Althin film solar cells was described [Ingo et al. 2014]. It was found that the band gap value becomes smaller for the thinner sample (increasing of saturation current density).

Electro deposition is becoming popular deposition technique due to able to grow films over large areas of substrates and lower cost of production. A typical set-up as indicated in Figure-1. It is an electrochemical process in which the anode and a cathode are immersed in a suitable electrolyte and the passage of electric current serves to deposit the material on the cathode. Generally, electro deposition was performed in electrochemical cell contains three electrode system, namely working electrode, reference electrode (silver/silver chloride, and standard hydrogen electrode) and counter electrode. Therefore, it is used to synthesize Cu(In,Ga)(S,Se)$_2$ thin films [Taunier *et al*,2005]. A wide range of sulphur content was observed (S/(S+Se) atomic ratio from 0 % to greater than 90 %.). The grain size becoming bigger as the sulphur content increased.

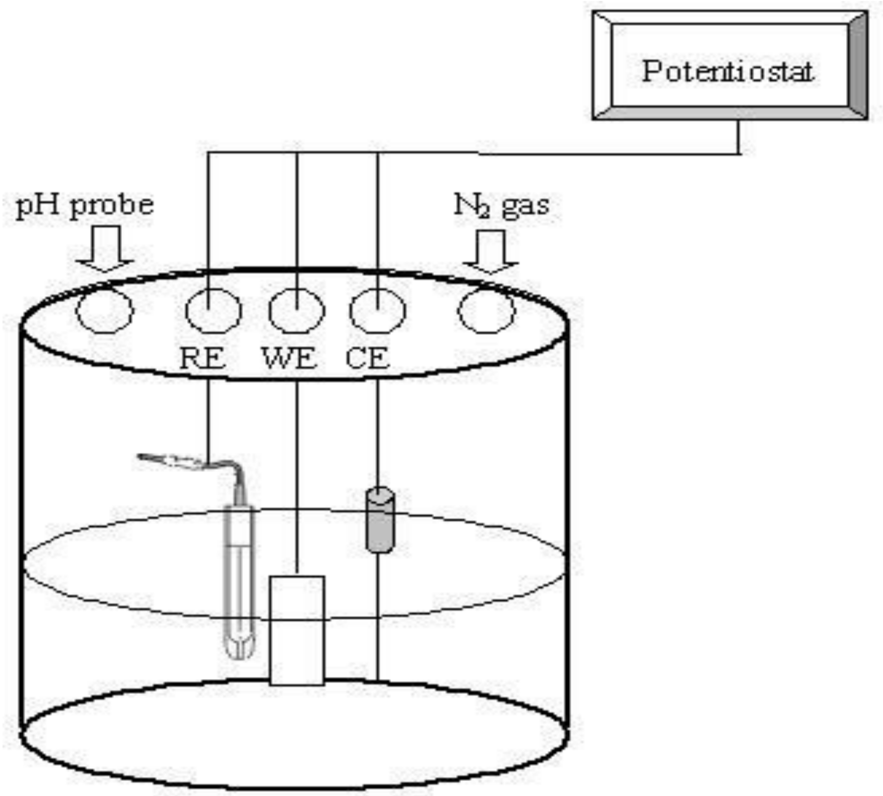

Figure-1. The electrodeposition method set-up.

Lower toxicity solution based process was employed to grow $Cu_2ZnSn(SSe)_4$ thin films on substrates [Guo *et al.*2015]. The grain sizes about 0.5 to 2 µm with a thickness of 1640 nm were determined using scanning electron micrograph. The 6.4 % (power conversion efficiency) and 0.554 (fill factor)in the selenized films based on photo electrochemical cell measurements. Other PCE values also revealed by other scientists including Wang *et al.*, 2013 (6.03 %), Jiang et al., 2013 (6.2 %), Schnabel *et al.*, 2013 (7.5 %), Kim et al., 2014 (12.4 %), Tian *et al*., 2014 (6.83 %) and Zhang et al., 2015 (7.86 %).

The well crystalline $Cu_2ZnSnS_xSe_{4-x}$ thin films have been prepared by using solution technique and heating process [Chen *et al,* 2014] to enhance power cell efficiency in dye sensitized solar cell. They observe that porosity and grain boundaries were decreased with the enhanced crystallization, lead to improve the charge transport as well.

Hot injection method was used to prepare $Cu_2CoSn(SeS)_4$ and $Cu_2ZnSn(SeS)_4$ films as counter electrode in dye-synthesized solar cells. Better electro catalytic behaviour (reduction of iodime/iodide electrolyte) could be observed for the $Cu_2CoSn(SeS)_4$ films if compared to $Cu_2ZnSn(SeS)_4$ films, even though these materials have single crystalline, stoichiometric ratio and 18-25 particle size.The $Cu_2CoSn(SeS)_4$ films have an efficiency of 6.47 %as shown in photovoltaic result and band gap of 1.18 eV, while $Cu_2ZnSn(SeS)_4$ films achieved efficiency of 3.18 % and band gap value of 1.45 eV as shown in optical property [Ozel *et al*, 2016].

CONCLUSIONS

There are several deposition methods such as solution deposition method, electrodeposition and evaporation technique have been used to prepare various types of penternary thin films (including Cu(In,Ga)(S,Se)$_2$ and $Cu_2ZnSn(SSe)_4$ thin films). The obtained films were characterized by using different tools such as x-ray diffraction, scanning electron microscopy, UV-Visible spectrophotometer. The experimental results indicate that these samples could be used in photovoltaic applications (as cheaper absorbing materials).

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