



Improving properties and performance solar cell of nanostructured tin sulfide by aluminum doping

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Aluminum dopant Tin Sulfide than (p-SnS /n-Si and p-SnS: Al / n-Si) thin films are deposited with thickness (500 ± 20) nm onto glass and Si substrates by thermal evaporation procedure under a vacuum of 5×10^{-6} mbar with deposition rate 1nm/sec. The effect of different Al ratios (1%,2% and 3%) on the nanostructure SnS thin film and (p-SnS /n-Si) hetero junction characteristics are examined. The structural films properties examined through X-ray diffraction showed that SnS film are poly crystallin with increase crystallite size from (21.04 to 27.7) nm with increasing Al ratio. The optical properties demonstration excellent absorbance for film doped with 3% Al beside reduced energy gaps from 1.6 toward 1.44 eV. Hall effects measurement indicated wholly film equipped pure besides doping had p-type conductive, Further, the lower resistivity of 4.77×10^2 ($\Omega \cdot \text{cm}$) besides maximum carrier's concentrate of 2.81×10^{16} (cm^{-3}) for doped 3% (Al) films. Below illumination the (I-V) characteristics of hetero junction via occurrence 100 mW/cm² powers density support presented that heterojunction (p-SnS: 3%Al / n-Si) has maximum efficiency ($\eta = 5.512$ %). which is enhanced by the optimized nanostructure and uniform nanoparticle distribution.

Keywords: Al doped Tin Sulfide; Thin films; Thermal evaporation; Structural; Optical.

1. INTRODUCTION

Nowadays, friendly technologies environmentally and increase energy efficiency led to discoveries of new material [1,2]. Thin film binary compound materials based solar cells attract interest with their composed of earth- abundant elements and lower cost [2]. Tin mono-sulfide (SnS) is one among the binary compounds that be a member of IV - VI group. SnS is considered a promising material for optoelectronic applications owing to its exceptional properties, for example inert both chemically and environmentally, friendly source materials non-toxic, have metastable cubic (CUB) or a stable orthorhombic(ORT) crystalline structure, shows strong anisotropic vibrational properties due to the layered structure, a suitable direct band gap (1.3 eV to 1.8 eV) with high absorption coefficient ($>10^4 \text{ cm}^{-1}$) making it ideal for photovoltaic application, own p-type electrical conductivity, long-term stability and cost-effectiveness [2-7]. Exterior doping is generally preferred to improve the physical properties of SnS, current studies have designated doping SnS films with elements such as cadmium (Cd) [8], neodymium (Nd) [9], selenium (Se) [10], bismuth (Bi) [11], iron (Fe) [12], copper (Cu) [13], indium (In) [14], nickel (Ni) [15], silver (Ag) [2,16,17] and Aluminum (Al) [18] making SnS more fit for photovoltaic devices additionally for optoelectronic applications. Different methods has been used to deposit SnS films either pure or doped such as thermal evaporation [10,11,16], sol-gel technique [8], chemical bath deposition technique [15], atomic layer deposition (ALD) [19,20], chemical vapor deposition (CVD)[21], RF sputtering [22], spray pyrolysis technique [9,23], electrochemical deposition[14, 24] and spin coating [25,26]. In current times, engineering orientation has confirmed to have high potential to get great power conversion efficiency in antimony chalcogenide with a low-dimensional crystal-structural (LDCS) like Sb_2S_3 and Sb_2Se_3 -based solar cells [5]. Growing in the direction of this research to further (LDCS) absorber materials especially SnS, thermal evaporation technique is used to fabricate the initial SnS-based solar cells, displaying a extremely low efficiency of 0.29% [5]. In this study, SnS thin films produced pure and doped with Al by thermal evaporation from alloy source on glass and Si substrate with 500 nm in thickness, varied dopant ratio in steps of 1 wt.% from 0 to 3 wt.%. The aim of this work is to explain how a dopant ratio of Al improves the nano structural, optical and electrical films properties and heterojunction characteristics with enhancing solar cell efficiency. The novelty of the present work where prepares the alloy and then uses it as an evaporation source to obtain homogeneous films, study the effect of aluminum doping on structural, optical and Hall Effect results for thin films prepared with solar cell performance in a single study. Control the optimal aluminum doping ratio that achieves the best energy conversion efficiency.

