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Thin films of copper antimony sulfide: A photovoltaic absorber material

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ABSTRACT

In this work, we report preparation and characterization of $CuSbS_2$ thin films by heating glass/Sb_2S_3/Cu layers and their use as absorber material in photovoltaic structures: glass/SnO_2:F/*n*-CdS/*p*-$CuSbS_2$/C/Ag. The Sb_2S_3 thin films of 600 nm were prepared by chemical bath deposition on which copper thin films of 50 nm were thermally evaporated, and the glass/Sb_2S_3/Cu multilayers were heated in vacuum at different temperatures. X-ray diffraction analysis showed the formation of orthorhombic $CuSbS_2$ after heating the precursor layers. Studies on identification and chemical state of the elements were done using X-ray photoelectron spectroscopy. The optical band gap of the $CuSbS_2$ thin films was 1.55 eV and the thin films were photoconductive. The photovoltaic parameters of the devices using $CuSbS_2$ as absorber and CdS as window layer were evaluated from the *J–V* curves, yielding J_{sc}, V_{oc}, and FF values in the range of 0.52–3.20 mA/cm^2, 187–323 mV, and 0.27–0.48, respectively, under illumination of AM1.5 radiation.

1. Introduction

New easily available, less expensive, non-toxic thin film solar cell materials are to be developed to solve various issues which limit the cost and capacity of the current thin film solar cell technologies based on CdTe and CIGS. Copper antimony sulfide ($CuSbS_2$) is suggested as an alternative to $CuInS_2$ considering its non-toxicity, relative abundance, and nearly equal ionic radius of indium [1–6]. Due to their direct optical band gap of 1.5 eV, the optimum value for an absorber material in solar cells, this material can be one among the new photovoltaic materials [1–3,6]. Solar cell applications of this material were illustrated in photovoltaic structures: SnO_2:F–(*n*)CdS:In–Sb_2S_3–$CuSbS_2$–Ag, giving V_{oc} = 330 mV and J_{sc} = 0.3 mA/cm^2 [7] and TCO/denseTiO_2/$CuSbS_2$/graphite resulting V_{oc} = 90 mV and I_{sc} = 0.02 mA [8]. Thin films of $CuSbS_2$ were deposited by spray pyrolysis from an aqueous solution of copper chloride, antimony acetate and thiourea at substrate temperatures of 200–350 °C and using polymer additives of sodium maleate-methyl metacrylate (hydrophilic polymer, HFL) and sodium maleate-vinyl acetate (hydrophobic polymer, HFB) as complexing agents in the solution [8,9]. In thermally evaporated $CuSbS_2$ thin films, their band gap variation with substrate temperature as well as non-degenerate semiconducting behavior were reported [10]. A two stage process approach was described very effective for the preparation of polycrystalline ternary semiconductor thin films [11]. Photoactive $CuSbS_2$ thin films of optical band gap 1.2–1.5 eV were prepared *via* a two-stage process of thermal evaporation of Cu–Sb precursor thin films followed by sulfurization at temperatures 200–400 °C [2]. In a different approach of two stage process, thermal evaporation of copper on chemical bath deposited Sb_2S_3 thin films followed by a thermal treatment resulted in *p*-type $CuSbS_2$ thin films [3,7]. Further studies are needed to explore the full potential of this material for PV applications.

In the present work, our aim is to prepare and characterize $CuSbS_2$ thin films for their application as absorber material in PV structures. Moreover, we fabricated PV structures using chemical bath deposited CdS thin films as window layers. $CuSbS_2$ thin films of 600 nm in thickness were prepared by heating glass/Sb_2S_3/Cu

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layered structure in low vacuum. The thin films formed at different heating conditions were characterized using X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), atomic force microscopy (AFM), UV–vis spectrophotometry and electrical measurements. Photovoltaic devices of structure: glass/SnO_2:F/CdS/$CuSbS_2$/C/Ag were prepared at various conditions. Analysis of *J–V* characteristics of the PV structures showed V_{oc} from 187 to 323 mV and J_{sc} 1.52 to 3.77 mA/cm^2 under illumination by AM1.5 radiation from a solar simulator.

