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Study and Preparation of Optoelectronic Properties of $AgAl_{1-x}In_xSe_2$ /Si Heterojunction Solar Cell Applications

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Abstract. In this research, $AgAl_{1-x}In_xSe_2$ (AAIS) compound alloys prepared for different x (0.3, 0.6 and 0.9) by melting them in an evacuated quartz tube (2.5*10-3 torr). AAIS with different Indium concentrations x (0.3, 0.6 and 0.9), t=750±30nm thin films deposited by thermal evaporation method with deposition rate (5±0.1) nm sec-1 on glass and p-type silicon substrates to study structural, optical, electrical properties and to fabricate solar cells. From energy dispersive x-ray spectrometer (EDS) found the amount or (concentration) of the elements (Ag, Al, In, Se) of alloys, to studying the topography, estimate the surface roughness and average grain size was used AFM technique, the results show that the grain size increase with In-content increase. Optical measurements show that the preparation AAIS thin films have high absorption in visible range. The allowed direct energy gap was decreases with increase In-content. All films were N-type while the concentration of the charge carriers increases. The optimum condition of efficiency (η =5.32). Under illumination found where (x=0.6, t=750±30 nm)

Keywords: n- $AgAl_{1-x}In_xSe_2$, energy gap, photovoltaic cells.

INTRODUCTION

This research focus on chalcopyrite compound for the development of thin films solar cells, which are both cheap and highly efficient. Ternary chalcopyrite semiconductor has received much interest as the absorbing layer in the polycrystalline thin films solar cells [1]. The band gap of AgAlSe2 is comparatively high, 2.5 eV, but it can be used to better coincide the solar spectrum either by replace part of Al by Ga, In or part of Se by Te, S. The flexibility of the material allows in principle the band gap variation from 1.00 eV of AgInTe2 to 3.13 eV of AgAlS2 [2, 3]. The first researcher start to work in Ag chalcopyrite was Keiichirou Yamada et al. (2006) have investigated Ag (In,Ga)Se2 thin films deposited on glass substrate by using a molecular beam epitaxy [4]. AgInSe2 prepared by annealing conventional thermal vacuum evaporation [5]. To increase in solar cells efficiency made the thin films (AgCu)(In,Ga)Se2 by films elemental co-evaporation[6]. The thin film AgAlS2 deposited by by thermal spray pyrolysis [7]. AgAlS2 thin films prepared by chemical spray pyrolysis [8]. AgInSe2 crystals were grown by Bridgman technique [9]. AgInSe2 thin films prepared by electrodeposition processdil[10]. Cu–Ag–In–Se thin film deposited by thermal evaporation technique [11]. The chalcopyrite compound Ag (Al, In) Se2 have been used as thin film absorber layers for solar cells in this work.

EXPERIMENTAL

The Alloy Preparation

$AgAl_{1-x}In_xSe_2$ alloys were prepared for three values of (x=0. 3, 0.6, 0.9). The exact amount of elements (Sliver, Aluminum, Indium, Selenium) according to atomic percentages weighed using an electronic balance (10^{-4} g). The

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materials were then sealed in evacuating (10^{-3}Torr) quartz ampule 25 cm length. The ampules containing material are heated to 1200 K for 5 hours, the sample was left to cool slowly in the oven, and the tubes broke them to extract samples and then grinding samples by laboratory glass mortar. (EDS) was using to analyzed the elemental composition.