2. EXPERIMENTAL

Films of pure Tin Sulfide (SnS), Aluminum (Al) doped SnS with (1, 2, 3) % percentages and p-SnS: Al/ n-Si hetero junction have been deposited on glass and Silicon single crystal (111) substrate by thermal evaporation process under 5×10^{-6} mbar vacuum per thickness (500 ± 50) nm thru deposition rate 1nm/sec after preparation SnS alloy. Tin Sulfide alloy is prepared by put sulfide (S) and tin (Sn) elements with (99.999%) purity and (1:1) weight percentages in quartz tube under (4×10^{-4}) mbar pressure then placed them in an electric oven at temperature 1000 K for 5 hours, finally left them cool slowly. To get films by thermal evaporation using Edward coating unit model (E 306) put the mixture obtained from alloy in molybdenum boat, 18 cm is the distance from the boat to substrate, then doped SnS films with Al (1, 2, 3) % percentages. X-ray diffraction (XRD) technique is used to study the crystal structure for thin films SnS pure and SnS: Al by using (SHIMADZU Japan XRD 600) with

CuK_α where (λ = 1.5418 Å) , 20 mA currents and 40Kv, thru greatest the values of 2Θ from 20° to 80°. Bragg law is used to determine the interplanar spacing (d) for miller index (hkl) [27,28]

$$2d\sin\theta=n\lambda \quad (1)$$

where θ is Bragg's angle and λ is X-ray diffraction wavelength. To determine for all films the crystalline size (C.S) by Scherer's Formula [29,30]

$$C.S = \frac{0.94 \times \lambda_{(x-ray)}}{\beta_{(FWHM)} \cos \theta} \quad (2)$$

Somewhere β: FWHM of the peaks. Dislocations density (δ) values calculate by equation [31,32]

$$\delta = \frac{1}{(C.S)^2} \quad (3)$$

Although (ε) micro strain can cautious since equality [32]:

$$\epsilon = \frac{\beta_{(FWHM)} \times \cos \theta}{4} \quad (4)$$

The absorbance (A) and transmittance (T) values are recorded in the range of (400 – 1000) nm to evaluate the optical property for all films using UV-vis spectrophotometer, the absorptions coefficient (α) is intended since equivalence [33,34].

$$\alpha = (2.303A/t) \quad (5)$$

Wherever film thickens is t. Tauc's equation is used to estimate the energy gap (E_g) [34,35].

$$(\alpha h\nu) = B (h\nu - E_g)^n \quad (6)$$

where n is transition type number, hν is incident photon energy and B is constant. Van der Pauw Ecopia-HMS -3000 is used to evaluate the electrical properties (Hall measurement) for all films prepared by record resistivity (ρ) and Hall voltage (V_H) values. Hall coefficient R_H, types of major carriers, Carrier concentrations (n_H) and Hall mobility (μ_H) are investigated [36].

I-V Current voltage characteristics are analyzed under dark and illuminated conditions to estimate the opto-electrical properties of p-SnS /n- Si and p-SnS: Al /n-Si heterojunction using halogen lamp (100 mW/cm²) by employed Shockley equation [37]. Additionally, the solar cells efficiency is determined [38]. Below a schematic diagram of the fabricated p-SnS:Al/n-Si heterojunction device.

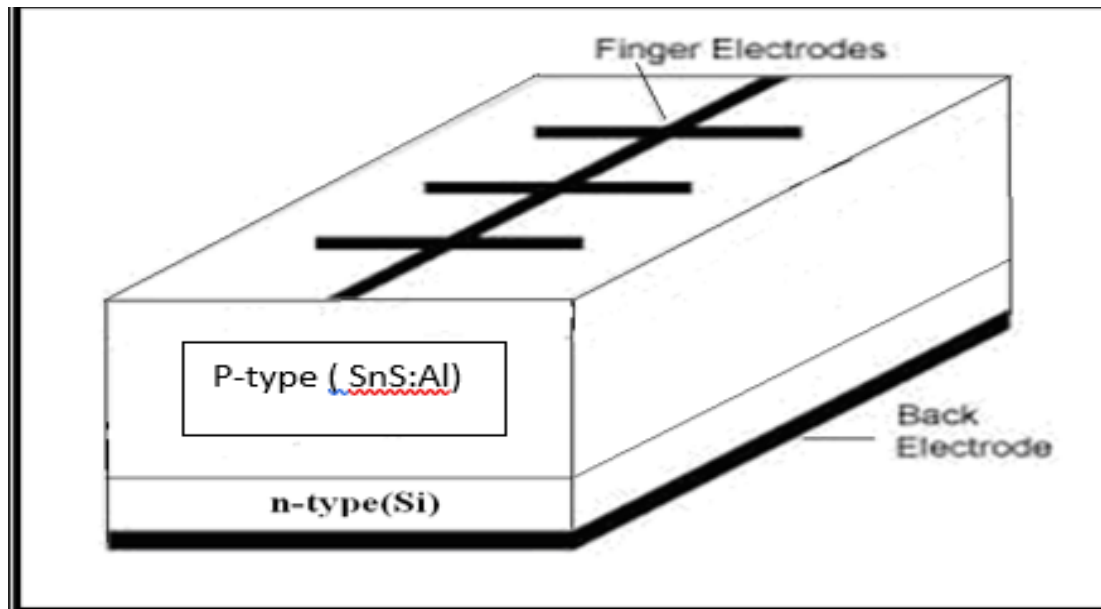


Figure 1 System design.

