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R. Abderrassoul

Electronics and Communications Engineering Department, Faculty of Engineering, Mansoura University, Mansoura, Egypt.

S. Rassoul

Faculty of Engineering, Mansoura University, Mansoura, Egypt.

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II - VI Compound Semiconductor Polycrystalline Thin Film Solar Cells : A Review

استعراض لخلايا الطبقات الرفيعة الشمسية المصنوعة من أشباه الموصلات II-VI

R. A. Abderrassoul and S. M. A. Rassoul,
Faculty of Engineering, Mansoura University,
Mansoura, E G Y P T.

الخلاصة - نعطي استعراضا موجها لخلايا الطبقات الرفيعة المبنية على أشباه الموصلات المركبة II-VI والسياتلك ثلاثية العناصر المشتقة منها . وتعتبر هذه المواد أحسن المرشحين للتطبيقات الأرضية للخلايا الشمسية . نستعرض طرق الترسيب المختلفة وتأثيرها على الخصائص الإلكترونية والضوئية للمادة ، وكذا تأثير المعالجات الحرارية ، ونقارن الأداء الجهد - ضوئي للخلايا الشمسية المختلفة المنشورة المبنية على هذه المواد .

Abstract: Thin - film polycrystalline solar cells are very promising for terrestrial applications. II - VI compound semiconductors are very good candidates, both in homojunction and heterojunction structures. Various techniques are reported for the deposition of thin films of these materials. Ternary semiconductors have gained wide application in thin film solar cells, and the highest photovoltaic conversion efficiency for a thin film compound solar cell has been reported for a $CuInSe_2$ - based cell. Quaternary alloys are starting to be considered for tailoring the device using varying energy bands and getting optimum lattice match. Postdeposition heat treatment is very important in determining the electrical characteristics of the film.

I. Introduction :

The photovoltaic (PV) effect (the direct conversion of photon energy into electricity) was first discovered in the last century, and the first silicon photovoltaic cell was made in Bell Laboratories, USA, in 1954. However, the application of solar cells has been limited to space applications, mainly because of their high cost.

To develop an alternative source of renewable energy low - cost high - efficiency PV systems had to be developed. Worldwide research was conducted to meet that goal, including research to develop low - cost high-efficiency thin film solar cells (amorphous silicon and polycrystalline non - silicon thin films), to improve the efficiency of single crystalline silicon solar cells, and to develop high efficiency concentrator solar cells.

II. Thin - film Solar cells :

Thin - film polycrystalline solar cells are very promising because of their inherently low fabrication cost , adaptability to high - speed mass production, and potential of moderate - to high conversion efficiency. II - VI compound semiconductors (with bandgaps of 1.1 - 1.6 eV) offer very good candidates for thin - film solar cells because they are direct bandgap semiconductors and because of their excellent match to the solar spectrum [1], as shown in figure (1), and therefore can be used as collectors or window materials for solar cells. Three of them (Cu_2S / CdS , $CuInSe_2 / CdS$, and $CdTe/CdS$) showed conversion efficiencies in the 5 - 8 % range in 1977 in an all thin film form [2]. Also because of their large, direct bandgaps, they are useful in luminescent devices and are used in CRT's [3], as well as in the emerging electroluminescent (EL) devices, while $HgCdTe$ has been extensively used for infrared detectors.

Figure (2) shows the optical absorption coefficient for some highly absorbing compound semiconductors, as well as for amorphous and polycrystalline silicon [4].

