International Journal of Research
Available at https://edupediapublications.org/journals

e-ISSN: 2348-6848
p-ISSN: 2348-795X
Volume 04 Issue 13
October 2017

Optimization of type and concentration of dopant (Sb and Al) for ZnO thin films prepared by spray pyrolysis technique and their applications in perovskite solar cells

Nitu Kumari[1], Sanjaykumar R. Patel[2], Jignasa V. Gohel[3]

[1,2,3] *Chemical Engineering Department S. V. National Institute of Technology, Surat-395007 (Gujarat), India.*

Email: nitujhabit@gmail.com, srp@ched.svnit.ac.in, sjn@ched.svnit.ac.in

Abstract:

In the present study, doped and pure ZnO films are prepared by low cost spray pyrolysis technique. Three types of dopants (Al and Sb) are used and their doping concentration is optimized successfully. The absorption spectra of prepared films are carried out by UV-VIS spectrophotometry. The films are characterized by X-ray diffraction (XRD) for structural analysis. Both the pure and doped ZnO films are consequently applied in perovskite solar cell device preparation. Additionally, solar cell device is made with low cost CuI (as hole transport material) and carbon (as counter electrode). The solar cell efficiency was measured by solar simulator connected with Kiethly source meter under one sun illumination. The power conversion efficiency (PCE) obtained for 8% Al-doped ZnO is maximum (3.47%) among all devices.

Keywords

ZnO films; Doping; Copper iodide; carbon; Perovskite Solar Cell

1. Introduction

Oxide materials are very easy to produce in thin as well as thick forms with various conducting as well as non conducting substrates. These oxide materials have various advantages. ZnO is the most important n-type semiconductor having hexagonal wurtizite structure. It is an important optoelectronic device material among II-IV semiconductors [1]. During the past few years, ZnO has attracted the interest of many researchers due to its high chemical stability, low dielectric constant, high electron mobility, good transparency, high electrochemical coupling coefficient and high luminous transmittance [2]. ZnO has some advantages over other oxide materials such as In_2O_3, $CdSnO_4$, TiO_2 and SnO_2 due to its unique combination of properties like low cost, non- toxicity, as well as, better electrical, optical and piezoelectric performance. These advantages make ZnO as a promising material for many different applications such as solar cells, gas sensors [3]. Doping of metals like silver, cadmium, aluminum, and antimony can be used to enhance the properties of ZnO films [4-7].

ZnO thin films of high quality for solar cell applications can be formed by various techniques. Spray pyrolysis technique is preferred among all techniques, because it is simple, non-vacuum, less expensive and versatile than other methods [8].

Energy demand to complete our requirement is massively increasing day by day and this situation will continue till a final solution exists [9]. In order to maintain economic and environmental stability, it is necessary to get an abundant supply of energy [10]. Moreover, the development of carbon- free sources of energy becomes one of the major scientific challenges for us. Solar energy has become a potential alternative energy source after the energy crisis in 1970s [11]. Solar cells convert solar energy directly into electricity, which has become a major research interest within academia and industry. Today almost 90% of the PV units produced are made from crystalline silicon which is composed of large area p-n junctions and others from thin films devices based on amorphous silicon [12]. These devices based on p-n hetero-junctions absorb part of the sunlight (absorption from 0.8–0.9 eV) and convert this light energy into electrical energy by the photovoltaic effect [13]. Laboratory devices with efficiencies over 25% have been demonstrated, and the best commercial cells have now reached efficiencies of 17%–18% [14]. However, the processing cost of silicon solar cell is very high and it drives people to develop low cost solar cell. Perovskite solar cell (PSC) is a third generation solar cell and it is a promising potential candidate in this field. Generally, PSC contains four major layers (electron transport layer, a photoactive layer, a hole transport layer and counter electrode). Basically, solar radiation is absorbed by perovskite to generate electron-hole pairs. ZnO films are most favourably used as ETL in PSC, owing to its wide band gap, stability and simple synthesis processes. ZnO thin film is basically responsible for collection of electrons from excited perovskite layer and its transfer to the external load.

In this paper, study is focused on preparation of doped and pure ZnO thin films using low cost spray pyrolysis technique. The prepared ZnO films are applied in PSC fabrication. Additionally low cost CuI and carbon are used as hole transport material and counter electrode respectively.

