

Effects of Na doping on CdS thin films and n-CdS/p-Si solar cells via chemical bath deposition method

Vagif Nevruzoglu^a, Murat Tomakin^b, Eyup Fahri Keskenler^{c,*} and Gokturk Ozturk^a

^aDepartment of Energy Systems Engineering, Recep Tayyip Erdoğan University, Rize 53100, Turkey

^bDepartment of Physics, Recep Tayyip Erdoğan University, Rize 53100, Turkey

^cDepartment of Material Science and Nanotechnology Engineering, Recep Tayyip Erdoğan University, Rize 53100, Turkey

Un-doped and Na doped CdS thin films were deposited by chemical bath deposition method. The effects of Na doping on structural, optical and electrical properties of CdS thin films and n-CdS/p-Si heterojunction were investigated. Crystal structure of all CdS thin films was cubic with (111) preferred direction. High Na doping deteriorated crystal structure and amorphous structure obtained for 3% Na doping. The grain size of thin films was decreased from 6.5 nm to 4.8 nm and surface homogeneity increased with Na doping. All samples had high band gap for CdS (2.42 eV) due to quantum size effect and band gap of the samples was increased 3.65 eV to 3.84 eV as a function of Na content. Na doped CdS samples had higher resistivity and carrier concentration than that of un-doped CdS. Ideality factors of Na doped n-CdS/p-Si heterojunctions were greater than unity due to nanostructured CdS, which indicates that the diode exhibits a nonideal behavior. It was determined that photovoltaic behavior of n-CdS/p-Si prepared with high Na doped CdS dependent on different semiconductor structure (such as depend on function of Na₂SiO₃ phase) occurred interface of CdS and Si.

Key words: CdS, Na doping, Heterojunction, CBD Method, Solar Cell.

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Introduction

Cadmium sulfide (CdS) with a direct band gap of 2.42 eV has been adopted as one of the most significant II-VI semiconductors and has been studied extensively during the past decades [1]. n-CdS is commonly employed as window layer on p-CdTe solar cell because of its wide band gap (~2.42 eV) since solar light is minimally absorbed in the n-CdS layer with the wide band gap [2]. CdS has also been investigated for Si based optoelectronic devices and photovoltaic applications [3,4]. Considering the leading position of single crystal silicon in large-scale device integration, the study on Si-based CdS systems is highly expected [5]. In recent years, nanostructural materials like nanowire and quantum-dot-based devices [6] have been widely investigated due to its novel properties. So, nano-CdS is a hopeful material for semiconductor applications. Also, impurities in semiconductors often cause dramatic changes in their structural, electrical and optical properties. Therefore, CdS thin films have often included atoms in a metallic character, such as Cu [7], Ag [8], B [9], Ni [10], Mn [11] etc. But studies on alkali metal like Na doped CdS nano structures are

very limited and CdS thin film doped with alkali metal exhibits interesting changes in the optical properties [12]. CdS thin films can be grown through many techniques, such as physical vapor deposition (PVD) [13], close-spaced-sublimation (CSS) [14], spray pyrolysis [15], and chemical bath deposition (CBD) [16]. Among these techniques CBD method is the most widely used for preparing of nanostructured thin films and also, it is easy to control the growth rate and to produce reproducible, uniform and adherent film [17-19]. In this paper we report the effects of Na addition to CdS thin films prepared by chemical bath deposition method on structural, optical and electrical properties of the films and on electrical properties of n-CdS/p-Si heterojunction. I-V characteristics of these devices were measured to establish a correlation between the Na doping and the electrical properties of n-CdS/p-Si heterostructure.

According to our best knowledge, there is no study about Na doping effect on interface processes occurring between the sandwich structures of Si/CdS solar cell.

