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Compositional Influence in the Photovoltaic Properties of Dual Ion Beam Sputtered $Cu_2ZnSn(S,Se)_4$ Thin Films

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Abstract : The optimal band gap (~ 1 to 1.5 eV) and high absorption coefficient ~104 cm^{-1} has made $Cu_2ZnSn(S,Se)_4$ (CZTSSe) films as one of the most promising absorber materials in thin-film photovoltaics. Additionally, CZTSSe consists of elements that are abundant and non-toxic, makes it even more favourable. The CZTSSe thin films are grown at 100 to 500°C substrate temperature (Tsub) on Soda lime glass (SLG) substrate by Elettrorava dual ion beam sputtering (DIBS) system by utilizing a target at $2.43x10^{-4}$ mbar working pressure with RF power of 45 W in argon ambient. The chemical composition, depth profiling, structural properties and optical properties of these CZTSSe thin films prepared on SLG were examined by energy dispersive X-ray spectroscopy (EDX, Oxford Instruments), Hiden secondary ion mass spectroscopy (SIMS) workstation with oxygen ion gun of energy up to 5 keV, X-ray diffraction (XRD) (Rigaku Cu Kα radiation, λ=.154nm) and Spectroscopic Ellipsometry (SE, M-2000D from J. A. Woollam Co., Inc). It is observed that from that, the thin films deposited at Tsub=200 and 300°C show Cu-poor and Zn-rich states (i.e., Cu/(Zn + Sn) < 1 and Zn/Sn > 1), which is not the case for films grown at other Tsub. It has been reported that the CZTSSe thin films with the highest efficiency are typically at Cu-poor and Zn-rich states. The values of band gap in the fundamental absorption region of CZTSSe are found to be in the range of 1.23-1.70 eV depending upon the Cu/(Zn+Sn) ratio. It is also observed that there is a decline in optical band gap with the increase in Cu/(Zn+Sn) ratio (evaluated from EDX measurement). Cu-poor films are found to have higher optical band gap than Cu-rich films. The decrease in the band gap with the increase in Cu content in case of CZTSSe films may be attributed to changes in the extent of p-d hybridization between Cu d-levels and (S, Se) p-levels. CZTSSe thin films with Cu/(Zn+Sn) ratio in the range 0.86–1.5 have been successfully deposited using DIBS. Optical band gap of the films is found to vary from 1.23 to 1.70 eV based on Cu/(Zn+Sn) ratio. CZTSe films with Cu/ (Zn+Sn) ratio of .86 are found to have optical band gap close to the ideal band gap (1.49 eV) for highest theoretical conversion efficiency. Thus by tailoring the value of Cu/(Zn+Sn), CZTSSe thin films with the desired band gap could be obtained. Acknowledgment: We are thankful to DIBS, EDX, and XRD facility equipped at Sophisticated Instrument Centre (SIC) at IIT Indore. The authors B. S. S and A. K. acknowledge CSIR, and V. G. acknowledges UGC, India for their fellowships. B. S. S is thankful to DST and IUSSTF for BASE Internship Award. Prof. Shaibal Mukherjee is thankful to DST and IUSSTF for BASE Fellowship and MEITY YFRF award. This work is partially supported by DAE BRNS, DST CERI, and DST-RFBR Project under India-Russia Programme of Cooperation in Science and Technology. We are thankful to Mukul Gupta for SIMS facility equipped at UGC-DAE Indore.

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