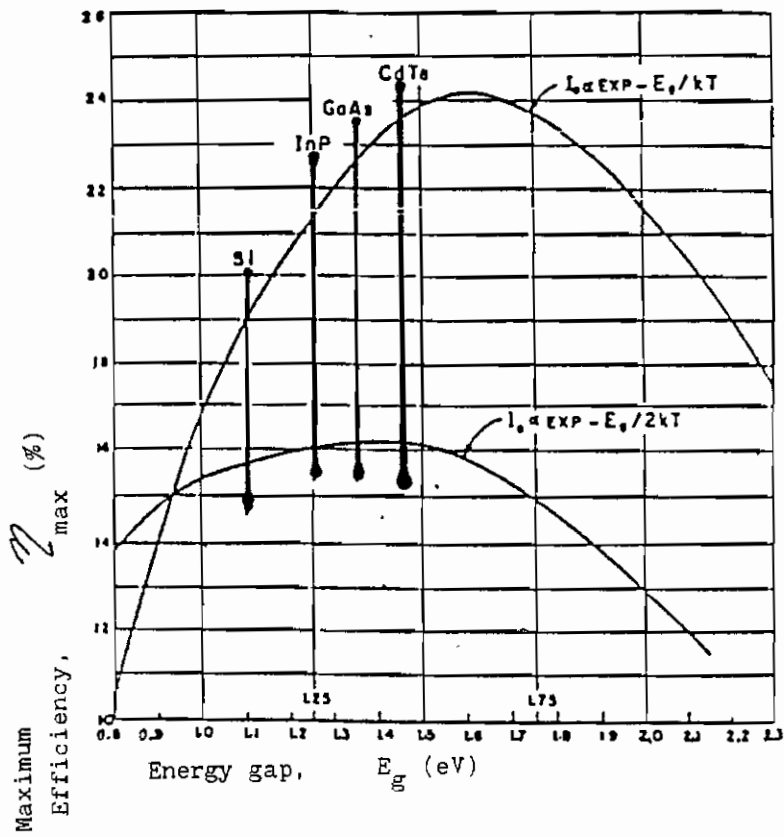


Figure (1). The maximum photovoltaic conversion efficiency, η_{\max} vs. energy gap, E_g . [1].

III. Solar Cell Materials :

i) Cadmium Telluride :

One of the important materials considered for photovoltaic applications is Cadmium Telluride, CdTe, because it has a near optimum bandgap energy (1.47 eV), and a high optical absorption coefficient at the band edge ($3 \times 10^4 \text{ cm}^{-1}$) [5], as shown in figure (2), and because it can be prepared in thin film form of both conductivity types (p and n). CdTe films for photovoltaic applications have been prepared by chemical - vapor deposition (CVD) [6], close - space vapor transport (CSVT) [7,8], vacuum evaporation [9,10], electrodeposition [11], screen-printing [12,13], metalorganic chemical - vapor deposition (MOCVD) [14], sputtering [15,16], spray pyrolysis [17,18], and molecular - beam epitaxy (MBE) [19,20].

ii) Other II-VI semiconductors :

Thin films of other compound semiconductor materials have been reported using different deposition techniques. ZnS films have been grown by plasma - assisted metal-organic chemical vapor deposition [21], and by low-pressure MOCVD [22], ZnSe had been formed using evaporation [23], and vapor - phase epitaxy [24], atmospheric [25,26] and low - pressure [27] MOCVD, and close - spaced vapor transport [28]. ZnTe films were reported using vacuum evaporation [29] and low - pressure MOCVD [30], while CdSe films have been grown by the gas-solid process [31].

iii) Ternary compound semiconductors :

Ternary chalcopyrite compound semiconductors of the II - VI class are also very promising candidates. An example of these materials is CuInSe_2 (CIDS), and its variations ($\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2 / \text{Cd}_{1-x}\text{Zn}_x\text{S}$) [32-35], which offer important candidates for the absorber region in a thin - film PV device because of its very high optical absorption coefficient [4].