2. Experimental

2.1. Preparation of $CuSbS_2$ thin films

$CuSbS_2$ thin films were prepared by heating layered structures of Sb_2S_3/Cu deposited on glass substrates [3]. First, antimony sulfide thin films were chemically deposited on glass substrates from a bath containing 650 mg of $SbCl_3$ dissolved in 2.5 ml of acetone, 25 ml of $Na_2S_2O_3$ (1 M), and 72.5 ml of distilled water (40 °C). The solution was stirred well and the substrates were

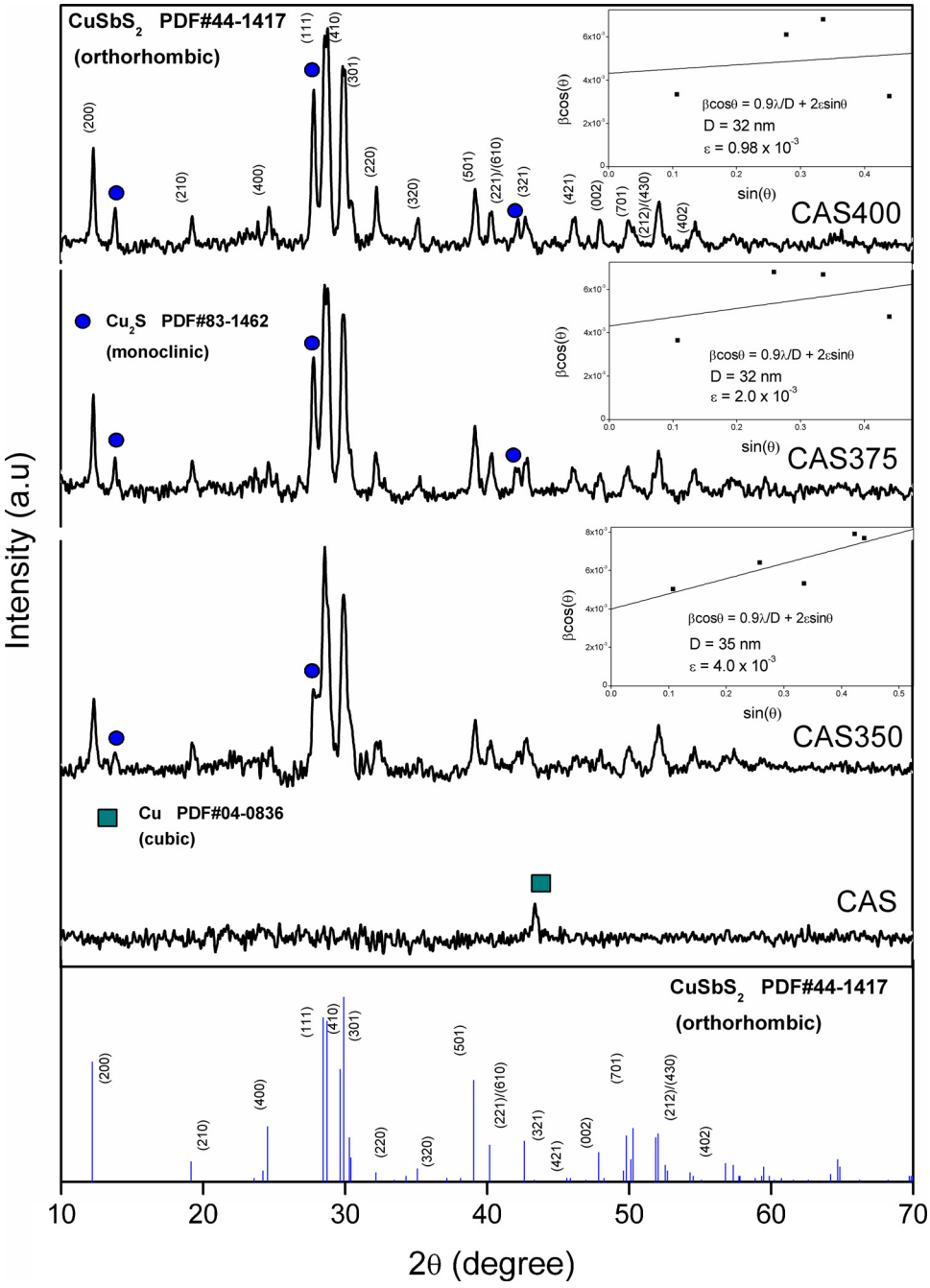

Fig. 1. XRD patterns for as-deposited glass/Sb_2S_3/Cu (CAS) and multilayers glass/Sb_2S_3/Cu annealed at 350 °C, 375 °C and 400 °C (CAS350, CAS375, CAS400) in low vacuum (10^{-3} Torr) 1 h. Standard patterns corresponding to orthorhombic $CuSbS_2$ are also included.