$AgAl_{1-x}In_xSe_2$ Thin Films Preparation

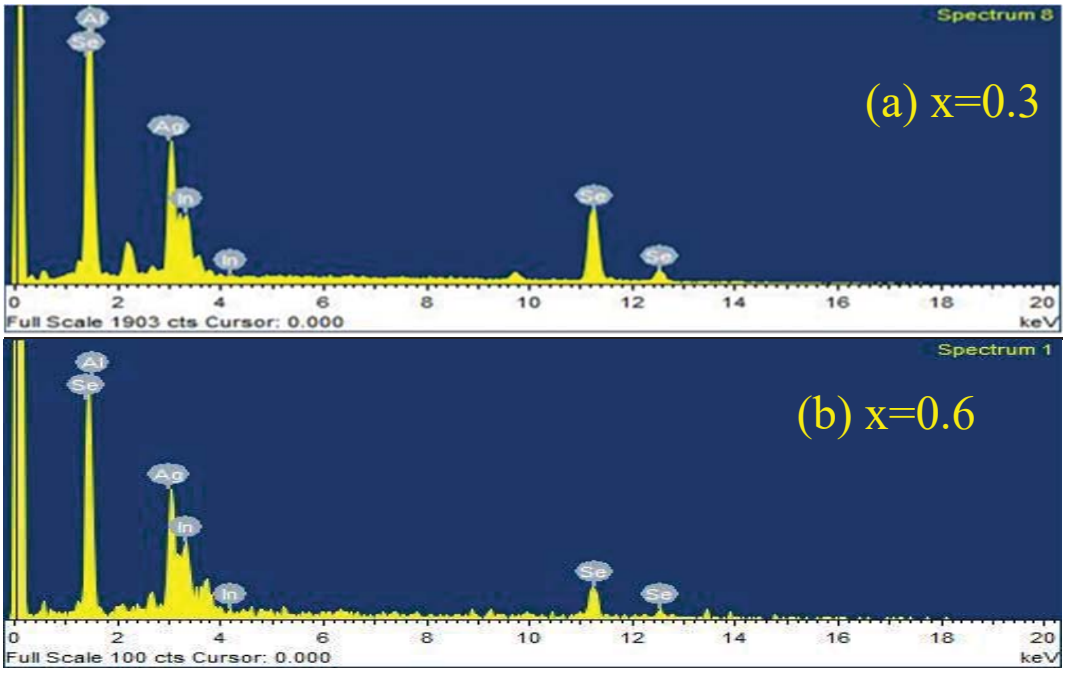
Vacuum thermal evaporation method used to deposition of AAIS thin film with Indium concentrations x (0.3, 0.6 and 0.9). The optical interferometer method was using to determine the thickness of AAIS.

Characterization

The deposition at R.T on glass and p-type silicon substrates, glass substrate to investigation the structural. Structure of the (AAIS) thin films examined by X-ray diffraction. The scanning angle 2θ was vary in the range of 10°-60° in order to know the crystal structure using X-ray diffraction system, the average crystalline size (C.S) calculated by using the Scherrer's Formula with (λ = 1.5418 Å) [12]. The (AFM) technology used to study the effect of mixing ratios on the surface topography, grain size, surface roughness for prepared thin films, as the use device type of (SPM-AAA3000 contact mode spectrometer, Angstrom [13]. Optical parameters were measured, which include absorbance (A) and transmittance (T) using a spectrometer type (UV-Visible 1800 spectra photometer), the calculation of the absorption coefficient by lambert law[14], the optical energy gap determined using Tauc equation[15]. Hall Effect measurements have been used in determining concentration, type of majority carrier and mobility in thin film materials by Van der Pauw, the (AAIS) thin films deposited on silicon substrate to making AAIS /Si, Shockley equation used to calculate I-V characteristics[16,17].

RESULTS AND DISCUSSION

The amount or (concentration) of the elements (**Ag, Al, In, Se)** in the alloys is examined by (EDS) technique depending on the standard of these elements. The results are shown in FIGURE (1) and TABLE (1).



