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Low-cost hydrothermal synthesis and characterization of pentanary $\text{Cu}_2\text{Zn}_x\text{Ni}_{1-x}\text{SnS}_4$ nanoparticle inks for thin film solar cell applications



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ABSTRACT

In the present investigation, $\text{Cu}_2(\text{Zn}_x\text{Ni}_{1-x})\text{SnS}_4$ nanoparticles were successfully synthesized by facile and low-cost hydrothermal method for different concentration of x=1, 0.75, 0.50, 0.25, 0 at 190 °C for 24 h. The probable reaction mechanism for the formation of $\text{Cu}_2\text{Zn}_x\text{Ni}_{1-x}\text{SnS}_4$ nanoparticles is reported. The synthesized nanoparticles were characterized by x-ray diffraction (XRD), Raman spectroscopy, field emission scanning electron microscope (FESEM), transmission electron microscope (TEM), x-ray photoelectron spectroscopy (XPS) and ultra violet-visible (UV-vis) spectrometric studies. The phase pure kesterite and stannite structures of synthesized nanoparticles were confirmed by XRD and Raman spectroscopy analysis. The structural transition from tetragonal kesterite copper zinc tin sulfide (Cu₂ZnSnS₄ - CZTS) to stannite Cu₂Zn_xNi_{1-x}SnS₄ (x=0) occurs in the Cu₂Zn_xNi_{1-x}SnS₄ samples with nickel concentration (x) in the range of 0.25–1. The results of SEM and TEM show the morphological features of the prepared Cu₂Zn_xNi_{1-x}SnS₄ nanoparticles were reduced from 1.63 eV (x=1) to 1.36 eV (x=0). It falls well within the suitable band gap range of absorber materials to be useful for thin film solar cell applications. Photoresponse behavior of Cu₂Zn_xNi_{1-x}SnS₄ (x=1 and 0.75) thin films under dark and light illumination indicates the potential of Cu₂Zn_xNi_{1-x}SnS₄ nanoparticles as an alternate absorber material in low-cost thin film solar cells applications.

1. Introduction

Thin film photovoltaic (PV) technology attracts the attention of many researchers because it offers large area thin-film solar cell (TFSC) fabrication at low-cost compared to other solar cell fabrication technology. TFSC based on chalcogenide compounds is a developing alternative field to high-cost crystalline and multi-crystalline silicon (Si) solar cell technologies [1]. The chalcogenide compounds are present in the form of a binary, ternary and quaternary combination of metals with their corresponding sulphides, selenides or tellurides [2]. The key advantage of these chalcogenide materials are excellent light absorbing properties and low thicknesses of absorber layer leads to saving materials and also reducing the environmental impact. In order to construct a TFSC, selection of a suitable absorber layer is an important criterion. It plays a vital role in of the primary absorption of light photons from sunlight. Absorber materials should exhibit a direct

band gap well matched to the solar spectrum along with other favorable electronic properties [3]. Copper indium gallium sulfide or selenide (CIGSSe) and cadmium telluride (CdTe) are two important absorber materials which are currently used in large scale TFSC fabrication [4-6]. The main drawbacks of these materials are high toxicity, environmental pollution and scarcity of materials (In, Ga) on earth [7]. So, the worldwide extensive research focused on discovering alternative absorber materials for TFSC application that are low-cost, abundant and environment-friendly [8]. Quaternary chalcogenide material CZTS satisfies the criteria and its derivatives were used as a promising absorber material for TFSC applications [9]. It has a high absorption coefficient ($\alpha > 10^4 \text{ cm}^{-1}$), optimum optical band gap values (~ 1.5 – 1.6 eV), environmental friendly compared to CIGS and CdTe, potentially low-cost precursors, and Earth-abundant elements [10,11]. CZTS is analogous with CIGSSe and has the same crystal structure and optical absorption coefficient. According to Shockley - Queisser photon

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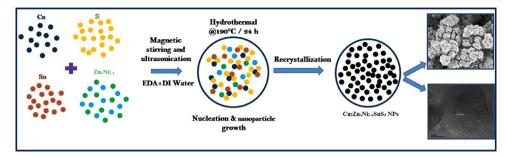


Fig. 1. Schematic representation of Cu₂Zn_xNi_{1-x}SnS₄ nanoparticles nucleation and growth.