3. RESULTS AND DISCUSSION

3.1. Structural measurement

Figure 1 shows structural characterization recorded by X-ray diffraction for thin films SnS un doped and doped with Al through various Al percentages (1, 2, 3) % in the range of 2θ amongst 20° to 80° . It is clear from this figure that these films prepared by thermal evaporation per thickness (500 ± 50) nm have polycrystalline orthorhombic structure indicates sharp peaks with diffraction XRD patterns appear at 2θ (27.5° , 30.6° , 31.8° , and 45.6°) related to (201), (101), (111) and (411) with preferred orientation along (111) plane. Further diffraction peak meticulously agrees with standard XRD information (JCPDS cards 00-039-0354); no distinguishing phases of Al element appear it approves that Al ions have well united into Sn ion sites without varying the crystal structure [17] and these results are in good contract with [3,4,17,18]. The development along (111) plan for films (SnS:3%Al) might be due to substitution of Al ions in the Sn positions as well as the occupation of Al ions in the crystal vacancies [17]. Also observed in fig (1) in addition Table (1) here is slight shifts on location of major peak (111) toward the higher 2θ and become sharpest after doped with Al due to improved crystallinities because the lower ionic radius of Al ions, replaces the higher ionic radius of Sn ions. Table (1) and figure (2) show crystallite size, dislocations density and strain which calculated using FWHM values in Radian for (111) peak. It is clear that the crystallite size values are improved from 21.04 nm to 27.7 nm while value of dislocations density besides strain decreased thru increased the Al content because of enhanced crystal growth. These results are great matched per [3,18].

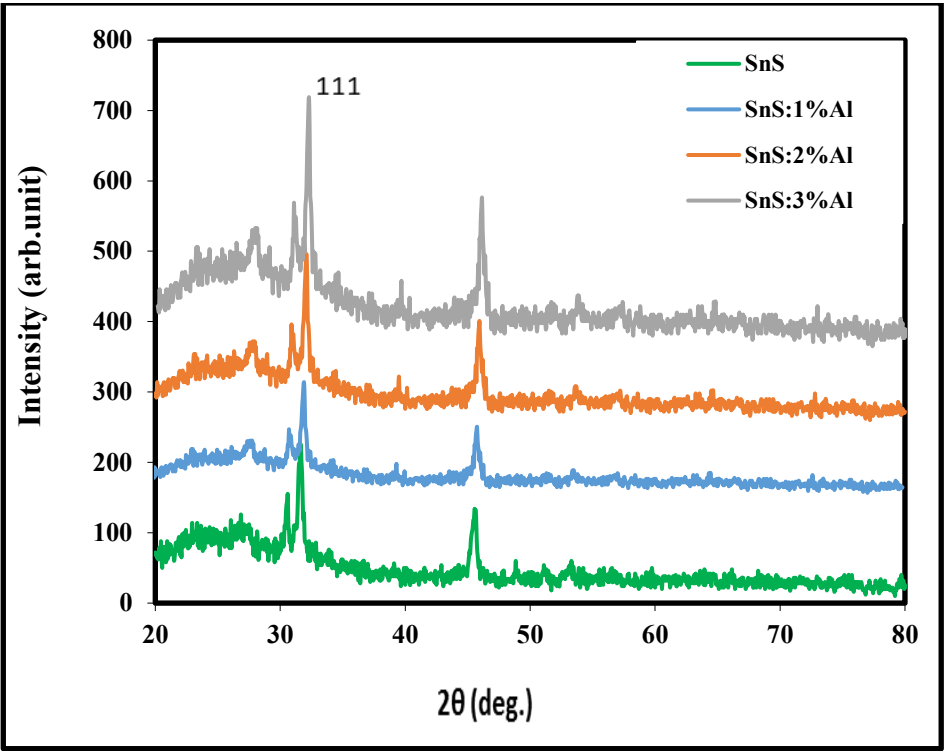


Figure 2 XRD pattern of SnS thin film pure and Al doped as a function of Al ratio.