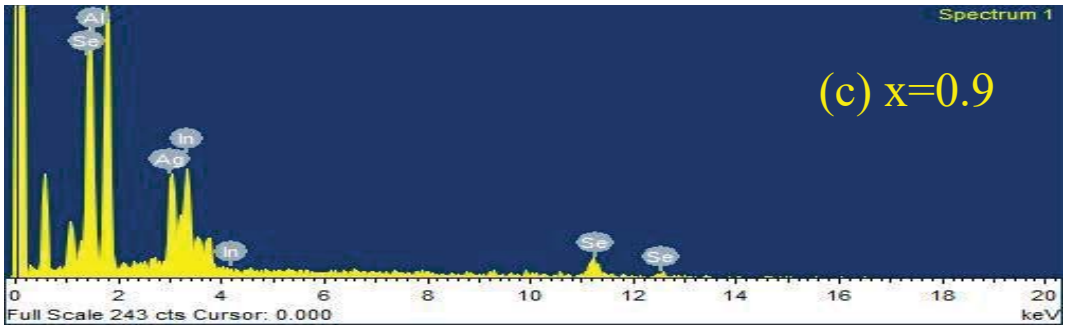


FIGURE 1. EDS patterns for $AgAl1_{-x}$ In_xSe_2 alloys with different In (x=0.3, 0.6, 0.9) (a, b, c).

TABLE 1. The composition of $AgAl1_{-x}$ In_xSe_2 alloys determined by (EDS)

alloys	Calculated (Wt%)				Test (Wt%)			
	Ag	Al	In	Se	Ag	Al	In	Se
$AgAl_{0.7}$ $In_{0.3}Se_2$	33.8	5.9	10.8	49.5	33.79	5.9	10.79	49.49
$AgAl_{0.4}$ $In_{0.6}Se_2$	31.23	3.12	19.94	45.71	31.23	3.12	19.93	45.70
$AgAl_{0.1}In_{0.9}Se_2$	29.01	0.73	27.79	42.47	29.0	0.73	27.78	42.46

FIGURE (2) displays the XRD for $AgAl_{1-x}$ In_xSe_2 thin film that deposited on glass substrates with different In x (0.3, 0.6 and 0.9). All films have two main crystalline peaks located at $2\theta \approx 26.6^o$ when preferred orientation (112), and peak appeared at $2\theta \approx 42.77^o$ with the (220). TABLE 2 display all the observed peaks in all films, this result show that the film have tetragonal crystalline structure and the degree of crystalline increasing when the In increase [18, 19, 20]. By increasing the In concentrations, the location of the measured diffraction peak do not difference significantly but the intensity of the peaks increases. That is mean development of crystalline of the thin film being and crystallite size C_S become greater as in TABLE 2, [19].

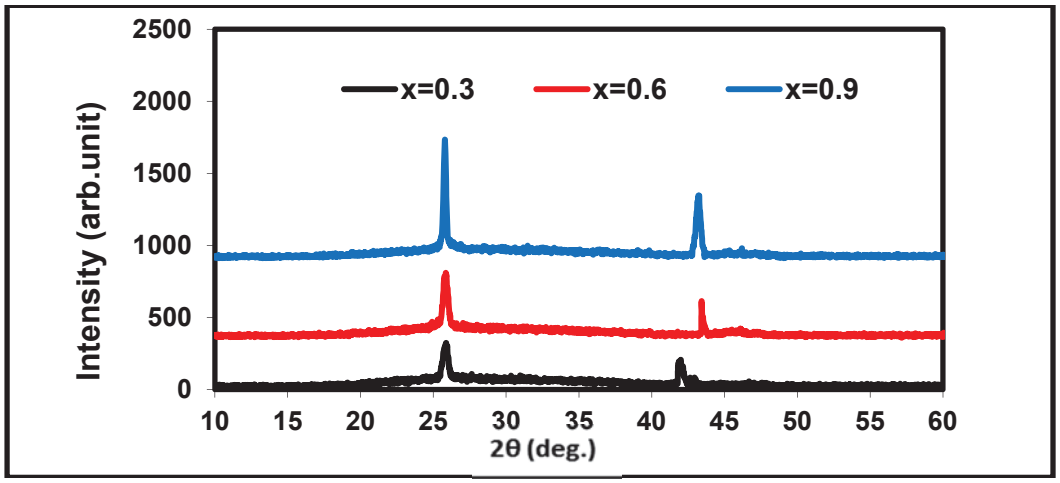


FIGURE 2. XRD for $AgAl_{1-x}$ In_xSe_2 films deposited on glass with different x.