Thin film CuInSe_2 (CIDS) is used for photovoltaic

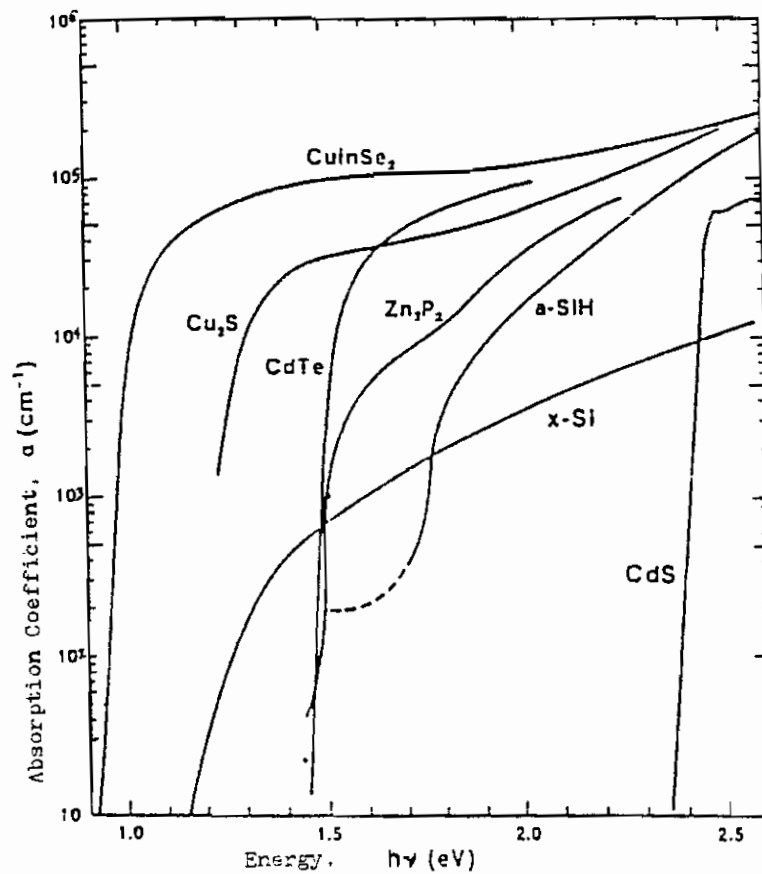


Figure (2). Optical absorption coefficients for some highly absorbing compound semiconductors. Also shown are the absorption coefficients for amorphous and crystalline silicon [4].

conversion both on its own and also in combination with larger bandgap semiconductors to make a more efficient use of the solar spectrum. Its bandgap is 1.04 eV at room temperature, significantly less than the optimum, approximately 1.4 eV [1], required to match to the solar spectrum.

To date, the material quality of these semiconductors remains inferior to that of Si and III - V semiconductors, but air mass one (AM1) efficiencies of about 12% have been achieved, both in photoelectrochemical (PEC) cells [4,36-40] and in heterojunction photovoltaic cells with CdS or $Cd_x Zn_{1-x} S$ as the window material [41], and recently 14.1% active area efficient test cells have been reported [42].

Polycrystalline $CuInSe_2$ thin films have been prepared for photovoltaic applications using various deposition techniques, including metalorganic chemical vapor deposition [43], flash evaporation [44], co-evaporation [45], sputtering [46-48], selenization of an electrodeposited CuIn layer [49], spray pyrolysis [50], and by a two - stage process utilizing e - beam evaporation [51]. Recently large grain polycrystalline $CuInSe_2$ ribbons have been grown for photovoltaic use [52], and 3900 cm^2 $CuInSe_2$ - based photovoltaic modules have been reported [42]. A detailed knowledge of the defect chemistry of this compound is necessary in order that $CuInSe_2$ can be efficiently used in economic terrestrial solar cells.

Other ternary compound semiconducting thin films reported were $Cd_{1-x} Mn_x Te$, grown by MOCVD [53,54], $PbSnTe$ and $PbSeTe$ grown by CSVT [55], $CuGaS_2$ grown by MOCVD [56], and $Zn_{0.99} Cd_{0.01} S$ grown by RF sputtering [57].

The band gap and the lattice constant of the compound semiconductor can be adjusted, within limits, by forming quaternary alloys. An example is the copper ternary system, resulting in five - element alloys like $Cu_x Ag_x In Se_{2-y} S_{2(1-y)}$, [58]. Figure (3) shows how such alloys can be formed among eight chalcopyrite semiconductors of the type I-III-VI₂. Points inside the cube correspond to six - element compounds. Iso - lattice constant, iso - energy gap, and iso - electron affinity maps can be constructed for such alloy systems. Figure (4) shows such a topological (E_g , a) map for the Cu-Ag-In-S-Se system [58]. These maps can be used to select

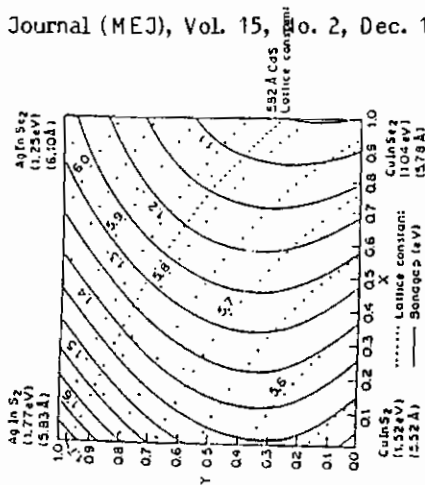


Figure (3). Sketch showing the ways alloys are formed between three or more ternary materials [58].

