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Effect of Cu content in $CuSbS_2$ thin films using hybrid inks: Their photovoltaic properties and defect characteristics

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ABSTRACT

$CuSbS_2$ (CAS) thin films were grown with different Cu/Sb precursor ratios using a non-vacuum-based hybrid ink method. The prepared precursor inks were coated on Mo-coated soda lime glass and a subsequent sulfurization process was carried out. X-ray diffraction (XRD) patterns confirmed the presence of the $CuSbS_2$ phase for an optimal Cu/Sb precursor ratio; for other ratios, additional secondary phases were also observed. Then, $CuSbS_2$ solar cells were fabricated to compare the photovoltaic performances. The sample with the pure $CuSbS_2$ phase showed better photovoltaic properties than those films containing secondary phases. The best $CuSbS_2$ solar cell with optimum Cu/Sb ratio exhibited the conversion efficiency of 2.75%. The temperature-dependent light and dark current density–voltage (JV-T) characteristics were measured to investigate the recombination process, and R–J curves were extracted to study the photovoltaic characteristics, including parasitic resistances. The depletion width and carrier concentrations were also compared from capacitance–voltage (C-V) measurements of the solar cells prepared with the films of various Cu/Sb ratios. Using admittance spectroscopy (AS), the dominant defect energy level was deduced and determined as 0.23 eV above the valence band maximum. Drive-level capacitance profiling (DLCP) was applied to characterize the defect behavior of the device with optimum Cu content.

1. Introduction

The development of cost-effective photovoltaic solar cells requires inexpensive and highly efficient absorber materials. Inorganic chalcogenide-based semiconductor absorbers, especially $Cu(InGa)Se_2$ (CIGS) and CdTe, have received immense attention for this application due to their excellent photoelectrical properties. The highest power conversion efficiencies achieved for CIGS and CdTe thin film solar cells were 22.6% [1] and 22.1% [2], respectively. However, the elemental scarcity and relatively high price of In and Ga in CIGS and the toxicity of Cd in CdTe limit their widespread use. This has motivated the development of other earth-abundant, low-cost, and low-toxicity absorber materials. For example, the quaternary kesterite compound $Cu_2ZnSn(SSe)_4$ (CZTSSe) meets these requirements and has been demonstrated as a promising alternative absorber in terms of the power conversion efficiency, of which a maximum of 12.6% [3] has been reported to date. However, the need to precisely control the cation stoichiometry and its complex structural polymorphism [4] are challenges for further development.

To overcome the aforementioned hurdles, earth-abundant and low-toxicity constituent materials with relatively simple structures, such as $CuSbS_2$ [5,6], Fe_2S [7,8], Sb_2S_3 [9], SnS [10,11], and Cu_2SnS_3 [12], have been investigated as future absorber materials. Among these, $CuSbS_2$ (CAS) has emerged as one of the most promising materials due to its suitable optoelectronic properties for thin-film solar cells. It has a favorable band gap of 1.38–1.65 eV [5,13,14] for photovoltaic applications and a strong absorption coefficient ($\alpha > 10^5$ cm^{-1}) in the visible region [14,15]. It possesses p-type conductivity due to dominant Cu vacancies [13,16], with a hole concentration of 10^{16}–10^{18} cm^{-3} [5,13,16] and a hole mobility of 0.1–1.0 cm^2 V^{-1} s^{-1} [14]. Moreover, $CuSbS_2$ is composed of earth-abundant and inexpensive elements and exhibits a high spectroscopic-limited maximum efficiency (SLME) of 22.9% [15].

Despite these favorable fundamental optoelectronic properties, $CuSbS_2$ photovoltaic (PV) devices currently show poor reproducibility and a relatively low efficiency. To date, the highest efficiency using this absorber material is only 3.22% [5]. Few studies of its optoelectronic

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