TABLE 2. Structural parameters of ($AgAl_{1-x}In_xSe_2$) thin films.

X	2θ(Degrees)	d(Exp.)(Å)	(hkl)	FWHM (Degrees)	C_S(nm)
0.3	25.88	3.438	(112)	0.456	18.67
	42.0	2.148	(220)		
0.6	25.86	3.441	(112)	0.407	20.917
	43.42	2.081	(220)		
0.9	25.8	3.449	(112)	0.205	41.524
	43.2	2.091	(220)		

In order to study the surface topography, roughness of thin film and effects of film concentration (x) used the AFM, to obtain very precise statistical values about the grain size and surface roughnes depending on the root mean square (r.m.s). FIGURE (3) appear AFM images in three dimensional for films $AgAl_{1-x}In_xSe_2$ with different values of In (x = 0.3, 0.6, 0.9), t = 750 nm. The study of the material films, surfaces is very important to know how the distribution and arrangement of atoms on surface and to identify the homogeneity characteristics of structures [21].

TABLE (3) the surface roughness and grain size vary with the values of In contain that effect on the properties of films surface and optical properties of the material, the highest value of roughness when the ratio 0.6 that an agreement with [22]. This value mean possibility to use AAIS thin films in fabricated solar cell or use it as a cover antireflection coating as it reduces light reflection and increases film absorption for energy photons in visible region, this variation have an effect on the electrical and optical characteristic of the materials and in the appropriate application selection in the electronic devices industry [23].

TABLE 3 . The grain size, roughness average and r.m.s for $AgAl_{1-x}In_xSe_2$ at different In (x=0.3, 0.6, 0.9).

x	Grain size (nm)	Roughness average (nm)	r.m.s (nm)
0.3	3.22	2.78	99.14
0.6	5.77	4.82	106.42
0.9	3.77	3.2	109.68

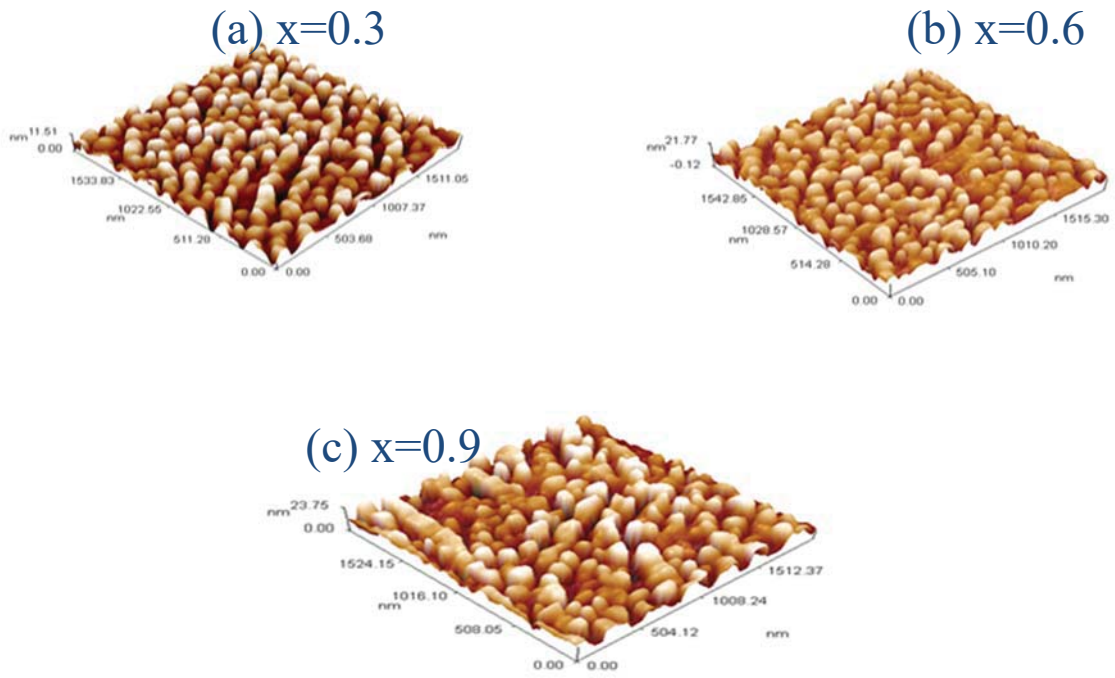


FIGURE 3. AFM image of thin $AgAl_{1-x}In_xSe_2$ film at different In (x=0.3, 0.6, 0.9).