Experimental Details

Un-doped and Na doped CdS thin films were prepared on glass (microscope slides) and boron doped p-Si (orientation=<111>, resistivity= 1.5-6 Ω-cm, thickness= 525 μm, doping concentration=10¹⁶ cm⁻³)

*Corresponding author:
Tel : +90-464-2236126 / 1235
Fax: +90-464-2237514
E-mail: keskenler@gmail.com

substrates by chemical bath deposition method. Cleaning procedure of Si and glass substrates were given in our previous work [20]. 0.035 M cadmium acetate ($\text{Cd}(\text{CH}_3\text{COO})_2 \times 2\text{H}_2\text{O}$), 0.07 M thiourea $((\text{NH}_2)_2\text{CS})$ and 1 M ammonium acetate ($\text{NH}_4\text{CH}_3\text{COO}$) were used for the initial solution in distilled water. The doping was performed by adding sodium hydroxide (NaOH) into the solution. Doping concentrations (Molar ratios, CdAc./NaOH) were 1%, 2% and 3%. Solution pH was set to around 10 by the ammonium hydroxide solution (5.0 N NH_4OH). Deposition temperature of CdS thin films was 90 °C. Second process in this study was preparing of n-CdS/p-Si heterojunction. Therefore, Na doped and undoped CdS samples were prepared on p-Si substrates to investigate the n-CdS/p-Si heterojunction. Indium have used for ohmic electrical contacts. Indium contacts have been created with vacuum evaporation method. Obtained In/n-CdS/p-Si/In structure annealed at 450 °C for 15 minute in air. The crystal structure of thin films was examined by X-ray diffraction (XRD) using Rigaku Smartlab with CuK_α radiation ($\lambda = 1.5408 \text{ \AA}$) over the range $2\theta = 10\text{--}70^\circ$ at room temperature. Jeol JSM 6610 scanning electron microscope (SEM) was used for morphological information. Compositional analysis was studied by Oxford Instruments Inca X-act energy dispersive X-ray spectroscopy (EDS) attached to the SEM. The thickness of the films was determined by gravimetric method. The thickness of the samples is approximately 150 nm. The transmittance spectra of the films were measured by Perkin Elmer Lambda 35 spectrophotometer in the spectral range of 300–1000 nm at room temperature. Carrier concentrations and carrier type of CdS samples were found by Hall Effect measurements (in dark) at room temperature. Carrier type of CdS samples was found n-type. The resistivity of the samples was measured by Van der Pauw method in dark at room temperature. The current-voltage (I - V) characteristics of n-CdS/p-Si heterojunction were studied in dark and under standard light conditions (93 mW/cm², AM1.5D) using a Sciencetech Inc. solar simulator. The electrical measurements were performed using a Keithley 2410 sourcemeter and a Lakeshore 336 temperature controller.

Results and Discussion

X-ray diffraction results of undoped and Na doped CdS samples were given in Fig. 1. The XRD analyses indicated that the CdS thin films had polycrystalline nature and there is only one peak at $2\theta \sim 26.5^\circ$. On the other hand, the formation of wide diffraction peak about $\sim 26.5^\circ$ can be attributed some amorphous or nanostructure components [21–23]. This peak is (002) belonging to the hexagonal form or (111) peak of the cubic structure. So, it is difficult to index due to the proximity of the diffraction angle positions. However, CdS samples could be structured in cubic phase due to

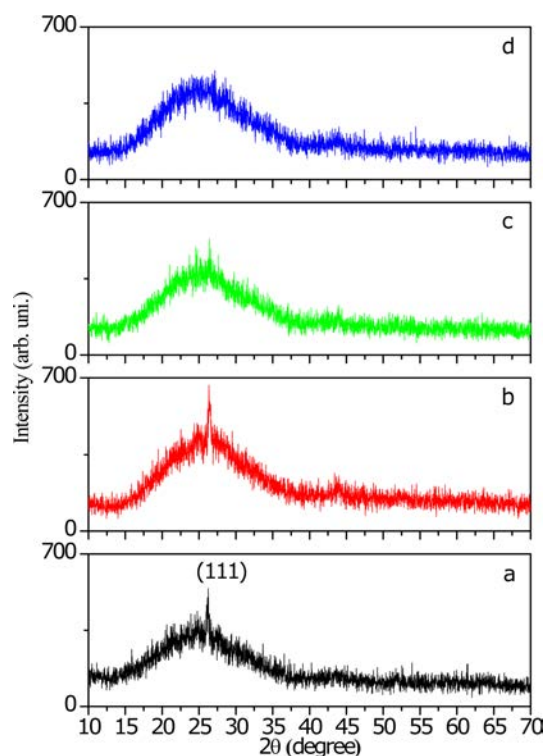


Fig. 1. XRD pattern of (a) pure, (b) 1% Na doped, (c) 2% Na doped and (d) 3% Na doped CdS thin films.