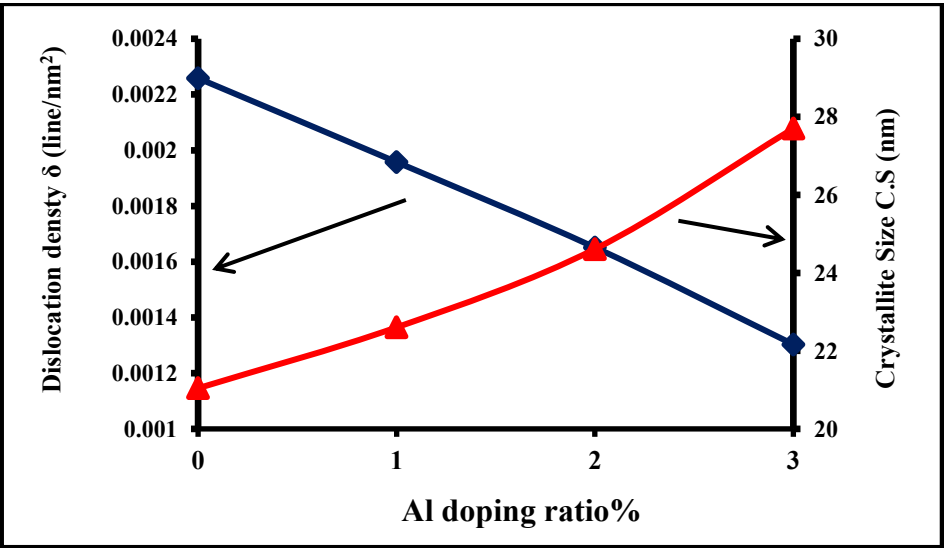


Figure 3 crystallite size then dislocations density aimed at pure besides Al dopant SnS in different doping ratio.

Table 1 Structure constraint for SnS thin films pure and diverse ratio Al dopant.

Sample	2θ	FWHM	C.S(nm)	δ	ε
SnS	31.6	0.41	21.04279	0.002258	0.00172
SnS:1%Al	31.9	0.382	22.60201	0.001958	0.001602
SnS:2% Al	32.1	0.351	24.61052	0.001651	0.001471
SnS:3% Al	32.3	0.312	27.70078	0.001303	0.001307

3.2 Morphological measurement

Figure 3 shows that the prepared films SnS and SnS:3%Al are free from micro-defects such as cracks or pin holes, and that the particles are of different shapes and sizes, and how the doping processes affected the particle size. It is clear that spherical nanoparticles distributed with great homogeneity and regularity over the surface area of the sample, and there are no surface cracks, gaps or voids, which indicates that the thermal evaporation method and doping ratios are suitable for preparation as well as the doping process has affected the size of the resulting particles.

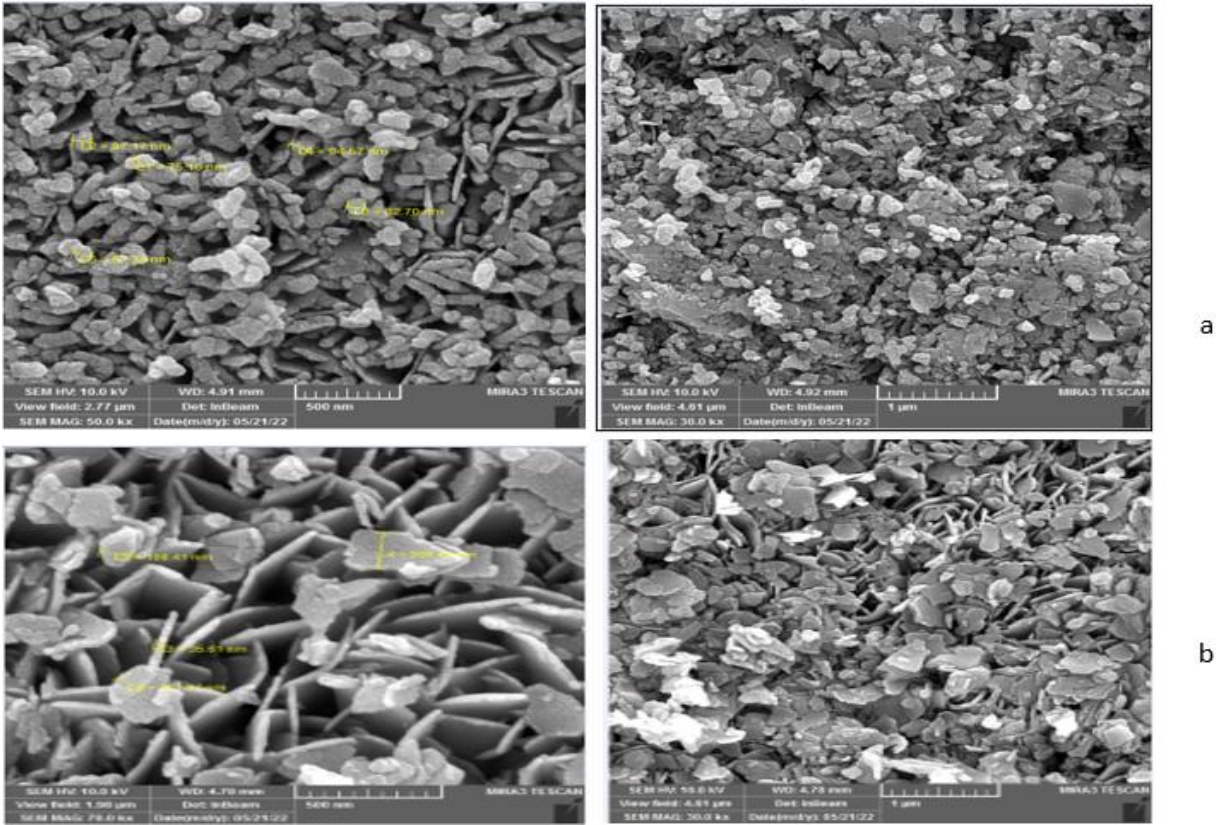


Figure 3 SEM image of SnS thin film a) pure and b) 3%Al doped.