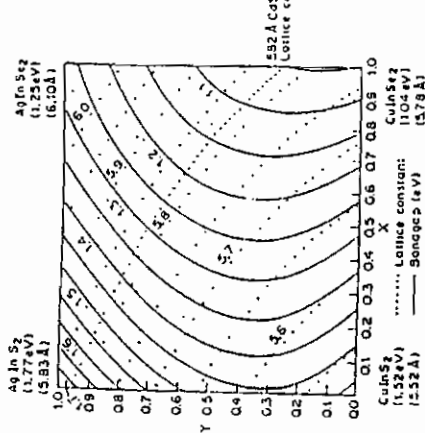


Figure (4). Empirical topological band gap and lattice constant map [56].

semiconductors of different band gap but the same lattice constant, for use in tandem solar cells.

iv) Cadmium Sulphide :

Polycrystalline cadmium sulphide, CdS, films have been extensively studied for photovoltaic applications in the last years [59,60].

CdS thin - film deposition techniques included vacuum evaporation [61-69], chemical vapor deposition [70], close - spaced techniques, CST, [71], sputtering [45,72-75], chemical spraying [76-80], cathodic electroplating [81], chemical bath deposition [82-85], sintering [86-88], and screen printing [10].

iv) Cuprous Sulphide :

Cu₂S / CdS heterojunction thin - film solar cells have been investigated, and is considered to be one of the best candidates for solar energy conversion. However, the disadvantages of lower photovoltaic efficiencies and instability limit their terrestrial applications.

Various deposition techniques have been reported, and research attempted to overcome these disadvantages. The deposition methods include vacuum deposition [89], RF sputtering [90], electrochemical and chemical methods [91], and organometallic chemical vapor deposition [92]. An extensive review of CdS/Cu₂S solar cells has been given in [93].

IV. Post - deposition heat treatment:

Heat treatment in different atmospheres (H₂, N₂, Ar,) is often carried out in order to improve and optimize the electronic and optical properties of the films deposited by various methods.

The effect of heat treatment in air and in different atmospheres on CdS/CdTe solar cells prepared by close - spaced vapor transport (CSVST) and hot wall vacuum evaporation (HWVE) has been investigated [94], and effective

grain boundary passivation in polycrystalline CdTe films by heat treatment in molecular hydrogen, a hydrogen plasma, and Li had been reported [95]. Oxygen has been found to have drastic effects on the performance of thin-film CdS/CdTe solar cells prepared by close-spaced sublimation (CSS), by enhancing the p-type characteristics of CdTe and thus enabling the fabrication of more efficient heterojunctions [96]. Annealing in N_2 and H_2 of undoped CdS thin films produced by RF sputtering was found to improve the electrical and optical properties [97], and the diffusion of copper from the $Cu_{2-x}S$ into the CdS layer due to heat treatment was analyzed in [98].

V. Photovoltaic materials' characterization:

The characterization of thin films used in photovoltaic applications includes studying the compositional and chemical, electrical, structural, and optical properties of the film.

Scanning electron microscopy (SEM) is used to study the morphology of the film, and X-ray diffraction to study its crystallinity. Electrical resistivity and Hall mobility (using the van der Pauw technique) are measured for the film at different temperatures. The optical absorption coefficient and the refractive index of the film are measured as a function of energy. Various chemical analysis techniques are used to characterize photovoltaic thin films, such as electron microprobe, Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS), and secondary ion mass spectroscopy (SIMS) [99].

VI. Photovoltaic devices:

Photovoltaic devices based on these materials have been reported. Important requirements on the PV devices are: i) Low cost of production, ii) Reasonable efficiency, iii) Stability, and iv) Suitability for large-scale manufacture.