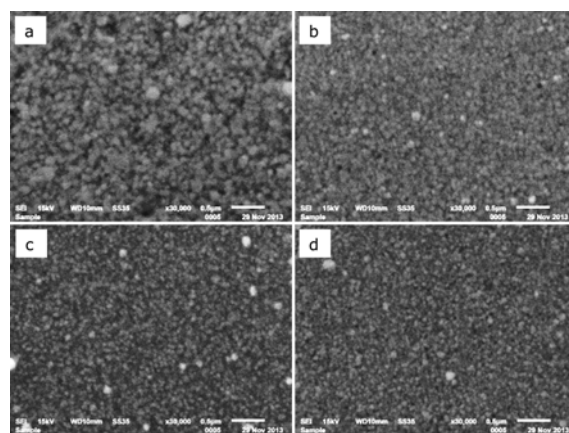


Fig. 2. SEM micrographs of (a) un-doped, (b) 1% Na doped, (c) 2% Na doped and (d) 3% Na doped CdS samples.

the absence of (100) and (101) plans of the hexagonal phase which are generally appear in hexagonal CdS structures [24]. Similar results were found by other researchers for chemical bath deposited CdS [8, 25, 26]. The intensity of (111) reflection increased for 1% Na doped CdS and then decreased for 2% Na doped samples. (111) peak for 3% Na doped CdS was disappeared, which shows that the crystallinity of the sample was deteriorated.

a lattice constant of CdS samples was determined from the diffraction angle of the (111) peak for undoped, 1% Na and 2% Na doped samples and the calculated results were 5.881 Å, 5.863 Å and 5.846 Å,

Table 1. Doping ratios, band gap values and grain sizes for undoped and doped CdS films obtained from EDS and Optical measurements.

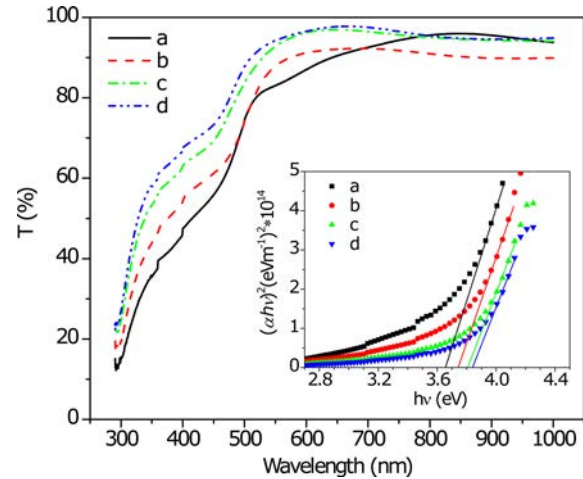
Samples	Cd (at.%)	S (at.%)	Na (at.%)	Cd/S	E_g (eV)	Grain size (nm)
Un doped	55.8	44.2	—	1.26	3.65	6.5
1% Na	56.5	42.8	0.7	1.32	3.74	5.5
2% Na	53.7	43.9	2.4	1.22	3.81	5.0
3% Na	50.5	44.8	4.7	1.13	3.84	4.8

respectively. The ionic radius of Na^+ ions (0.116 nm at tetrahedral sites) is larger than that of Cd^{2+} ion (0.097 nm at tetrahedral sites). Therefore, the decreasing of lattice parameter is not expected for substitutional incorporation of Na ions into Cd sites. The observed decrement in the a parameter may be arisen from the possible introduction of Na ions into interstitial sites. Similar result was found by Mageswari and co-worker for Na doped CdS sample prepared with CBD [27].

