



The International Conference on Technologies and Materials for Renewable Energy, Environment and Sustainability, TMREES14

Influence of Al-doped ZnO transparent contacts deposited by a spray pyrolysis technique on performance of HIT solar cells

F.Z. Bedia^{a,b,c*}, A. Bedia^{a,b,c}, M. Aillerie^{a,b}, N. Maloufi^d, F. Genty^{a,b}, B. Benyoucef^c

^aUniversité de Lorraine, LMOPS-EA 4423, 57070 Metz, France.

^bSupélec, LMOPS, 57070 Metz, France.

^cURMER, Abou-Bakr Belkaid University, 13000 Tlemcen Algeria.

^dUniversité de Lorraine, LEM3-UMR CNRS 7239, 57000 Metz, France.

Abstract

Transparent and conductive Al-doped ZnO (AZO) thin films were deposited by spray pyrolysis and analysed in the aim to improve optical and electrical properties involved in the efficiency of Heterostructure with Intrinsic Thin Layer (HIT) solar cell. X-ray diffraction measurement shows that AZO film grown on glass has (002) preferred orientation. High optical transmittance value of ~80% in the visible region was observed and the optical band gap was found to be 3.31eV at room temperature. The influence of AZO thin films as transparent conductive oxide TCO on heterojunction with intrinsic thin-layer (HIT) solar cell performance was investigated using software simulation. The beneficial effect of implementing AZO front contact for increasing electrical energy conversion properties of HIT solar cell compared to the reference cell without the AZO layer.

© 2014 Elsevier Ltd. This is an open access article under the CC BY-NC-ND license

(<http://creativecommons.org/licenses/by-nc-nd/3.0/>).

Selection and peer-review under responsibility of the Euro-Mediterranean Institute for Sustainable Development (EUMISD)

Keywords: AZO, TCO, HIT solar cell, Efficiency;

1. Introduction

Transparent conductive oxide (TCO) films have high transmittance in the visible region of the electromagnetic spectrum combined with reasonable high conductivity. TCO films have therefore been used extensively as

* Corresponding author. Fatima Zohra Bedia.

E-mail address: f_bedia@yahoo.fr.

components in optoelectronic devices with applications in Heterostructure with Intrinsic Thin Layer (HIT) solar cells, light emitting diodes (LEDs), flat and touch panel displays [1-3]. Zinc oxide (ZnO) has been actively investigated as an alternative to the standard Indium Tin Oxide (ITO) material used in these above mentioned applications, because ZnO is cheaper, non-toxic and readily available. Impurity-doped ZnO is more appropriate for TCO applications because the electrical conductivity of undoped ZnO is not high enough to be used as TCO [4, 5]. Doping ZnO with Group III elements (Al, In and Ga) drastically lowers the electrical resistivity [6-8] of the material due to partial substitution by dopant element of Zn ions, thereby creating additional energy levels in the band gap. These energy levels play significant role in the observed optical transmittance of ZnO films. Therefore, the optimization of the concentration of the dopant element is of prior importance in the aim of an improvement of optical and electrical properties of ZnO films. In addition, doped ZnO thin films, such as AZO and Ga-doped ZnO (GZO), with a textured surface structure as well as a high transmittance in the near infrared region have recently attracted much attention for transparent electrode applications in silicon solar cells [9, 10].

Hetero-junction with intrinsic thin layer (HIT) cells, first developed by Sanyo Ltd., are silicon solar cell that combine the high efficiency of crystalline silicon (c-Si) cells with the low temperature deposition technology of hydrogenated amorphous silicon (a-Si:H). This structure produces both the emitter and the back surface field (BSF) layers. The high-quality performance of HIT solar cells has been demonstrated with efficiencies over 22% in laboratory cells and over 20% in mass production cells based on textured n-type Czochralski (CZ) crystalline silicon (c-Si) [11,12].

