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Biren Patel, Manmohan Singh Waldiya, Abhijit Ray



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Highly phase-pure spray-pyrolysed Cu_2SnS_3 thin films prepared by hybrid thermal treatment for photovoltaic applications

Biren Patel, ManmohansinghWaldiya and Abhijit Ray *

Department of Solar Energy & Solar Research and Development Center, Pandit Deendayal Petroleum University, Raisan, Gandhinagar, Gujarat 382-007, India

* E-mail: abhijit.ray1974@gmail.com; Fax: +91-079-23275030

Abstract

The structure and physical properties of thin films of ternary photovoltaic absorber Cu_2SnS_3 on soda-lime glass substrate deposited by cost effective spray pyrolysis following a hybrid thermal annealing (HTA) have been investigated. The advantage of HTA-sulfurization over standard thermal annealing (STA) and rapid thermal annealing (RTA) is established for Cu_2SnS_3 (CTS). The X-Ray diffraction (XRD) and Raman spectroscopy reveals highly phase pure Cu_2SnS_3 is produced through effective relaxation of compressive stress and increased crystallinity by the HTA-sulfurization step. Additionally, scanning electron microscopy (FE-SEM) and energy dispersive x-ray spectroscopy (EDX) reveals that the evolution of secondary phases, Cu_xS_y (x, y : 116, 64; 2, 1; 1.8, 1; 51, 27; 6, 6 etc) could be effectively prevented by the HTA process over the STA and RTA. The optimized HTA-CTS thin films obtained are of p -type conductivity with the carrier concentration, resistivity and mobility of $5.5 \times 10^{20} \text{ cm}^{-3}$, $9.8 \times 10^3 \Omega\text{-cm}$ and $1.15 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, respectively. The optical band gap of about 1.65 eV with large absorption coefficient of 10^5 cm^{-1} demonstrates it as an ideal candidate for thin film solar cells. A solar cell configuration of glass/F:SnO₂/CTS/CdS/ZnO:Al/Al shows an open circuit voltage of 225 mV.

Keywords: CTS; thin film; spray pyrolysis; rapid thermal annealing

1. Introduction

A number of copper chalcogenide based compound semiconductors, that includes ternary $\text{CuIn}(\text{S}/\text{Se})_2$ (namely, CIS) [1,2], Cu_2SnS_3 or Cu_3SnS_4 (namely, CTS) [3] as well as quaternary $\text{Cu}(\text{In},\text{Ga})(\text{S}/\text{Se})_2$ (namely, CIGS) and $\text{Cu}_2\text{ZnSn}(\text{S}/\text{Se})_4$ (namely, CZTS) [4] has been of great interest globally for last two decades for their application in thin film solar cells and modules. Several groups have demonstrated that their efficiencies are in great pace of improvement towards of that of the commercial Silicon technologies [5-7] and these compounds have a great potential for scaling up owing to their easy process-ability through solution techniques [8-10]. While the expensive CIS and subtle phase pure quaternary CZTS have been widely studied, the development of less expensive and ternary CTS is at the nascent stage. From the chalcogenide class, CTS is one of the ternary semiconductor compounds which contain all earth abundant and non-toxic elements available on the earth crust. Tetragonal and cubic Cu_2SnS_3 has been

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