The absorption coefficient value calculated by using the lambert law depending on the absorption spectrum. FIGURE (4) represent change in absorption coefficient (α) as a function of photon energy (*hv*). At the high values α greater than $10^4 cm^{-1}$ it is pointed that the bandgap is direct transition and this is agree with [2,3]. The value of (α) of the thin films is increasing in visible spectrum region, when (x = 0.6 , t=750nm) the α has highest value, while when x = 0.9 of the prepared film the absorption is decreasing. FIGURE (5) shows the band gap values decrease with In for $AgAl_{1-x}In_xSe_2$ thin films. This is due to adding In to $AgAlSe_2$ made material more opaque . The decrease of the band gap in agreement with study [24].

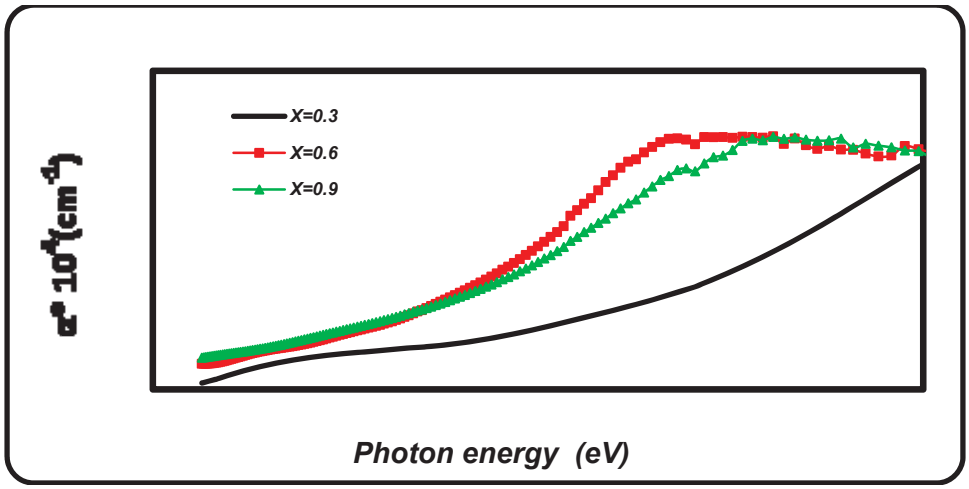


FIGURE 4. Variation of absorption coefficient α respect to photon energy for $AgAl_{1-x}In_xSe_2$ at different x=0.3,0.6,0.9.

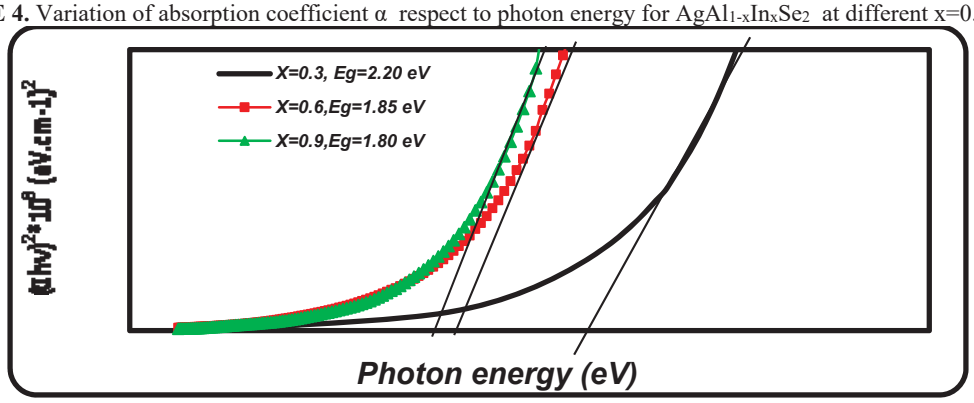


FIGURE 5. Variation of $(\alpha h\nu)^2$ respect to photon energy for $AgAl_{1-x}In_xSe_2$ at different x=0.3,0.6,0.9.