Modeling of HIT cells was started by van Cleef et al [13] using the AMPS computer code [14], which however does not have a proper built-in optical model; and using the derivative of the AMPS program (D-AMPS), where a fairly good optical model has been introduced [15]. The numerical PC program AFORS-HET [16, 17] has been developed especially for simulating HIT solar cells. The PC-1D program [18], developed at the University of New South Wales, Australia for modeling textured mono-crystalline silicon solar cells, has also been fairly successful in modeling HIT cells. The program ASDMP by Chatterjee *et al.* [19], has also been extended to model N-a-Si:H/P-c-Si type front (with a heterojunction only on the emitter side) HIT cells [20].

For our study, we have used the numerical device simulator SCAPs [21] to simulate the output characteristics of HIT's on P-c-Si substrates. SCAPs is a one-dimensional solar cell device simulator, developed at ELIS, University of Gent, which is freely available to the PV research community. The user can define a solar cell as a series of layers with different properties, such as thickness, doping densities and defect distribution. It is then possible to simulate a number of common measurements: I-V, QE, C-f, and C-V.

This paper is devised into two parts, first starting with the presentation of the results of investigations of the structural and optical properties of AZO film prepared by spray pyrolysis method. The second part presents the simulation studies used to evaluate the performance of the so-obtained AZO/a-Si:H(n)/a-Si:H(i)/c-Si(p)/Al-BSF/Al HIT solar cell. Finally, the resulting discussion associated to the experimental and simulation works, mainly concerns the conversion efficiency η , the short circuit current density J_{sc} , the open circuit voltage V_{oc} and quantum efficiency (QE) as a function of the AZO sprayed characteristics.

2. Experimental part. Elaboration then structural and optical properties of AZO film

2.1. Elaboration of AZO film

The AZO film was sprayed on glass substrates by the spray pyrolysis method. A solution of 0.08M of dehydrated zinc acetate $Zn(CH_3COO)_2$ (with 99.99% of purity) is dissolved in methanol (CH_3OH) and simultaneously deposited at 350 °C under an air condition. Aluminum chloride $AlCl_3$ (with 99.999% of purity) was added as dopant source with a ratio Al/Zn equal to 1%. The crystal structures and the preferential orientation of the AZO films were analyzed by X-ray diffraction (Bruker D8 Advanced Diffractometer) in $\theta-2\theta$ scan mode. The scanning electron microscope (SEM) was employed for the observation of the surface morphologies. The optical transmission spectra were measured using UV-Vis-NIR spectrometer (Perkin-Elmer Lambda 900 spectrometer).

2.2 Structural characterizations of AZO film

The X-Ray Diffraction (XRD) pattern of AZO film is shown in Fig. 1. The XRD diffraction peaks of $2\theta = 31.09$, 34.54 , 36.34 , 47.64 , 56.63 , 62.94 , and 78.10 , which can be attributed to (100), (002), (101), (102), (110), (103), and (112) planes of the hexagonal structure, respectively [22]. Additionally, the intensity of the (002) peak is obviously stronger than the other peaks, suggesting that AZO crystallites are highly oriented with the c-axis being perpendicular to the substrate. The lattice parameter values obtained for our ZnO films are found equal to $a = 3.317$ Å and $c = 5.188$ Å, which is in good agreement with values reported in literature [22]. Scanning Electron Microscopy (SEM) images of the AZO film grown by spray pyrolysis are shown in Fig. 2. We observed an arrangement of the nanoparticles mostly uniform and only slightly irregular confirming the existence of additional diffraction peaks in the XRD pattern of the film. The average size of ZnO nanoparticles is in the range of 100–150 nm.

