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Spray Pyrolysed Cu_2ZnSnS_4 Solar Cell Using Cadmium Free Buffer Layer

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Abstract Cu_2ZnSnS_4 (CZTS) thin film solar cells were fabricated using CZTS as absorber layer and In_2S_3 as buffer layer. Both of these layers were prepared by using chemical spray pyrolysis (CSP) technique. Since the resistance of the In_2S_3 buffer layer plays an important role in the performance parameters of the cell we have done excitu doping of In_2S_3 by using indium. For an optimum doping, the cell exhibited an open circuit voltage of 430 mV, a short circuit current density of 8.02 mA/cm^2, a fill factor of 45% and a conversion efficiency of 1.5%. We avoided the usual cyanide etching and CdS buffer layer, both toxic, for the fabrication of the cell.

Keywords: Semiconductors, Spray coating techniques, Doping, solar cell
PACS: 68.55.ag, 81.15.Rs, 68.55.Ln, 88.40.H-

INTRODUCTION

The aim to develop thin film solar cells using eco-friendly, abundant and cost effective materials is satisfied by fabricating the cells with Cu_2ZnSnS_4 (CZTS) as the absorber. With an absorption coefficient of 10^4 cm^{-1} and band gap close to 1.45 eV, the quaternary compound contains neither rare metals nor toxic materials. Combining CZTS thin film with Cd-free buffer layer, we can develop next generation solar *cells.* Ito and Nakazawa reported, for the first time, the photovoltaic effect in the heterodiode that consists of cadmium-tin-oxide transparent conducting film and CZTS thin film on a stainless steel substrate. They reported an open circuit voltage of 165mV [1]. Katagiri et al. have reported an efficiency of 6.77% on the cell having structure Al/ZnO:Al/CdS/CZTS/Mo/SLG substrate [2]. Teodor K. Todorov et al. reported the highest conversion efficiency of 9.66% in mixed sulfoselenide [Cu2ZnSn(S,Se)$_4$] device[3]. Nowadays lot of works is going on for the effective replacement of CdS. The motivation behind this is not only to eliminate toxic cadmium but also to improve light transmission in the blue wavelength region by using a material having band gap wider than that of CdS. Our aim is to develop a Cu_2ZnSnS_4-based solar cell, free from Cd and prepared using a simple technique. In the present paper we report the fabrication and characterization of Cu_2ZnSnS_4/In_2S_3 solar cell deposited using chemical spray pyrolysis (CSP) technique.

EXPERIMENTAL

We used automated CSP machine for depositing both absorber and buffer layers. Glass plates coated with ITO were used as the substrate. Aqueous solution containing cuprous chloride (0.01875 M), zinc acetate (0.0125 M), stannic chloride (0.0125 M) and thiourea (0.1 M) was sprayed at a rate of 8ml/min onto the substrate kept at 623 K using compressed air (pressure~1.5 bar) as the carrier gas. Concentration of thiourea was 2 times the stoichiometric requirement (0.05 M) to compensate for the loss of sulphur during pyrolysis. Indium chloride (0.025 M) and thiourea (0.1 M) were used as precursor solutions for depositing In_2S_3 layer over this. Over the indium sulfide layer, some area is doped with indium to reduce the resistance of the top of the buffer layer. Doping was achieved by allowing an optimum quantity of vacuum evaporated indium (9mg) to diffuse in to the film by thermal annealing [125^{0}C for 2 hours]. Ag electrodes having an area of 0.1cm^2 were given over the indium sulphide layer on the doped as well as undoped regions by vacuum evaporation.

RESULTS AND DISCUSSION

At first we prepared a CZTS/In_2S_3 bilayer on ITO coated glass substrate. Silver (Ag) was deposited over this using physical vapour deposition to act as the top electrode. ITO was the bottom electrode. The cell

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parameters obtained were V_{oc}= 0.4 V, J_{sc}= 3.7 mA/cm^2, FF = 26.2 %, η= 0.38 %. The series resistance (R_s) and shunt resistance (R_{sh}) were found to be 95.51 Ω/cm^2 and 115.34 Ω/cm^2 respectively (Rs and R_{sh} were the effective values estimated from the slopes of the J-V curve at the vicinity of the intersections of the J-V curve with V and J axis[4]). The high series resistance might be the reason for low value of J_{sc} and FF and hence the efficiency of the device. High series resistance of the heterojunction is caused by the high resistance of In_2S_3 buffer layer. As we know, in a heterojunction, the resistance near the junction must be high in order to reduce the minority carrier recombination while the resistance should be low near the surface in order to improve the collection of the light generated carriers.

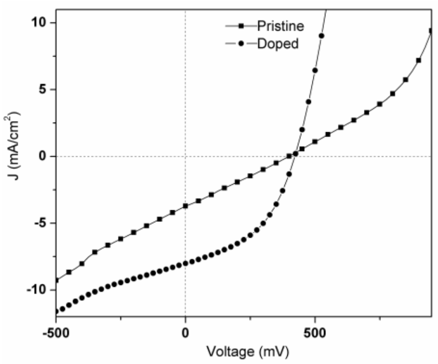

Figure 1. J-V curves under illumination, for the pristine cell and the cell with doped buffer layer

Hence to reduce the resistance near the surface, we doped In_2S_3 with an optimum quantity of indium as described earlier. Then Ag electrodes were deposited over the doped buffer layer.

The J-V curves under illumination, for the pristine cell and the cell with doped buffer layer are shown in Figure 1. The cell parameters obtained were V_{oc}= 0.43 V, J_{sc}= 8.02 mA/cm^2, FF = 44.3 %, η= 1.5 %, R_s= 15 Ω/cm^2 and R_{sh}= 178 Ω/cm^2. Improvement in cell parameters is due to the significant reduction in series resistance caused by the doping of In_2S_3 buffer layer with In. Again the resistivity measurements on In_2S_3 layer deposited on glass substrates before and after doping with indium proved that the resistivity decreased from 2.3×10^5 Ωcm to 4.7 Ωcm after doping. Hence the doping decreased the resistance of buffer layer thereby increasing the collection of light generated carriers. This is the reason for increase in short circuit current density.

CONCLUSION

We have successfully fabricated a bilayer heterojunction photovoltaic device using automated spray machine. Here Cu_2ZnSnS_4 was absorber layer and In_2S_3 was the buffer layer. Using Ag as top electrode J-V characteristics of the cell was recorded. The performance parameters were not so good. By doping the In_2S_3 buffer layer with indium, we could enhance the performance parameters. For the optimum doped cell, the cell parameters obtained were V_{oc}= 0.43 V, J_{sc}= 8.02 mA/cm^2, FF = 44.3 %, η= 1.5 %, R_s= 15 Ω/cm^2 and R_{sh}= 178 Ω/cm^2.

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