The type of carriers (n), Hall coefficient $\mathbf{R_{(H)}}$, conductivity and Hall mobility (μ_H) have been estimated from Hall measurements. The negative sign of $\mathbf{R_{(H)}}$ indicates that the conductivity type is n-type [25]. Which means that the type of conduction was n-type, i.e. electrons are majority charge carriers in the conduction process. TABLE (4) show that when the increasing the Indium element ratio x for (AAIS) films lead to a decrease in the value of the Hall coefficient (RH), increase in the carrier concentration (n) and mobility (μH) reach maximum value at x=0.6 for t=750nm and then decrease. This result is related to the structure which mean that the structure reach the most stable state. This may decrease trapping centers which increases number of charge carriers and mobility due to band gap decreasing. The **n (cm^{-3})** of the order $10^{17}cm^{-3}$ is very good agreement with refs [26, 27].

TABLE 4. Hall parameters for $AgAl_{1-x}In_xSe_2$ at different x=0.3, 0.6, 0.9.

x	$\sigma(\Omega.cm)^{-1}$	$R_{(H)}$	$\mu_H(cm^2/V.S)$	n (cm^{-3})
0.3	54.32	-13.888	7.54E+02	4.50E+17
0.6	120.9	-8.0128	9.61E+02	7.80E+17
0.9	90	-12.019	1.08E+03	5.20E+17

The study of the (current - voltage) characteristics is important parameter of junction measurement to describe the performance of the heterojunction solar cell, which explains the behavior of the resultant current with the applied forward and reverse bias voltages [28]. The cell characteristics in the light are the same in the dark with a shifted by the amount of the photocurrent. In order to determine the performance of a solar cell device. The measurements was with incident power density 100 mW/cm^2. It could be seen that the (Jph) increases with increasing x to 0.6 and then decrease because the energy levels within the energy gap act as active recombination centers when increasing x more than 0.6. Also the decrease in short circuit current density may be attributed to higher series resistance increasing

when x >0.6. The ratio (x = 0.6, t=750nm) is the best among the others and this is consistent with the researcher [29], The Rs has small values compared with the Rsh values for all the prepared samples, the lowest value for Rs, also the highest value for Rsh when the thickness (750nm) and the ratio (x = 0.6).