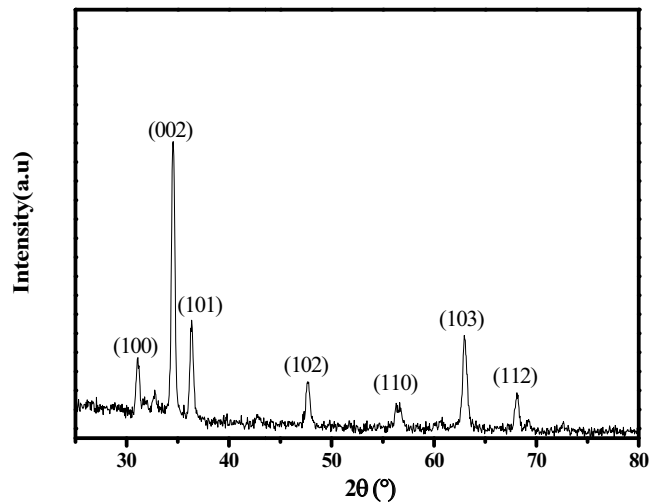


Fig. 1. XRD pattern of the AZO thin film on a glass substrate.

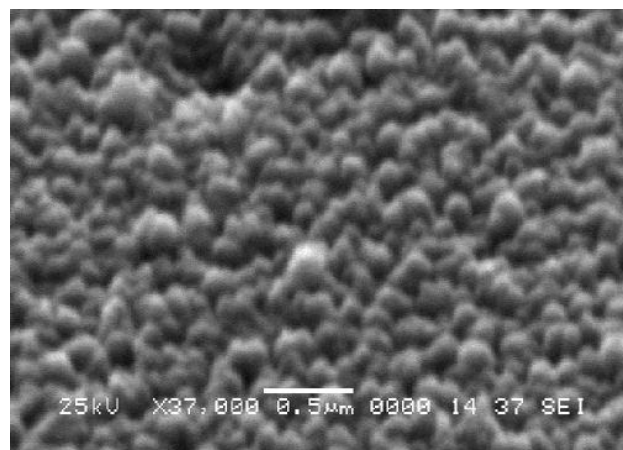


Fig. 2. SEM image of the AZO thin film.

2.3 Optical characterizations of AZO film

We report in Fig. 3, the optical transmittance of AZO films grown glass substrate using glass spectrum as reference. The absorption coefficient behaviour is shown in the Inset of Fig. 3. We have experimentally found that the transmission values of the film are low at short wavelengths and the values are high at long wavelengths. So, the film behaved as an opaque material because of its high absorbing properties at short wavelengths and as a transparent material at long wavelengths. The AZO film shows a good transparency with the average transmittance beyond 80% in the visible range and above 90% in the near infrared.

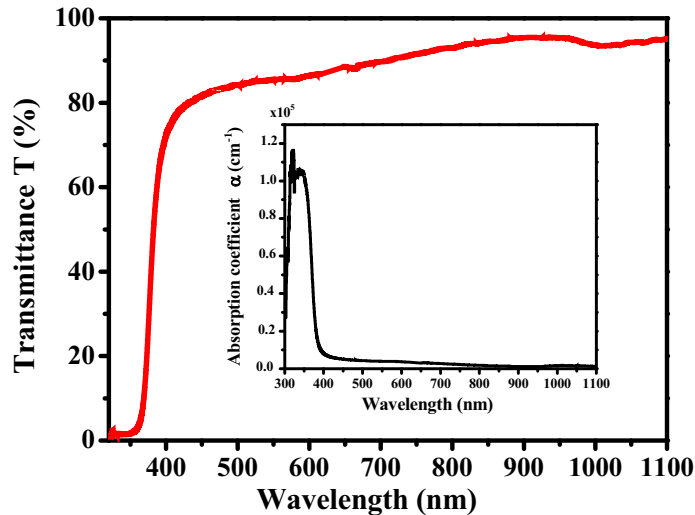


Fig. 3. Optical transmission spectra of AZO film.