3.3 Optical characterization

The optical characterization absorbance and transmittance spectra of manufactured thin films of SnS and SnS:Al via various ratio of Al using thermal evaporation are recorded in the wavelengths from 400 nm to 1000 nm showing in figure 4. This figure shows increased films absorbance values from (40% to 90%) in the range (400-700) nm though decreased transmittance values with rise concentration of Al (1, 2 and 3) % for all range. This behavior may be due to Al dopant level make as fine absorption position leads to increase absorbance values of these films connection with increase crystallite size values, the optical absorbance and transmittance of thin films are effects by many elements, such as crystallinity, film thickness, surface homogeneity, doping concentration, structural defects, and scattering grain boundary, jointly influence on the absorbance and transmittance level of the material [2].

Absorption coefficient calculated from absorbance according to equation (5) for all films prepared un doped and doped is presented in Figure 5 verses wavelength (400-1000) nm, this figure shows increase absorption coefficients value after doping with Al for all rang of wavelength as well as all allowed direct transition can be occurred because the absorption coefficients value for these films is high ($\alpha > 10^4 \text{ cm}^{-1}$) for similar reasons we discussion before.

Furthermore, this figure demonstration reduction optical energy gap values which calculated from Tauc equation from (1.6 to 1.44) eV as in Table 2 with increase Al ratio (0, 1, 2 and 3) % this result is as good as with studied [2, 3] furthermore films doped with Al are good absorber layer for solar cell. Figure (6) show the variation $(\alpha h\nu)^2$ verses $h\nu$ for un doped SnS and Al doped, it is clear from this figure a red shift after doping films with Al concentration in band gaps values.

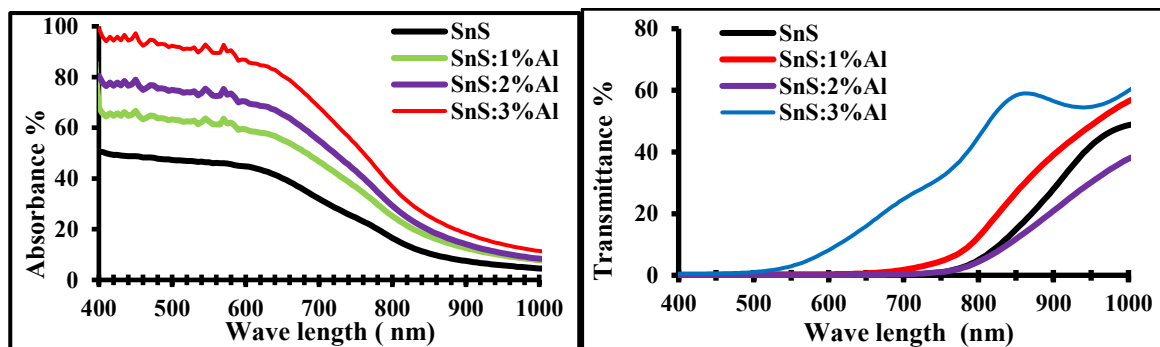


Figure 4 Absorbance besides Transmittance against wavelength for thin film SnS besides Al dopant SnS in different doping ratio.

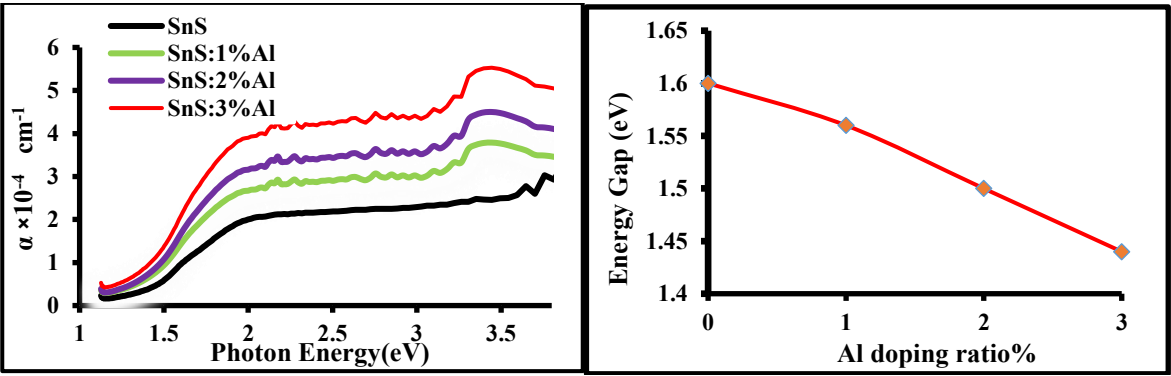


Figure 5 Optical energy gap plus absorption coefficients of pure SnS thin film then Al doped.