Different photovoltaic structures have been reported, including homojunctions, heterojunctions, metal-semiconductor (Schottky) junctions, and metal-oxide-

semiconductor junctions (e. g. Indium tin oxide (ITO)/CdTe, SnO_2/CdTe , ZnO/CdTe). Lattice mismatch and the proper selection of bandgaps are important considerations in heterostructures. Table (1) lists the bandgaps, lattice constants, and electron affinity of the II - VI and ternary compound semiconductors [100].

Heterojunctions play an important role in photovoltaic devices because of their versatility as compared to homojunctions. Many of these heterojunctions are based on II - VI semiconductors or closely related I-III-VI₂ semiconductors. The most promising of these devices have had CdS as one of the semiconductors in the junction, mainly because of the ease with which high quality thin films of CdS can be produced by a wide variety of methods. The lattice mismatch between CdS and CdTe is about 9%, while that between CdS and CuInSe₂ is about 1%.

Table (2) summarizes the solar cell characteristics for CdTe - based heterostructures reported in the literature, while table (3) is for CuInSe₂ and table (4) is for Cu₂S devices. Important characteristics shown in the tables are the photovoltaic conversion efficiency, solar cell structure, cell area, open circuit voltage, short circuit current density, and fill factor.

Table (1) Bandgap, Lattice constant, and Electron affinity of II - VI and ternary semiconductors [100].

Material	Bandgap (eV)	Lattice constant (Å)	Electron affinity (eV)
II - VI :			
ZnS	3.6	5.41	3.9
ZnSe	2.6	5.67	4.09
ZnTe	2.3	6.10	3.5
CdS	2.4	5.83	4.5
CdSe	1.8	6.06	4.95
CdTe	1.5	6.47	4.28
HgS	2.0	5.86	--
HgSe	0.6	6.07	--
HgTe	-0.02	6.43	--
Ternary Compounds :			
CuInSe ₂	1.04	5.78	--
CuInS ₂	1.52	5.52	--
AgInSe ₂	1.25	6.10	--
AgInS ₂	1.77	1.77	--
CuGaSe ₂	1.68		--
Cu ₂ O	2.0		--

Table (2) Summary of CdTe thin-film solar cells

Structure	Deposition method	Active area (cm ²)	Efficiency (%)	V _{oc} ¹ (V)	J _{sc} ² (mA/cm ²)	F.F. ³ (%)	Ref.
CdS/CdTe	Screen print	0.78	12.8	0.754	27.9	61	101
CdS/CdTe	CSVT*	0.1	10.5	0.75	22.7	62	102
CdS/(CdHg)Te	Electroplating	1.48	10.6	0.62	27.03	63	103
CdS/CdTe	CSVT	1.2	10.6	0.748	22.2	63.4	8
CdS/CdTe/ZnTe	Spray-electro-plate-evaporate	4.0	10.4	0.745	22.0	63.6	104
SnO ₂ /CdTe	CSVT	4.0	10.5	0.663	28.1	56	105
CdS/CdTe	Evaporation	0.09	8.7	0.612	20.5	60.5	106
CdTe/CdS	CVD**	1.0	6.5	0.67	16.4	59	107
CdTe/ITO***	CVD	1.0	8.2	0.72	19	60	"
CdTe/CdS		0.14	6.9	0.52	17.3	77	108
ITO/CdS/CdTe	CSS*		10.5***	0.75	17	62	102
CdS/CdTe	Screen printing-sintering	0.78	12.8	0.75	22	61	109
CdS/CdTe	Screen printing	500	8.5*	0.275	34.7	50.6	110
CdS/CdTe	CSVT	0.8	4.5	0.65	13	44	111
CdS/CdTe	Sintering	0.3	12.5				"
CdS/CdTe	Pyrolytic spray	small	8.7	0.747	17.3	67	18
CdS/InP*	CVD	0.55	13.5***	0.78	17.1	75	70
Au/CdTe	Electro-	1.0	6.2	0.58	17.3	62	11
" "	deposition	--	8.6	0.723	18.7	64	"
ITO/CdS/CdTe	Cathodic	0.02	8.0	<0.62	<21		112
" "		4.2	6.5-6.8				"
ITO/CdS/CdTe	Spray pyrolysis	0.024	4	0.635	12.9	49	16
" "	" "	1.1	3	0.607	10.9	49	16
Cd _{1-x} Zn _x Te	Electrodepo-	0.02	3.8*	0.55	15	45	113
x = 0.1	deposition/evaporation						
ITO/InSe [®] /Au	(ITO) Sputtering	---	>10	0.58	32	63	114