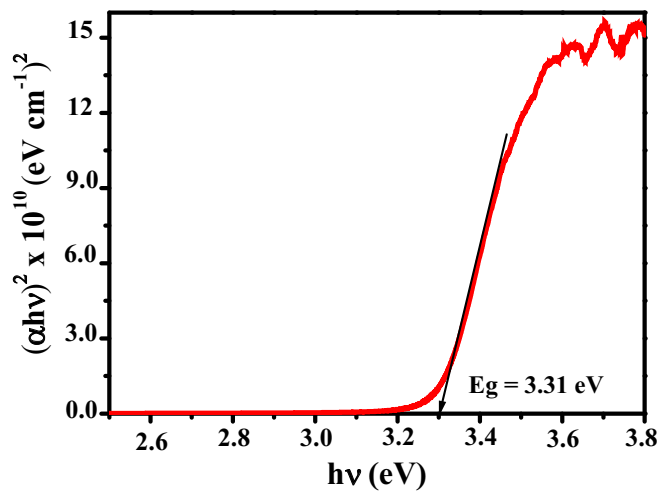


Fig. 4. The plots of $(\alpha h\nu)^2$ as function of photon energy of the AZO thin film.

In order to calculate the bandgap of the AZO film, we used the Tauc's relationship [23] as follows:

$$(\alpha h\nu) = [A(h\nu - E_g)]^{1/2} \quad (1)$$

where A is a constant, h is Planck's constant, ν is the photon frequency, E_g is the optical band gap, and α is the absorption coefficient (inset Fig.3), which is calculated from transmittance spectrum [24]. We have plotted in Fig. 4 the curve $(\alpha h\nu)^2$ as function of the photon energy ($h\nu$).

An extrapolation of the linear region of the curve of $(\alpha h\nu)^2$ on the y-axis against the photon energy ($h\nu$) on the x-axis gives the value of the bandgap E_g . According to the measured absorption spectra reported in Fig. 4, the bandgap value was evaluated equal to 3.31 eV.

3. Simulation part. Integration of AZO film in HIT solar cell

3.1. Solar cell structure

The simulated solar cell structure is a AZO/a-Si:H(n)/a-Si:H(i)/c-Si(p)/Al-BSF/Al, as shown in Fig. 5. In simulation of the HIT solar cell, the transparent conductive oxide, TCO, is the AZO layer prepared in the present study by the spray pyrolysis method and thus, we have taken in consideration parameters found during the structural and optical characterization, as presented above. The SCAPS software environment, which is a one-dimensional solar cell device simulator, was employed to simulate this cell.

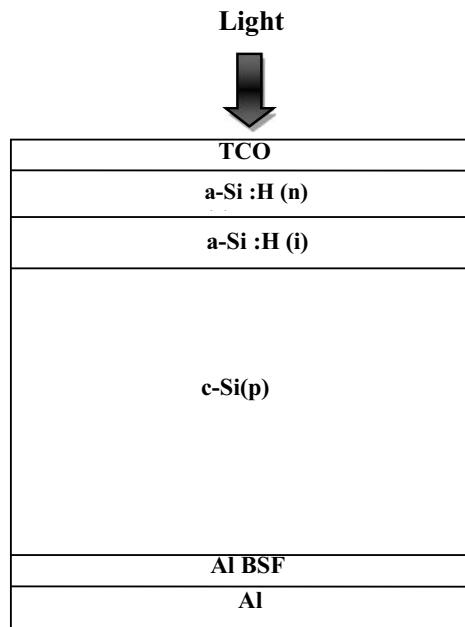


Fig. 5. Schematic structure of TCO/a-Si(n)/ a-Si:H(i)//c-Si(p)/Al BSF/Al solar cell.