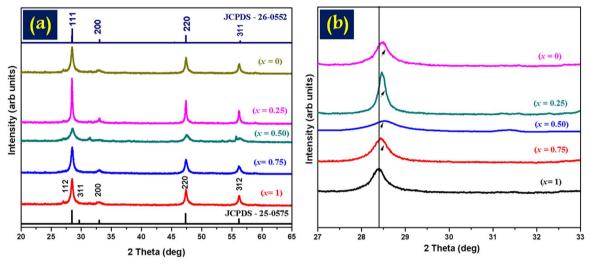


Fig. 2. (a). X-ray diffraction patterns of the $Cu_2Zn_xNi_{1-x}SnS_4$ (x=1,0.75,0.50,0.25,0) nanoparticles, (b) expanded (112) peaks. Relevant standard JCPDS patterns are also included for comparison.

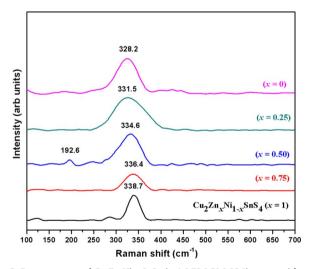


Fig. 3. Raman spectra of $\rm Cu_2Zn_xNi_{1-x}SnS_4$ (x=1,0.75,0.50,0.25,0) nanoparticles prepared by hydrothermal method at 190 °C for 24 h.

balance calculations, the theoretical conversion efficiency of CZTS based solar cell is 32.2% which represent an improvement in power conversion efficiency (PCE) of more than 12.6% over toxic hydrazine-based CZTSSe thin films fabricated by vacuum based method [12,13]. Based on material aspects, CZTS is available in nature in three mineral structures, namely kesterite, stannite, and wurtzite [14–16]. The kesterite structure is preferred over other structures for TFSC applications because it offers high thermal stability, high electron affinity and tunable direct band gap suitable for solar spectrum [17,18]. There have been various vacuum and non – vacuum based techniques adopted to

fabricate CZTS/CZTSSe thin films, such as evaporation, sputtering, chemical vapor deposition, physical vapor deposition, electrodeposition and nanoparticle based solution-film cast method [19–23]. Among the all fabrication methods, thin film fabrication by nanoparticle based solution casting is the easiest way to prepare low-cost films on a large scale [24]. The nanoparticle ink can be dip coated, spin coated or roll-to-roll deposited method on conductive glass substrates.

CZTS nanoparticles were synthesized by different methods such as, sol-gel, microwave assisted colloidal synthesis, hydrothermal, solvothermal and hot-injection methods are reported in the literature [25-28]. The hydrothermal synthesis has been a useful technique to synthesize high-quality nanomaterials under various pressures and temperatures. In the case of CZTS, this hydrothermal method offers better crystallinity and phase pure CZTS nanoparticles without sulfurization [29,30]. In hydrothermal synthesis, the role of solvents and surfactants are very important, which actively take part to induce and impact the crystalline nature and morphological property of the CZTS nanoparticles. This highly influences the PCE of thin film photovoltaic devices [31]. Phase pure CZTS nanoparticles have been obtained by aqueous and non-aqueous solvent route methods. There have been a limited number of reports on non - aqueous solvent based hydrothermal method for synthesis CZTS nanoparticles [32,33]. Some of the solvents used for CZTS nanoparticles are polyethylene glycol (PEG)+de-ionized (DI) water, polyvinylpyrrolidone (PVP)+DI water, 2-methoxyethanol+DI water, monoethanolamine (MEA)+DI water and ethylenediamine (EDA)+DI water, etc [34]. Among this, EDA+DI mixed solvent system is more efficient for hydrothermal synthesis of CZTS nanoparticles. EDA is a strong chelating agent and it forms metal - chelate complexes when they react with the precursor materials. These metal - chelate complexes can easily reduce and separate it into their metal complexes, after the treatment at high-temperature hydro-

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