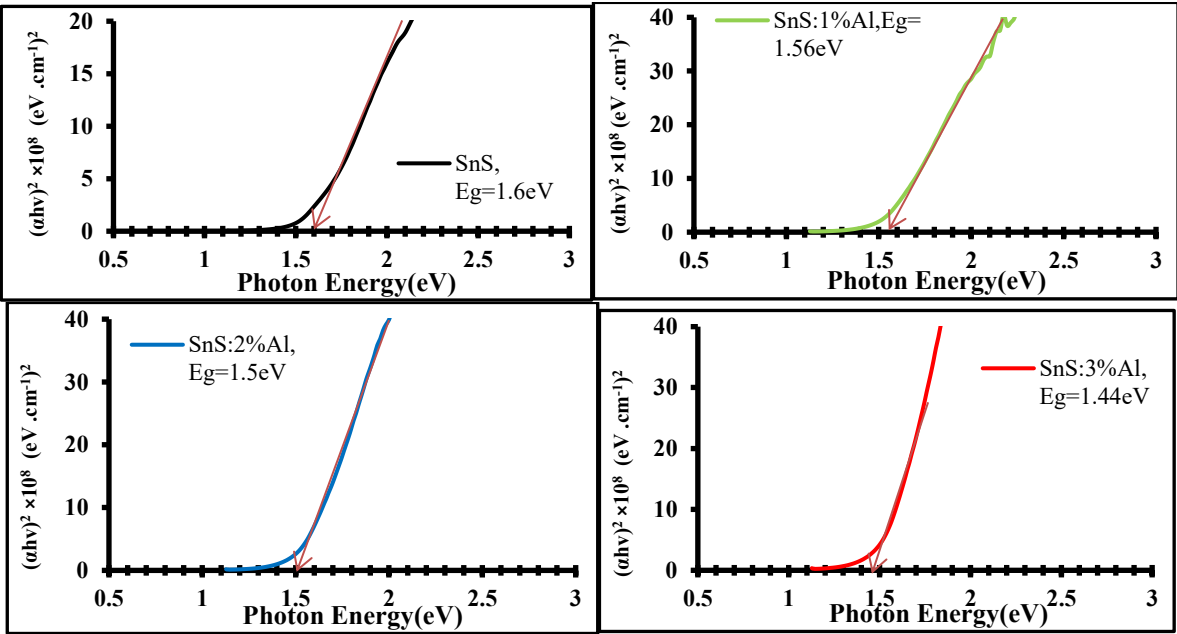


Figure 6 Distinction $(\alpha h\nu)^2$ besides Al dopant SnS in different doping ratio.

Table 2 Directs optically gaps and absorption coefficient of pure SnS also dopant Al.

Sample	Egopt (eV)	$\alpha \times 10^4 \text{ (cm}^{-1}\text{) at 550 nm}$
SnS	1.6	2.254
SnS:1%Al	1.56	2.883
SnS:2% Al	1.5	3.414
SnS:3% Al	1.44	4.208

3.4 Electrical properties

The electrical parameters like carriers concentrate (n_H), resistivity (ρ), types of major carriers, mobility (μ_H) and Hall coefficient R_H for SnS pure and doped with Al are determined using Hall Effect measurement as in Table (3). This figure displays all SnS films undoped and doped have positive signs of Hall coefficients which means that they had (p-type) conductivity [1,2,17], this behavior as result Tin vacancies which generally noticed in SnS thin films, led to make acceptor states neighboring the valence band, permitting hole conduction [1,2]. It is clear from table (3) and figure (7) that all parameters affected by doping Al ratio where the electrical characteristic improvement after doping. Carrier concentration values for SnS films increased after doping with Al as a result decreased in the values of Hall coefficient whereas the values of resistivity and mobility decreased. When the current and voltages which transient in SnS films increase, perhaps collisions between carriers increase makes possible drop the values of mobility. Pure SnS film shows low carrier concentration ($3.6 \times 10^{10} \text{ cm}^{-3}$) with high resistivity ($1.34 \times 10^5 \text{ } \Omega \cdot \text{cm}$), some studies similarly result for SnS films high values of resistivity with little hole concentration [2, 3,39-41] with this result remains a challenge for use it as absorber layers for photovoltaic applications. Hole concentration enhanced by Al doping with different ratio reached maximum value of ($2.81 \times 10^{16} \text{ cm}^{-3}$), as well as resistivity reductions and conductivity improvement, may be Al substitution at Sn sites, lead to presents acceptor levels [1]. Also, Al doping creates localized states within the band gap that sustains carrier generation but increase scattering events, reducing mobility. This result correlates with the optical properties of these films.

