DEVELOPMENT OF Sb_2Se_3 AND Sb_2S_3 SOLAR CELLS BY CLOSE-SPACED SUBLIMATION

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Solar photovoltaics (PV) are already one of the cheapest sources of electricity and will play a crucial role in global energy transition to carbon-free electricity production. Due to increasing demand for electricity, however, research in new generation PV technologies are being researched for applications in multi-junction tandem cells, solar windows, building-integrated PV, and internet-of-things (IoT) devices. Emerging photovoltaic absorber materials, such as binary antimony chalcogenides – Sb_2Se_3, Sb_2S_3, and $Sb_2(S,Se)_3$ – have shown promise due to their favorable optoelectronic properties, low processing temperatures, and earth-abundant constituents. This work focuses primarily on fabricating Sb_2Se_3 and Sb_2S_3 solar cells by close-spaced sublimation (CSS) and characterizing the defects in Sb_2Se_3 and Sb_2S_3 devices using admittance spectroscopy (AS) and photoluminescence (PL) techniques. The CSS deposition approach, comprising optimal processing conditions, seed screening, and suitable buffer layers, is demonstrated for growing high-quality Sb_2Se_3 and Sb_2S_3 thin films. PV device performance was directly related to the grain morphology and microstructure of Sb_2Se_3 and Sb_2S_3 films. Optimal film structures had a compact large-grain morphology with increased texture along the [001] crystal direction. Advanced pole figures and electron backscatter diffraction (EBSD) maps were measured for Sb_2Se_3 films to improve accuracy of determining grain orientation in polycrystalline Sb-chalcogenide thin films, in addition to the common texture coefficient (TC) analysis. Ultimately, Sb_2Se_3/TiO_2 and Sb_2S_3/CdS devices in superstrate configuration achieved efficiencies of 5.7% and 3.8%, respectively. Nevertheless, high defect densities severly limit performance of Sb-chalcogenide PV devices. AS and PL techniques were employed to characterize the best Sb_2Se_3/TiO_2 and Sb_2S_3/CdS devices to increase experimental knowledge of defect present within these absorber films. For both Sb_2Se_3 and Sb_2S_3, AS study revealed deep defect states with activation energies >0.3 eV, while PL found bands related to donor-acceptor-pair (DAP) recombination. By developing the comprehensive deposition strategy and improving knowledge of performance-limiting defects in Sb_2Se_3 and Sb_2S_3 absorbers, this work helped to lay the groundwork for future development of $Sb_2(S,Se)_3$ solar cells by CSS.

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