Oxygen defects in c-Si were fixed at 0.55 eV above the edge of the valence band, and the capture cross-sections for electrons and holes were both $1.0 \times 10^{-14} \text{ cm}^2$ [16]. The interface state density of the a-Si:H/c-Si interface was chosen as $1.0 \times 10^{11} \text{ cm}^{-2} \text{ eV}^{-1}$. The front and the back contacts were assumed as flat band ones allowing to neglect the contact potential influence. The surface recombination velocities of electrons and holes were both set as $1.0 \times 10^7 \text{ cm/s}$. The recombination mechanisms (band to band recombination, Auger recombination and Shockley–Read–Hall

recombination) were considered during this simulation, and results are reported in Table 1. The other parameters used in the simulation and summarized in Table 1 are taken from Ref [16, 17]. The sunlight illuminates the right side of the solar cell. An AM1.5G solar spectrum is used for the optical generation to simulate current–voltage (I–V) curves under standard one-sun illumination conditions at intensity of 100 mW/cm². The light reflection of front contact and back contact was set to be at standard values for such devices, i.e. 0.1 and 1, respectively. The optical absorption of a-Si:H layer was set to be the absorbance a-Si-WronskiAdachi extracted from a default parameter file in SCAPs software and, c-Si is obtained from the compendium of the open source computer program PC1D, commonly used in the solar cell community for simulation of crystalline semiconductor devices. The optical absorption of AZO layer used our experimental data (inset Fig.3). In the solar cell device physics, the important parameters to be discussed are the open circuit voltage (Voc), the short circuit current density (Jsc) and the efficiency (η). Voc is the voltage when no current pass through the cell, where as Jsc is the current when no voltage is applied.

Table 1. Performance of HIT solar cells for simulation.

	c-Si(p)	a-Si :H (i)	a-Si:H (n)	Al-BSF
Layer thickness (nm)	3x10 ⁵	3	10	5x10 ³
Dielectric constant	11.9	11.9	11.9	11.9
Electron affinity (eV)	4.05	3.9	3.9	4.05
Band gap (eV)	1.12	1.72	1.74	1.12
Effective conduction band density (cm ⁻³)	2.8x10 ¹⁹	1x10 ²⁰	1x10 ²⁰	2.8x10 ¹⁹
Effective valence band density (cm ⁻³)	1.04x10 ²⁰	1x10 ²⁰	1x10 ²⁰	1.04x10 ²⁰
Electron mobility (cm ² V ⁻¹ s ⁻¹)	1040	5	5	202.4
Hole mobility (cm ² V ⁻¹ s ⁻¹)	412	1	1	77.15
Doping concentration of acceptors (cm ⁻³)	1x10 ¹⁶	0	0	1x10 ²⁰
Doping concentration of donators (cm ⁻³)	0	0	1x10 ²⁰	0
Thermal velocity of electron (cm s ⁻³)	1x10 ⁷	1x10 ⁷	1x10 ⁷	1x10 ⁷
Thermal velocity of hole (cm s ⁻³)	1x10 ⁷	1x10 ⁷	1x10 ⁷	1x10 ⁷
Auger recombination coefficient for electron (cm ⁶ s ⁻¹)	2.2x10 ⁻³¹	0	0	2.2x10 ⁻³¹
Auger recombination coefficient for hole (cm ⁶ s ⁻¹)	9.9x10 ⁻³²	0	0	9.9x10 ⁻³²
Direction band to band recombination coefficient (cm ⁶ s ⁻¹)	1.1x10 ⁻¹⁴	0	0	1.1x10 ⁻¹⁴

3.2. Simulation results

Reliable solar cell modelling must be based on accurate and reliable optical and electrical modelling parameters of the device. The AZO film grown on glass substrate was applied as transparent front contact for a HIT solar cell device and its properties and efficiency were compared with those of a reference cell, as proposed in the simulation software environment.

Table 2 lists corresponding HIT solar cells characteristics found during the simulation. We also report in Fig. 6 and Fig. 7, the current density - voltage (J - V) characteristics and the quantum efficiency as function of the wavelength, respectively, for the reference HIT solar cell and the HIT solar cell with our AZO film.