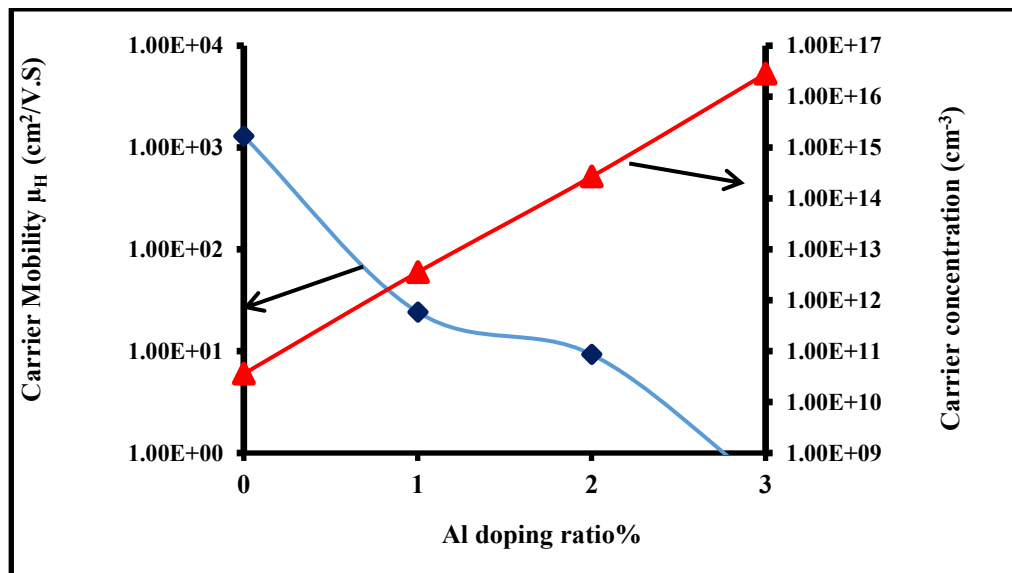


Figure 7 Carriers concentration then mobility aimed at pure besides Al doped for SnS thin films.

Table 3 Hall effects constraint at pure besides Al doped for SnS thin films.

Sample	Carrier concentration (cm ⁻³)	Carrier Mobility μ_H (cm ² /V.S)	σ ($\Omega.cm$) ⁻¹	Resistivity ρ ($\Omega.cm$)	R_H (cm ³ /C)
SnS	3.60E+10	1.29E+03	7.44E-06	1.34E+05	1.74E+08
SnS:1%Al	3.55E+12	2.42E+01	1.38E-05	7.26E+04	1.76E+06
SnS:2% Al	2.69E+14	9.30E+00	4.00E-04	2.50E+03	2.32E+04
SnS:3% Al	2.81E+16	4.67E-01	2.096 E-3	4.77E+02	2.23E+02

3.5 *I-V characteristics*

Current density voltage curve below dark and illumination (100 mW/ cm²) conditions of p-SnS/n-Si and p- SnS:Al/n-Si heterojunction as a function of Al ratio are shown in Figure (8). This figure shows under dark conditions the value of dark current is low due to the lowest production of charge carriers [17], as well as dark forward current is formed by major carrier movements while extremely weak reverse current. The values of open-circuit voltage (V_{oc}), short-circuit current density (J_{sc}), V_m , I_m , fill factor (F.F) and the conversion efficiency of fabricated solar cell (η) are calculated by equations [17,42] and shown in Table 4:

$$F.F = \frac{J_m \times V_m}{J_{sc} \times V_{oc}} \times 100\% \quad (7)$$

$$\eta = \frac{F.F \times J_{sc} \times V_{oc}}{P_{in}} \times 100\% \quad (8)$$

The high value of efficiency η with V_{oc} , J_{sc} and F.F is found 5.512%, 0.56 mV, 16.96 mA/ cm² and 0.58, respectively, for p-SnS:3%Al/n-Si heterojunction due to many reasons [17]. When Al concentration increases in films, (1) the crystal quality improved which decreases internal defects, which cause decreases photo-excited carriers recombination leading to improve conversion efficiency with recorded lesser fill factor, (2) Increase SnS grain size after doping lead to increase hopping of particle-to-particle on the photo-induced carrier and small bandgap as a result of great absorption in visible light. Additionally, controlling the effect of Al doping ratio improved properties of charge carrier transport by reducing the recombination of electron-hole pair and caused in a higher open-circuit voltage and short-circuit current density, enhanced the conversion efficiency from 1.438% to 5.512%. These results are good matched with ref [3,5,17,43].