The surface properties of the CdS thin films were investigated by SEM and obtained results were shown in Fig. 2. It could be seen from Fig. 2 that the grain size and surface roughness of thin films was decreased and grains shows homogeneous size and more closed packet structure with Na doping.

The composition ratio of the films was determined by EDS analysis. The atomic percent (at.%) of Cd, S and Na in the films are listed in Table 1. The all samples are Cd-rich because of Cd/S ratio is larger than 1, meaning that the samples have sulfur vacancies. Cd/S ratio firstly increased from 1.26 to 1.32 for 1% Na doping and then decreased to 1.13 with higher Na doping. EDS analysis confirms the inclusion of Na dopant in the doped films.

The optical window transparency has a major impact for enhancing the efficiency of a solar cell. Fig. 3 shows the optical transmittance for all samples between wavelength range 300–1000 nm. Optical transmittance of the CdS thin films increased with Na doping. The reason of increasing in optical transmittance can be related to reduce of surface roughness and formation of more closed packet structure with Na doping. The band gap of CdS samples, E_g , was determined by $\alpha = A(h\nu - E_g)^{1/2}$, which is used for direct-gap materials, where A is a constant. The variation of $(\alpha h\nu)^2$ as a function of $h\nu$ is shown in the inset of Fig. 3. $(\alpha h\nu)^2$ curve as a function of $h\nu$ shows a linear behavior near the band gap. The value of E_g is given by the intercept of the straight line with the $h\nu$ axis. The obtained band gap values were listed in Table 1. The band gap of the as-deposited samples increased from 3.65 eV to 3.84 eV with Na doping which is due to decrease in grain size as increasing doping ratio. Similar increment for band gap was found by Yellaiah and co-worker [28] for Na

**Fig. 3.** Optical transmittance spectra and band gap value (inset figure) of (a) un-doped, (b) 1% Na doped, (c) 2% Na doped and (d) 3% Na doped CdS thin films.

doped $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{S}$ powders. These values were larger than band gap value of bulk CdS (2.42 eV). Observed large shift reveals quantum size effect in the CdS thin films. The increased band gap makes nanostructured CdS a more effective window material in photovoltaic applications. The band gap of nanostructure CdS film is related to the grain radius (R) [24]:

$$E_g^{\text{nano}} = E_g^{\text{bulk}} + \frac{\hbar^2 n^2}{2R^2} \left(\frac{1}{m_e^*} + \frac{1}{m_h^*} \right) - \frac{1.8e^2}{4\pi\epsilon_0\epsilon_r R} \quad (1)$$

Where $\hbar = \frac{h}{2\pi}$ is Planck constant, R is the radius of nanoparticles, $m_e^* = 0.19m_0$ is the effective mass of electron $m_h^* = 0.8m_0$ is the effective mass of hole, ϵ_r is the dielectric constant of material (5.7) and ϵ_0 is the permittivity of free space. The Coulomb term (third term in Eq. 1) of electron-hole interaction is generally small compared to electron-hole confinement kinetic energy (second term in Eq. 1). The obtained grain size values were listed in Table 1. It was determined from Table 1 that grain size decreased increasing Na content. We found same result from SEM images.

The resistivity (ρ) and carrier concentration (n) value of all films was listed in Table 2. The dark resistivity value increased from 487 $\Omega\text{-cm}$ to 784 $\Omega\text{-cm}$ for 1% Na doping and then resistivity slowly decreased to 665 $\Omega\text{-cm}$ with high Na doping. The electrical properties of CdS thin films prepared by chemical bath deposition method depend on Cd/S ratio [29], used chemical amount [30], solution temperature [31], characteristics of doping atoms [32], mobility of atoms in crystal structure [33]. Also decreasing in thin film thickness increases the resistivity [34, 35]. We can introduce scattering from grain boundaries in nano-sized samples for another mechanism about resistivity. As known decreasing or increasing of grain sizes in crystal structure affects distance between grain boundaries and so resistivity values are affected [36]. In this study