Table 2. Performance of HIT solar cells with and without AZO.

	Jsc (mA/cm ²)	Vco (mV)	η (%)
Hit solar cell with sprayed AZO	36.32	722	20.12
Reference solar cell without AZO contact	29.05	689	16.86

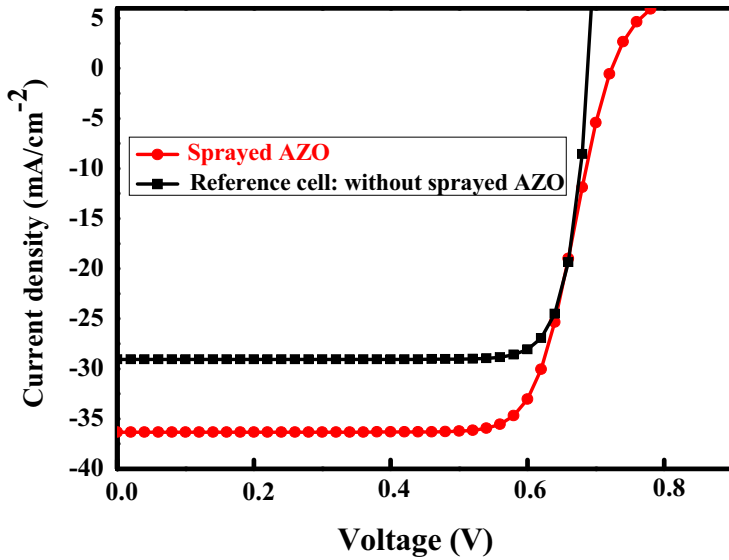


Fig. 6. Current density as a function of voltage for AZO/a-Si:H(n)/a-Si:H(i)/c-Si(p)/Al-BSF/Al solar cells with and without AZO.

The beneficial effect of integration AZO front contact for increasing the J_{sc} in comparison to the reference solar cell without the AZO layer is clearly observed in Fig.6.

The highest efficiency η , equal to 20.12 % was obtained for the HIT cell with sprayed AZO, where as the reference delivered only 16.86% under identical conditions. Also, the same high open circuit voltage, V_{oc} was achieved with the sprayed AZO. These results are in good arrangement with results of literature [16, 17].

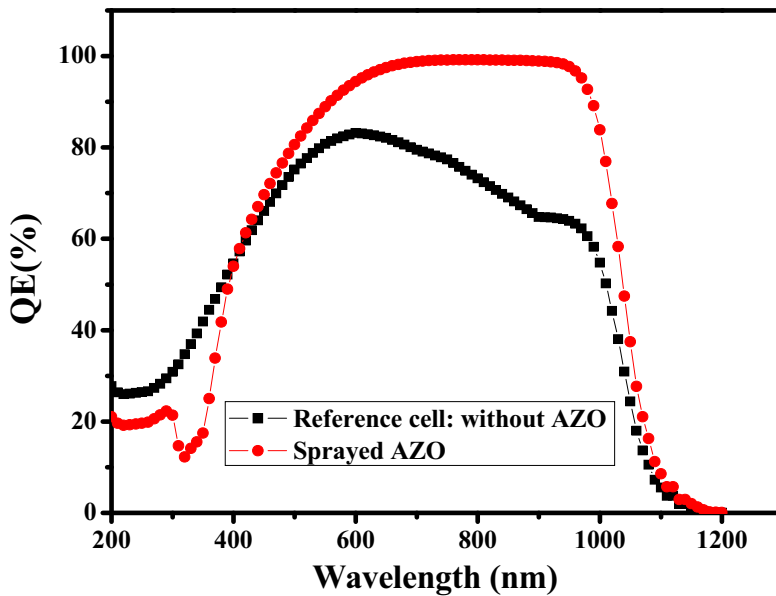


Fig.7. Quantum efficiency of AZO/a-Si:H(n)/a-Si:H(i)/c-Si(p)/Al-BSF/Al solar cells with and without AZO.