¹ Open-circuit voltage
² Short-circuit current density
³ Fill factor
* Closed-space vapor transport
** Chemical vapor deposition
*** Indium tin oxide
close-spaced sublimation
AM2 efficiency
\$ Single crystal
+ AM1.5 efficiency (under 100 mW/cm² illumination)

Table (3) Summary of CuInSe₂ thin-film solar cells

Structure	Deposition method	Reported efficiency (%)	Cell area (cm ²)	V _{o.c.} ¹ (V)	J _{s.c.} ² (mA/cm ²)	F.F. ³ (%)	Ref.
ZnO/CdS/CuInSe ₂ /Mo/glass		12.5					115
CdS/CuInSe ₂ /Mo/alumina	Coevaporation	12					116
ITO/CdS/CuInSe ₂ /Mo/glass	Vacuum evaporation	3-4*	0.09	0.22	25-30	46-50	117
ITO/CdS/CuInSe ₂ /Al ₂ O ₃	"	"	"	"	"	"	"
ZnO/CdS/CuInSe ₂ /Mo/glass	Coevaporation	14.1*	3.8	0.508	41	67.7	118
SiO ₂ /CdZnS/CuInSe ₂ /Mo/alumina(glass)	Vacuum evaporation	11	1.0	0.436	38.6	65.3	119
ZnO/CdZnS/CuInGaSe ₂ /Mo/alumina	Coevaporation	12.9	1.0	0.555	34.2	65.7	120
SiO ₂ /CdS/CuInSe ₂ /Mo/alumina	Coevaporation	9.53*	1.0	0.396	39	63	121
CuInSe ₂ /ZnSe	Sputtering	7-8.5	1.5-2.0	0.43		32.2	122
CuInSe ₂ /CdS	"	10-11.5					"
CuGaSe ₂ /(Zn,Cd)S	Evaporation	5.8		0.845	11.6	50	123
Cu(Ga _x In _{1-x})Se ₂ /(Zn,Cd)S	"						"
ZnCdS/CuIn _{1-x} Ga _x Se ₂	Evaporation	10.2	1.074	0.509	30.41	66	124
ZnO/CdS/CuInSe ₂ /Mo/glass	Electro-deposition (Cu,In)/H ₂ Se	7					125
CdS/CuGa _x In _{1-x} Se ₂ /Mo/glass	Evaporation	10					"

¹ Open-circuit voltage

² Short-circuit current density

³ Fill factor

* Under 87.5 mW/cm² intensity

+ AM1.5 (under 100 mW/cm² intensity).

Table (4) Summary of Cu_xS thin-film solar cells

Structure	Deposition method	Efficiency (%)	Area (cm ²)	V _{o.c.} ¹ (V)	J _{s.c.} ² (mA/cm ²)	F.F. ³ (%)	Ref.
Cu _x S/CDS	Sputtering	3.6	--	--	--	--	90
Cd _{1-y} Zn _y S/Cu _x S	Electrochemical	3.0 - 6.5	--	0.41-0.45	17-22	48-70	91
Cu _x S/CDS	MOCVD	--	--	--	--	--	92
	Evaporation/chemical	9.15	1.0	0.516	21.8	71.4	126

¹ Open-circuit voltage

² Short-circuit current density

³ Fill factor

VII. Conclusions:

Efficient utilization of the sun's energy in Egypt will be encouraged by the development of low - cost thin - film polycrystalline solar cells.

II - VI compound semiconductors - based thin-film solar cells have progressed considerably during the recent years, for the potential application in terrestrial photovoltaic energy conversion.

A good deal of materials research is still required in order to control and optimize the electronic and optical properties of II - VI based thin films and to understand the drastic effect of heat treatment on these properties.

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