increment in resistivity value can be due to grain size decreasing with Na doping. However, decreasing of resistivity for high doping concentration can occur from decreasing of surface roughness and formation of more closed packet structure on surface of samples with Na doping. The carrier concentration of samples increased from $5.34 \times 10^{13} \text{ cm}^{-3}$ to $5.20 \times 10^{15} \text{ cm}^{-3}$ up to 2% Na doping and for 3% Na doping the carrier concentration decreased to $6.42 \times 10^{14} \text{ cm}^{-3}$ value. In this study, observed changes in the carrier concentration can be attributed to natural behavior of Na atoms in CdS. As known, Na atoms have amphoteric characteristic in CdS crystal structure and in this structure they moves with disosiatif mechanism. Namely the movement of Na in CdS structure originates with two mechanisms: i) movement in empty lattice points and ii) movement in regions between lattice points. When Na atoms locate into lattice points of CdS crystal they have an acceptor characteristic, and when Na atoms locate into interstitial regions they have a donor characteristic, which increase the carrier concentration [27, 37]. In this study we found from XRD measurement that Na atoms located interstitial sites in CdS lattice. Therefore the carrier concentration increased with Na doping. In the production process of CdS thin films arises the defects and defects complexes which have various characteristics. One of these defects is vacancy Cd sites (V_{Cd}) [38, 39]. It was determined from EDS analysis that Na doping greatly reduced Cd content for especially high Na doping, which can cause some vacancy Cd sites. Na atoms in V_{Cd} sites gain acceptor characteristics and so decrease carrier concentration in 3% Na doped sample.

In the second phase of this study, un-doped and Na doped CdS thin films precipitated on single crystal p-Si with CBD method and n-CdS/p-Si structures obtained. Fig. 4 shows J - V curves of n-CdS/p-Si heterojunctions under dark condition. As seen from figure, all samples show rectifier feature and threshold voltage values were increasing depending on Na doping amount.

The thermionic emission current - voltage relation of a p-n heterojunction is given by [40];

$$I = I_s(e^{qV/nkT} - 1) \quad (2)$$

where I_s is the saturation current, q is electronic charge, V is applied voltage, n is the ideality factor, k is Boltzmann constant and T is the absolute temperature. The ideality factor can be calculated from the slope of

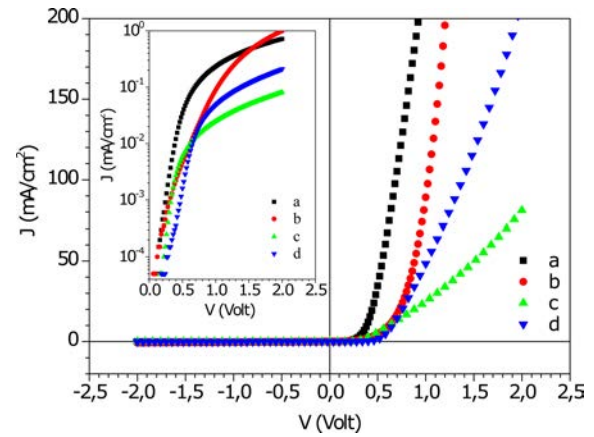


Fig. 4. J - V characteristics for (a) un-doped, (b) 1% Na doped, (c) 2% Na doped and (d) 3% Na doped n-CdS/p-Si structure. The inset figure shows $\ln J$ - V characteristics of the n-CdS/p-Si structure.

the straight line region of $\ln(I)$ - V graph and can be written as,

$$n = \frac{q}{kT} \frac{dV}{d(\ln I)} \quad (3)$$

I_s can be determined by an extrapolation of $\ln I$ - V curve to $V = 0$.

The variation of logarithmic current density as a function of bias voltage is shown in the inset of Fig. 4. The ideality factor and current mechanism were determined with the help of these spectrums for all samples. The calculated values of n and J_s ($= I_s/A$, A : diode area) were given in Table 2. Ideality factors of all samples are greater than unity, which indicates that the diode exhibits a nonideal behavior so that that thermionic emission is not only the conduction mechanism for current flow. If ideality factor varies between 1 and 2, the tunneling current mechanism is dominant. If $n = 2$, the generation-recombination current mechanism is the dominant. If $n > 2$, it means the leakage current mechanism is dominant [20]. Higher ideality factor values like in this study have been reported for several nanostructure device studies [41, 42].