In ref. [16], Zhao et al. have fabricated HIT silicon solar cells with conversion efficiency above to 19 % whereas the thin solar cells fabricated by Dao et al. [17] have a conversion efficiency of 17.86%. This comparison with results of literature points out the good quality of the AZO thin films obtained in the present study. In Fig.7, the QE measurements show that the solar cell with the AZO front contact presents a higher response in the near-UV-Visible-near-IR region (400 nm- 1200nm) than the reference cell. Thus, as it is well-known that the performance of a heterojunction cell critically depends on TCO front contact [16], the reported QE measurements confirm this possible improvement of the functioning of HIT solar cell by an integrating of AZO layer in the structure.

4. Conclusions

AZO films were deposited on glass substrates by spray pyrolysis in the aim to get optimized thin film for Heterostructure with Intrinsic Thin Layer or HIT solar cells. Structural characterizations of the AZO film, using XRD and SEM techniques point out an arrangement of the nanoparticles mostly uniform highly oriented with the c-axis being perpendicular to the substrate. The experimental determination of the band-gap, obtained by transmission measurements, gives a value of 3.31 eV. SCAPs (a Solar Cell Capacitance Simulator) simulation program was used to investigate the effect of AZO film properties on the operating parameters and efficiency of AZO/a-Si:H(n)/a-Si:H(i)/c-Si(p)/Al-BSF/Al HIT solar cells. The high efficiency η of 20.12% with a short circuit current density J_{sc} equal to 36.32 mA/cm² and a higher response in the Visible-near-IR region (400 nm- 1200nm), compared to HIT cell without AZO film were obtained for the HIT solar cell when implementing AZO at the back contact. This result demonstrates the feasibility of using spray-pyrolysis technique for the deposition of large area AZO contacts for solar cell applications

References

- [1] Kim D, Yun I, Kim H. Fabrication of rough Al doped ZnO films deposited by low pressure chemical vapor deposition for high efficiency thin film solar cells. *Current Appl. Phys.* 2010;10:S459-S462.
- [2] Fernández S, Gandía JJ. Texture optimization process of ZnO:Al thin films using NH₄Cl aqueous solution for applications as antireflective coating in thin film solar cells. *Thin Solid Films* 2012;520:4698-4702.
- [3] Szarko JM, Song JK, Blackledge CW, Swart I, Leone SR, Li S, Zhao Y, Optical injection probing of single ZnO tetrapod lasers. *Chem. Phys. Lett.* 2005;404:171-176.
- [4] Chen J, Chen D, Zhou Y, Li W, Yanjie R, Hu L. Electrochemical deposition of Al-doped ZnO transparent conducting nanowires arrays for thin-film solar cell electrodes. *Mater. Lett.* 2014;117:162-164.
- [5] Hirahara N, Onwona-Agyeman N, Nakao M. Preparation of Al-doped ZnO thin films as transparent conductive substrate in dye-sensitized solar cell. *Thin Solid Films* 2012;520:2123-2127.
- [6] Prasada Rao T, Santhosh Kumar MC. Physical properties of Ga-doped ZnO thin films by spray pyrolysis. *J. Alloys Compds* 2010; 506:788-793.
- [7] Wienke J, Booi AS. ZnO:In deposition by spray pyrolysis-influence of the growth conditions on the electrical and optical properties. *Thin Solid Films* 2008;516:4508-4512.
- [8] Chen XL, Wang F, Genga HX, Zhang KD, Wei CC, Zhang XD, Zhao Y. Natively textured surface Al-doped ZnO-TCO layers with gradual oxygen growth for thin film solar cells via magnetron sputtering. *Appl. Surf. Sci.* 2012,258: 4092-4096.
- [9] Yen WT, Lin YC, Ke JH. Surface textured ZnO:Al thin films by pulsed DC magnetron sputtering for thin film solar cells applications. *Appl. Surf. Sci.* 2010, 257: 960-968.
- [10] Crossay A, Buecheler S, Kranz L, Perrenoud J, Fella CM, Romanyuk YE, Tiwari AN. Spray-deposited Al-doped ZnO transparent contacts for CdTe solar cells. *Solar Energy Materials & Solar Cells* 101 (2012) 283–288 10-12.
- [11] Sawada T, Terada N, Tsuge S, Baba T, Takahama T, Wakisaka K, Tsuda S, Nakano S. High-efficiency a-Si:H/c-Si heterojunction solar cells, in: *Proceedings of the First WCPEC Conference, Hawaii, USA, 1994, 1219-1226.*
- [12] Taira S, Yoshimine Y, Baba T, Taguchi M, Kanno H, Kinoshita T, Sakata H, Maruyama E, Tanaka M. Our approaches for achieving HIT solar cells with more than 23% efficiency. In: *Proceedings of the 22nd European Photovoltaic Solar Energy Conference, Milan, Italy, 3–7, 2007; 932-935.*
- [13] Van Cleef MWM, Schropp REI, Rubinelli FA. Significance of tunnelling in p+ amorphous silicon carbide n crystalline silicon heterojunction solar cells. *Appl. Phys. Lett.* 1998;73:2609-2611.
- [14] Mc Elheny P, Arch JK, Lin HS, Fonash S. Range of validity of the surface photovoltage diffusion length measurement: A computer simulation. *J. Appl. Phys.* 1988; 64:1254-1265, ISSN 0021-8979.
- [15] Pla J, Tamasi M, Rizzoli R, Losurdo M, Centurioni E, Summonte C, Rubinelli F. Optimization of ITO layers for applications in a-Si/c-Si heterojunction solar cells. *Thin Solid Films* 2003; 425:185-192.