2. Materials and Methodology

2.1. Materials

Fluorine doped tin oxide (FTO) glass, lead iodide (PbI_2), methyl amine (CH_3NH_2, 33 wt% in ethanol), hydroiodic acid (HI, 57 wt% in water), copper iodide (CuI), propyl sulphide and chlorobenzene were purchased from Sigma Aldrich. Zinc acetate dihydrate ($Zn(O_2CCH_3)_2(H_2O)_2$) was purchased from Finar Chemicals Ltd., Ahmadabad, India. Isopropanol (C_3H_8O) was purchased from HPLC Pvt. Ltd., Mumbai, India. Antimony nitrate ($Sb(NO_3)_3$), and alumina (Al_2O_3) were purchased from S.D. Fine Chem, Mumbai, India. All chemicals were of analytical grade and were used without further purification. All experiments were carried out at ambient condition. Deionized (DI) water (Elix 10, millipore) was used for preparing all the aqueous solutions. Before use, FTO was cleaned with detergent and sonicated in acetone, iso-propyl alcohol (IPA) and de-ionised water in an ultrasonic bath for 10 minutes separately.

2.2. Methodology

2.2.1. Deposition of pure and doped ZnO films

In this study, ZnO thin films are deposited by the method reported in our previous study [8]. To make doped ZnO films, similar process was adopted. For doping, the weight percent of $Sb(NO_3)_3$ and Al_2O_3 was varied from 0.5-1.5% and 2-10% respectively.

2.2.2. Perovskite solar cell fabrication

Firstly, ZnO films were deposited as ETL on pre-treated FTO substrate. Further, a multilayer stack was prepared by adding further a photoactive layer of $CH_3NH_3PbI_3$. This layer was coated by the two-step spin coating method: (i) PbI_2 was dissolved in N, N-dimethyl formamide (DMF) under constant stirring to get clear solution. PbI_2 solution was added drop-wise on FTO/ZnO/Ag:ZnO and spin coated at 1500 rpm (for 30 s). (ii) CH_3NH_3I layer was further deposited. CH_3NH_3I was dissolved in IPA. It was further deposited by spin coating (at 2000 rpm for 30 s) by drop-wise addition of CH_3NH_3I solution upon PbI_2 layer. Subsequently, it was immediately kept on a hot plate at 90ºC for 30 min. Change in color was observed from yellow to black, which confirms the proper formation of perovskite layer. Copper iodide layer as HTL was further coated on FTO/ZnO/ /$CH_3NH_3PbI_3$. CuI solution was prepared by dissolving CuI in the mixture of propyl sulphide and chlorobenzene. The solution was subsequently spin coated on perovskite layer and annealed at 80ºC for 1 h [15]. Carbon paste was used as the low cost counter electrode. It was casted on FTO/ZnO/$CH_3NH_3PbI_3$ /CuI (by doctor blade), followed by drying. The final structure of device is FTO/ZnO/$CH_3NH_3PBI_3$/CuI/ carbon.

2.3. Characterization

The structural characterization was investigated with XRD method (Rigaku D/Max 2200). Surface morphology was studied by FESEM (Nova NanoSEM 450, FEI limited), and optical property of the films were measured by UV-Vis-NIR spectrophotometer (Hach-DR6000) respectively. The PCE of device was studied using SS150 solar simulator (ScienceTech, Canada) connected to the keithly (2401) I-V measurement system.

3. Results and Discussion

3.1. Optical characterization

Figures 1 and 2 shows the Tauc's plot of Sb and Al doped ZnO at different wt% of Sb and Al respectively. It is clear from the Fig. 1 that 1.2 wt% Sb:ZnO has lowest band gap among all prepared Sb:ZnO films. Figure 2 depicts that, the band gap of 8 wt% Al:ZnO is lowest among all prepared Al: ZnO films.