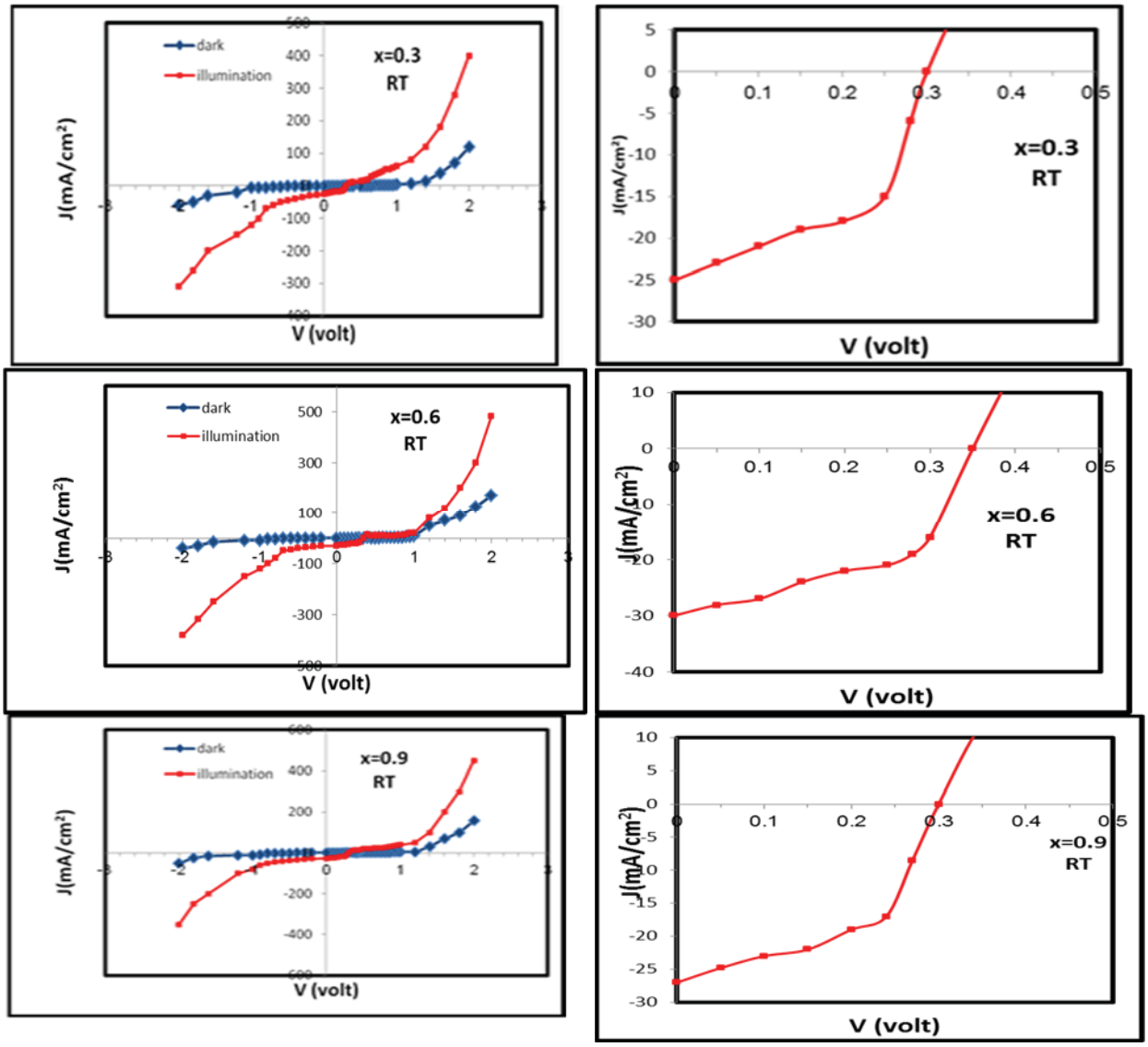


FIGURE 6. I-V characteristics under dark and illumination for $AgAl_{1-x}In_xSe_2$ /Si Solar Cell at different x(0.3,0.6,0.9).

TABLE 5. the parameter for $AgAl_{1-x}In_xSe_2$/Si heterojunction solar cell at different x(0.3,0.6,0.9).

X	J_{sc}(mA/cm^2)	V_{oc}(volt)	J_m(mA/cm^2)	V_m(volt)	$R_{sh(\Omega)}$	$R_{s\ (\Omega)}$	F.F.	η%
0.3	25	0.3	15	0.25	25.0333	3.33	0.5	3.75
0.6	30	0.35	19	0.28	26.3	3.12	0.506667	5.32
0.9	27	0.3	17	0.24	22.739	4.2	0.503704	4.08

CONCLUSIONS

X-ray diffraction results show that the structure of $AgAl_{1-x}In_xSe_2$ with Indium element ratios have polycrystalline Tetragonal structure in preferential orientation (112). The roughness and grain size increases with increasing In content. The direct transitions of $AgAl_{1-x}In_xSe_2$ and energy gap decreases with increasing of Indium content. Hall

measurement showed that all the films are n-type. Solar cell manufacturing using n-AAIS/p-Si heterojunction by vacuum evaporation technology at room temperature. The maximum value of efficiency (5.32) when x=0.6.

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