The CdS/Si junction exhibits a rectifying behavior that can be shown from the I - V measurements. However, the surface/interface properties of semiconductor materials play a major role in the rectifying behavior of the heterojunction. The saturation current and ideality factor

Table 2. The photovoltaic parameters measured under light condition of un-doped and Na doped n-CdS/p-Si heterojunctions.

	ρ (Ω -cm)	n (cm^{-3})	Ideality factor	J_s ($\mu\text{A}/\text{cm}^2$)	J_{sc} (mA/cm^2)	V_{oc} (V)	η (%)
Un-doped	487	5.34×10^{13}	5.02	7.4	13.65	0.26	0.99
1% Na	784	4.80×10^{15}	7.24	9.9	17.05	0.31	1.11
2% Na	655	5.20×10^{15}	3.98	2.2	8.25	0.23	0.54
3% Na	665	6.42×10^{14}	9.29	6.0	12.05	0.27	0.62

generally increase with increasing surface states. The CdS/Si junction exhibits a rectifying behavior that can be shown from the I-V measurements. Interface states density is occurred by lattice parameters mismatching (about 7%) in crystal structure when n-CdS and p-Si heterojunction are contacted. SiO_x , CdO_x , and SO_x etc. interface states can occur by contacting two sides. These states density of interface structure is in the range of 10^{14} - 10^{15} cm^{-3} [43-45]. In addition, it is known that indium contacts which performed on CdS surface diffuse into CdS layer due to relatively high annealing temperature (450°C) and so carrier concentration of CdS layer increases. Dzhaferov et al. found for similar study that carrier concentration of CdS increased from $1.4 \times 10^{13} \text{ cm}^{-3}$ to $1.3 \times 10^{16} \text{ cm}^{-3}$ due to In diffusion [46].

In this study, CdS thin films have nano-sized grain structure and grain size decreased with Na doping. In this situation, surface/interface states in CdS/Si junction occur. According to this result, it is expected that ideality factor and saturation current increase continuously with Na doping. However, the ideality factor showed fluctuated changing depending on Na doping. Therefore, it can be said that there are different effects on CdS/Si junction. For example, ideality factor depend on serial resistance of a p-n junction [47]. Also, indium used as ohmic contacts can diffuse into Si in annealing process of n-CdS/p-Si and thus acceptor levels occur in forbidden band gap [46]. Additionally, Cd and S atoms can diffuse into p-Si in annealing process. Indium is the shallow acceptor impurity in Si whereas the Cd and S are the deep-level acceptor and donor impurities, respectively [48, 49]. However, Dzhaferov and co-worker were found that the photovoltaic parameters of In/n-CdS/p-Si cells improved after In, Cd and S diffusion into Si [46]. In literature, studies on Na doped Si samples are very limited. It has been reported that Na atoms have complex action in Si crystal structure [50, 51]. As a result, one should consider the different competing processes in evaluating the rectifying behavior of n-

CdS/p-Si junction.