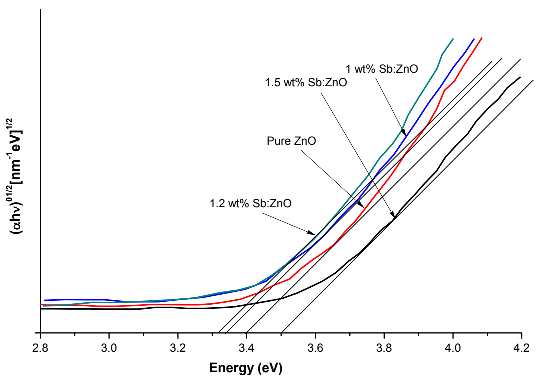

Figure 1. Tauc's plot for pure and Sb doped ZnO films with various Sb wt%.

e-ISSN: 2348-6848
p-ISSN: 2348-795X
Volume 04 Issue 13
October 2017

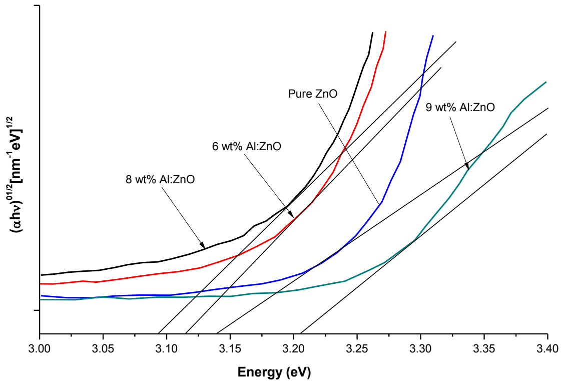

Figure 2. Tauc's plot for pure and Al doped ZnO films with various Al wt%.

3.2. PCE analysis

The prepared ZnO films (doped and pure) are further applied in perovskite solar cell fabrication. The current-voltage characteristics were measured by solar simulator connected with kiethly source meter under one sun illumination. The J-V curve is shown in Figs. 5 and 6. The data obtained by J-V curve is shown in Table 1.

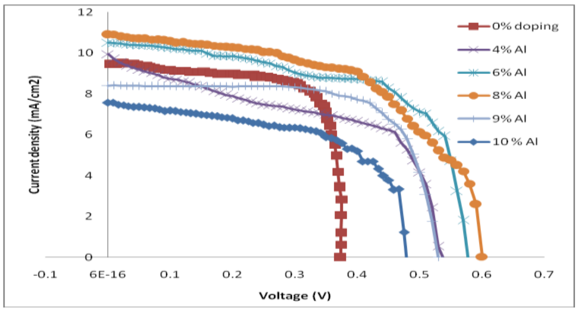

Figure 3. J-V curve of pure and Al-doped ZnO films

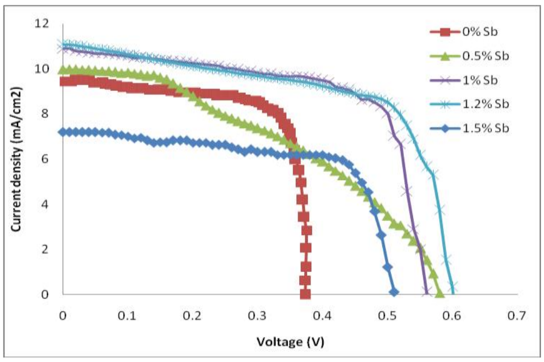

Figure 4. J-V curve of pure and Sb-doped ZnO films

Table 1. J-V data of device obtained at optimum doping condition

Doping in ZnO (weight %)	Current density, J_{sc} (mA/cm^2)	Voltage, V_{oc}(V)	Fill Factor (%)	PCE (%)
0	9.45	0.373	35.3	3.21
1.2 (Sb)	11	0.61	50	3.38
8 (Al)	10.9	0.59	54	3.47

4. Conclusions

Good quality pure and doped ZnO (Al and Sb) films are successfully made by spray pyrolysis method. The weight percent of $Sb(NO_3)_3$ and Al_2O_3 was varied from 0.5-1.5% and 2-10% respectively. Band gaps obtained by optical characterization for all films are in a good agreement with the standard ZnO thin film. The band gap of ZnO films are reducing by doping at some level. 1.2 wt% Sb:ZnO has lowest band gap among all prepared Sb:ZnO films and the band gap of 8 wt% Al:ZnO is lowest among all prepared Al: ZnO films. All prepared ZnO films are consequently applied in perovskite solar cell device preparation. Additionally, solar cell device is made with low cost CuI (as hole transport material) and carbon (as counter electrode). The solar cell efficiency was measured by solar simulator connected with Kiethly source meter under one sun illumination. The power conversion efficiency (PCE) obtained for 8% Al-doped ZnO is maximum (3.47%) among all devices.