- [16] Zhao L, Li HL, Zhou CL, Diao HW, Wang WJ. Optimized resistivity of p-type Si substrate for HIT solar cell with Alback surface field by computer simulation. *Solar Energy* 2009; 83: 812-816.
- [17] Dao VA, Heo J, Choi H, Kim Y, Park S, Jung S, Lakshminarayan N, Yi J. Simulation and study of the influence of the buffer intrinsic layer, back-surface field, densities of interface defects, resistivity of p-type silicon substrate and transparent conductive oxide on heterojunction with intrinsic thin-layer (HIT) solar cell. *Solar Energy* 2010; 84: 777-783.
- [18] Basore P. A Numerical modeling of textured silicon solar cells using PC-1D. *IEEE Transaction on Electron Devices* 1990; 37: 337-343.
- [19] Chatterjee P. A computer analysis of the effect of a wide-band-gap emitter layer on the performance of a-Si:H-based heterojunction solar cells. *J. Appl. Phys.* 1996; 79:7339-7347.
- [20] Nath M, Chatterjee P, Damon-Lacoste J, Roca I, Cabarrocas P. Criteria for improved open-circuit voltage in a-Si:H(N)/c-Si(P) front heterojunction with intrinsic thin layer solar cells. *J. Appl. Phys.* 2008; 103: 034506-1-9.
- [21] Burgelman M, Nollet P, Degraeve S. Modelling polycrystalline semiconductor solar cells. *Thin Solid Films* 2000; 361-362, 527-532.
- [22] JCPDS Card No. 36-1451 (ZnO hexagonal).
- [23] Pankove JI. *Optical Processes in Semiconductors*, Prentice-Hall Inc., Englewood Cliffs, NJ, 1971.
- [24] Shanmuganathan G, Shameem Banu IB, Krishnan S, Ranganathan B. Influence of K-doping on the optical properties of ZnO thin films grown by chemical bath deposition method. *J. All. Comps.* 2013;562:187-193.