The photovoltaic parameters measured under light condition of un-doped and Na doped n-CdS/p-Si heterojunctions have been given in Table 2 and Fig. 5. Open circuit voltage (V_{oc}), short circuit current (J_{sc}) and efficiency (η) showed fluctuated changing with Na doping. Photovoltaic parameters of n-CdS/p-Si cell increased after 1% Na doping. The low resistivity and high carrier concentration for CdS films decrease the device sheet and series resistance and so one obtain high V_{oc} and J_{sc} [52, 53]. In our study, resistivity and carrier concentration of 1% Na doped CdS sample increased. But increment of carrier concentration (order of $\sim 10^2$) is very large from increment of resistivity (times ~ 1.6). So it can be said that improvement in photovoltaic parameters in due to increasing carrier concentration. Also, increasing of photovoltaic parameters for 1% Na doped CdS sample can be attributed improvement of crystallinity and surface properties [54]. Photovoltaic parameters of n-CdS/p-Si cell for 2% Na doping decreased. Electrical properties of 2% Na doped CdS were relatively better than that of 1% Na doped CdS. Also, grain size and surface properties of 1% and 2% Na doped CdS samples were similar. Therefore, explaining of deterioration in photovoltaic parameters of n-CdS/p-Si heterojunction for 2% Na doped CdS is difficult. But, photovoltaic parameters for 2% Na doping can be explained with nearly amorphous structure. We found that 3% Na doped CdS sample had amorphous structure and lower grain size according to other samples. Also, 3% Na doped CdS sample had higher resistivity and lower carrier concentration than that of 2% Na doped CdS sample. Therefore we expect a decreasing in photovoltaic parameters of n-CdS/p-Si prepared with 3% Na doped CdS sample. Whereas, photovoltaic parameters of n-CdS/p-Si were improved again for 3% Na doping. Thereby we cannot explain increasing in photovoltaic parameters with properties of CdS for 3% Na doping. This behavior can arise from phase (such as Na_2SiO_3) formed interface of CdS and Si due to diffusion of Na. Na diffuses into the Si substrate with same time the other Cd and S atoms in Na doped CdS thin films which annealed at 450°C for 15 min. in air atmosphere [55]. It can be come out that Na_2SiO_3 structure can occur between the interfaces of films according to the results obtained from photo-electrical measurements.

Conclusions

In this study, Undoped and Na doped CdS thin films were deposited on glass and p-Si by chemical bath deposition method. XRD studies showed that some amorphous components were observed for all films and crystallinity of thin films was deteriorated with Na doping. It was determined from decrement in lattice parameter that Na ions introduced into interstitial sites.

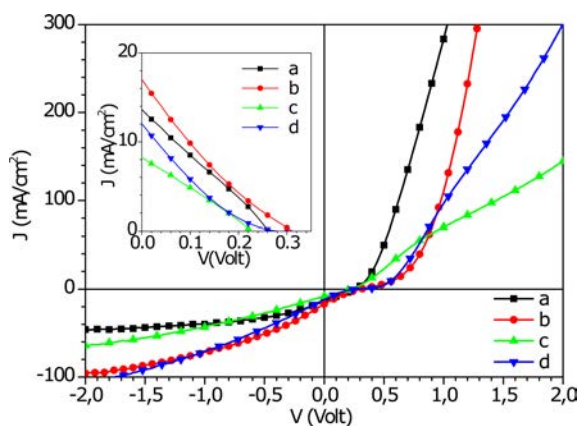


Fig. 5. J-V characteristics (with inset figure) for (a) un-doped, (b) 1% Na doped, (c) 2% Na doped and (d) 3% Na doped n-CdS/p-Si structure for under light condition.

The grains of all CdS films were nanosize and grains shows homogeneous size and more closed packet structure with Na doping. It was determined from EDX studies that the thin films were Cd-rich. Increasing or decreasing in resistivity value explained depends on grain size and surface properties of samples. Carrier concentration increased due to Na atoms located interstitial sites in CdS lattice. But in 3% Na doped sample carrier concentration decreased located Na atoms in V_{Cd} sites, which are gain acceptor characteristics. The experimental I-V characteristics of the n-CdS/p-Si structure were significantly affected from Na doping. It was determined that the different competing processes (such as surface/interface states, serial resistance, diffusion of In, Na, Cd and S atoms into p-Si) effected the rectifying behavior of n-CdS/p-Si junction. Improvement in photovoltaic parameters of n-CdS/p-Si junction for 1% Na doped CdS sample was explained with high carrier concentration, good crystallinity and low surface roughness. However, it was determined that dominant parameter which affected photovoltaic behavior of n-CdS/p-Si prepared with high Na doped CdS was different semiconductor structure (such as Na_2SiO_3) occurred interface of CdS and Si dependent.

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