5. Acknowledgements

The authors acknowledge S. V. National Institute of Technology, Surat-395007, Gujarat, India for rendering analytical services for this work.

6. References

[i] Aoun, Y., Benhaoua, B., Benramache, S. and Gasmi, B, "Effect of annealing temperature on structural, optical and electrical properties of zinc oxide (ZnO) thin films deposited by spray pyrolysis technique", Optik, 126, 2015, pp. 5407–5411.

[ii] Bedia, A., Bedia, F. Z., Aillerie, M., Maloufi, N., and Benyoucef, B., "Morphological and Optical properties of ZnO thin films prepared by spray pyrolysis on glass substrates at various temperatures for integration in solar cell", Energy procedia, 74, 2015, pp. 529 – 538.

[iii] Lehraki, N., Aida, M. S., Abed, S., Attaf, N., Attaf, A, and Poulain, M., "ZnO thin films deposition by spray pyrolysis: Influence of precursor solution Properties", current applied physics, 12, 2012, pp. 1283-1287.

[iv] Lanjewar, M., and Gohel, J.V., “Enhanced performance of Ag-doped ZnO and pure ZnO thin films DSSCs prepared by sol-gel spin coating”, Inorg. Nano-Met. Chem. 47, 2017, 1090–1096.

[v] Gohel, J.V., Jana, A.K., and Singh, M., “Highly enhanced photocurrent of novel quantum-dot-co-sensitized PbS–Hg/CdS/Cu:ZnO thin films for photoelectrochemical applications”, Appl. Phys. A 123. 2017, pp-1-12.

[vi] Taabouche, A., Bouabellou, A., Kermiche, F., Hanini, F., Sedrati, C., Bouachiba, Y., and Benazzouz, C., “Preparation and characterization of Al-doped ZnO piezoelectric thin films grown by pulsed laser deposition” Ceramics International 42, 2016, pp- 6701–6706

[vii] Briscoe, J., Diego, E.G., and Dunn, S., “In situ antimony doping of solution-grown ZnO nanorods” Chem. Comm., 10, 2009, pp-1273-1275

[viii] Kumari, N., Gohel, J.V., and Patel, S.R., “Multi-response optimization of ZnO thin films using Grey-Taguchi technique and development of a model using ANN”, Optik 144, 2017, pp- 422–435

[ix] Kojima, A., Teshima, K., Shirai, Y., and Miyasaka, T., “Organometal Halide Perovskites as Visible-Light Sensitizers for Photovoltaic Cells”, J. Am. Chem. Soc. 131, 2009, pp-6050–6051.

[x] Feng, L.U.J., Jie, B.A.I. Xiao, L.I., Kun, C.A.O., and Ming, W.A.N.G., “Alternate redox electrolytes in dye-sensitized solar cells”, Chin. Sci. Bull. 57, 2012, pp- 4131-4142.

[xi] Michael, M. L., Joel, T., Miyasaka, T., Takurou, N. M., and Henry, J. S., “Efficient Hybrid Solar Cells Based on Meso-Superstructured Organometal Halide Perovskites”, Science report, 338, 2012, pp-643-647.

[xii] Batniji, A., Monzir, S. A.L., Taher, M. E.A, Sofyan, A. T., and Hatem, G., “Dyes extracted from Trigonella seeds as photosensitizers for dye-sensitized solar cells”, J. Theor. Appl. Phys. 10, 2016, pp- 265-270.

[xiii] Reza, H. and Mohammad A., “Improving optical absorptivity of natural dyes for fabrication of efficient dye-sensitized solar cells”, J. Theor. Appl. Phys., 7, 2013, pp-1-7

[xiv] Gokilamani, N., Kumarasamy, M.N., Thambidurai, M., Ranjitha, A., Dhayalan, V., Senthil, T.S., and Prabhu, R., “Dye-sensitized solar cells with natural dyes extracted from rose petals”, J Mater Sci: Mater. Electron., 24, 2013, pp-3394–3402.

[xv] Zhu, G., Shen, Y., Xu, K., Huangfu, M., Cao, M., Gu, F., Wang, L.: Preparation of ZnO electron transport layers by spray technology for perovskite solar cells. J. Alloys Compd. 689, 2016, pp-192–198.