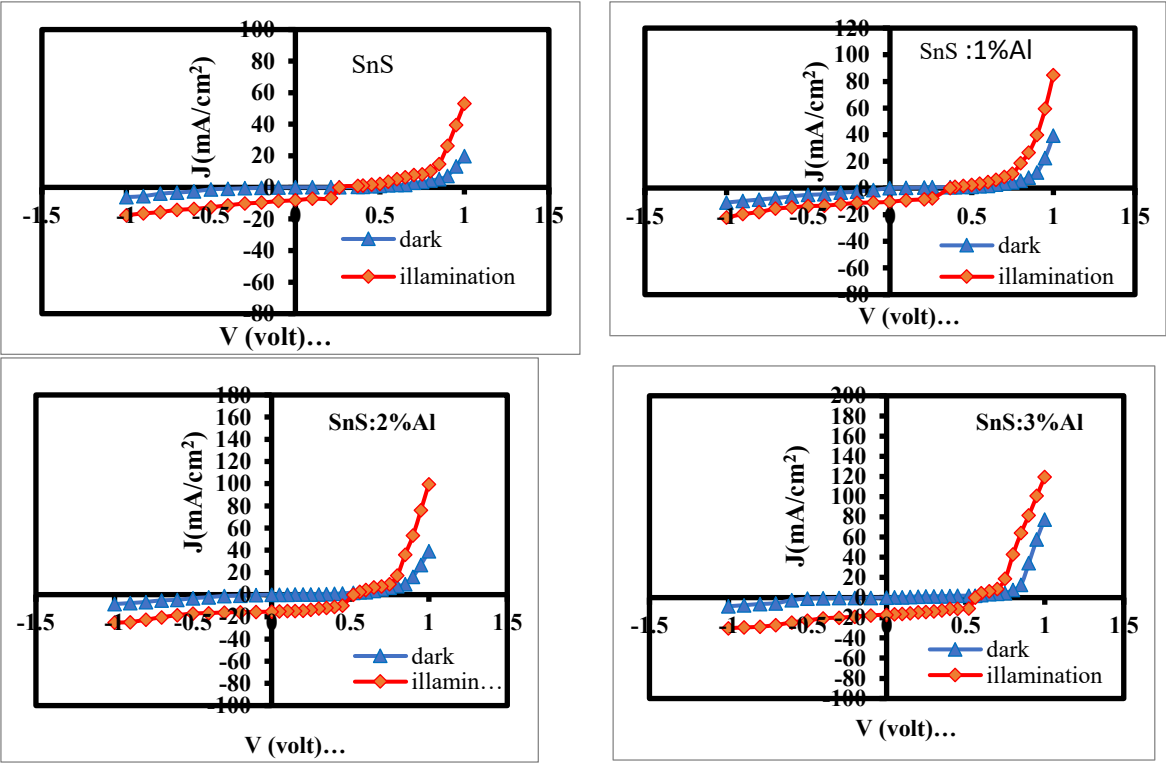


Figure 8 J-V characteristic for pure SnS and p-SnS:Al /n-Si heterojunction thin films.

Table 4 Solar cell factors for pure SnS and p-SnS:Al /n-Si heterojunction thin films.

Sample	Voc (V)	Jsc (mA/cm ²)	Vmax (mV)	Jmax (mA/cm ²)	F.F	η %
SnS	0.26	8.19	0.21	6.85	0.675542	1.4385
SnS:1%Al	0.37	10.4	0.26	7.77	0.525	2.0202
SnS:2% Al	0.52	15.25	0.45	9.89	0.561223	4.4505
SnS:3% Al	0.56	16.96	0.52	10.6	0.580357	5.512

Table 5 Summary of some results on SnS solar cells.

Efficiency	Reference
0.285%	17
5.41%	44
1.34%	45
6.27%	46
3.88%	47
2.9%	48
3.16%	49

4. CONCLUSIONS

Un doped SnS and (1,2,3) % of Al doped films were successfully synthesized on glass and Si substrate thru thermal evaporation method, the effect of Al ratio on SnS thin film properties (structural, optical, also electrical) and on heterojunction solar cell (p- SnS:Al/n-Si) were studied. XRD analysis confirmed all films prepared are poly crystal with single phase of orthorhombically structure with (111) as favored orientation, also increase films crystallite size with Al ratio where a higher value for the sample doped with 3%Al. At higher doping levels ($> 3\%$), a slight decrease in crystallinity was observed, which may be related with lattice distortion due to the difference in ionic radii between Al and Sn atoms. Optical analysis revealed the opposite dependence of energy gap and optical transmittance through Al concentration and 3% doping concentration results a band gap of 1.44 eV. Hall Effect measurements show all films possesses p-type conductivity and SnS:3%Al produced the high charge carrier concentration of $2.81 \times 10^{16} \text{ cm}^{-3}$, mobility of $4.67 \times 10^{-1} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and high conductivity of $2.096 \times 10^{-3} (\Omega \cdot \text{cm})^{-1}$ with lowest resistivity of $4.77 \times 10^2 \Omega \cdot \text{cm}$. I-V characteristic for heterojunction device (p- SnS:Al/n-Si) under 100 mW/cm^2 illumination were influenced by Al ratios and the best photovoltaic conversion efficiency of 5.512% was examined for the device fabricated with SnS:3%Al thin film due to improved structural, optical and electrical properties compared to other samples. Based on collective structural, optical, and electrical analyses, 3% Al doping SnS films provides the optimal balance for thin-film solar cell applications. Overall, the results reveal that moderate Al doping, particularly around 3%, enhances SnS thin films properties (the structural quality, optical absorption, and electrical conductivity), making them suitable for applications in optoelectronic devices and solar cells.

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