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Chemical Spray Deposited Nickel Sulphide Thin Films for Supercapacitor applications

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ABSTRACT
Pure kesterite phase thin films of Cu_2ZnSnS_4 (CZTS) were synthesized at different substrate temperatures using sulphate precursors by spray pyrolysis method. The significance of synthesis temperature on the structural, morphological and optical properties has been studied. The X-ray analysis assured that synthesized CZTS thin films showing pure kesterite phase. The value of crystallite size was found maximum at the substrate temperature 400 °C. At the same temperature, microstructural properties such as dislocation density, microstrain and stacking fault probability were found minimum. The morphological examination designates the development of porous and uniform CZTS thin films. The synthesized CZTS thin films illustrate excellent optical absorption (10^5 cm^{-1}) in the visible band and the optical band gap varies in the range of 1.489 eV to 1.499 eV.

Key words: XRD, NiS, CZTS thin film,

INTRODUCTION

Nickel sulfide (NiS) thin films have large applicability in solar selective coatings [1], photoconductors [2], antireflection coatings, polarizers, narrow band filters and supercapacitor etc. absorber layer of the material Cu_2Zn SnS_4 (CZTS) is incredibly promising because, it has a direct band gap energy (1.5 eV), an absorption coefficient ($>10^{-4}$ cm^{-1}) and their optoelectronic characteristics for the compact layered solar cells. And also, the materials used for preparing Cu_2ZnSnS_4 (CZTS) are low-toxic, earth-abundant and low-cost. Cu_2ZnSnS_4 (CZTS) is the great alternative to the CIGS thin film which contains expensive and scarce elements such as indium and gallium.

Also they are harmful to the human body and environment. The highest reported conversion efficiency of CZTS solar cells is ≈12.7% [1], which is still far from the commercialization range. Main reason is their structural complication. Hence CZTS contains more defects and impurities in the structural phase (like secondary phases CuS, ZnS, etc.). And these defects and impurities are dynamic in application of semiconductors in solar cells, successively they directly affect the generation, separation, and recombination of electron-hole pairs [2]. It is important to deposit the pure phase with less defective materials thin films for the efficient solar cells.

Several physical and chemical methods are used today to deposit CZTS thin films, in that electrochemical, co-sputtering, thermal co-evaporation, spin coating, spray pyrolysis, etc. shows good results. The simplicity and forthrightness of the spray pyrolysis technique makes it very smart for large scale industrial applications and grants many cheap advantages compared to other methods [3]. In the present article, the effect of the substrate temperature on the structural-microstructural, morphological and optical properties of the CZTS thin films deposited using sulphate precursors are studied.

METHODOLOGY

An aqueous solution contains cupric sulphate (0.025 M), zinc sulphate (0.025 M), stannous sulphate (0.025 M) and thiourea (0.20 M) used to deposit Cu_2ZnSnS_4 (CZTS) thin films by spray pyrolysis. To recompense the loss of sulfur in the course of spray deposition, excess amount of thiourea is taken. All the Cu_2ZnSnS_4 (CZTS) thin films were deposited at the substrate temperatures of 350, 375, 400 and 425 °C.

Structural and crystallographic properties of CZTS films were studied by x-ray diffractometer (XRD Bruker AXS D8 Advance Model) with the radiation source Cu-Kα in the range 10° to 80°. (FE-SEM SU8000 Hitachi) field emission scanning electron microscope was used to determine surface morphology. The optical parameters in the wavelength 400-900 nm were studied at room temperature through Uv-Visible spectroscopy (Perkin Elemer Lambda 1050 Uv-Vis-NIR Spectrophotometer).

RESULT AND DISCUSSION

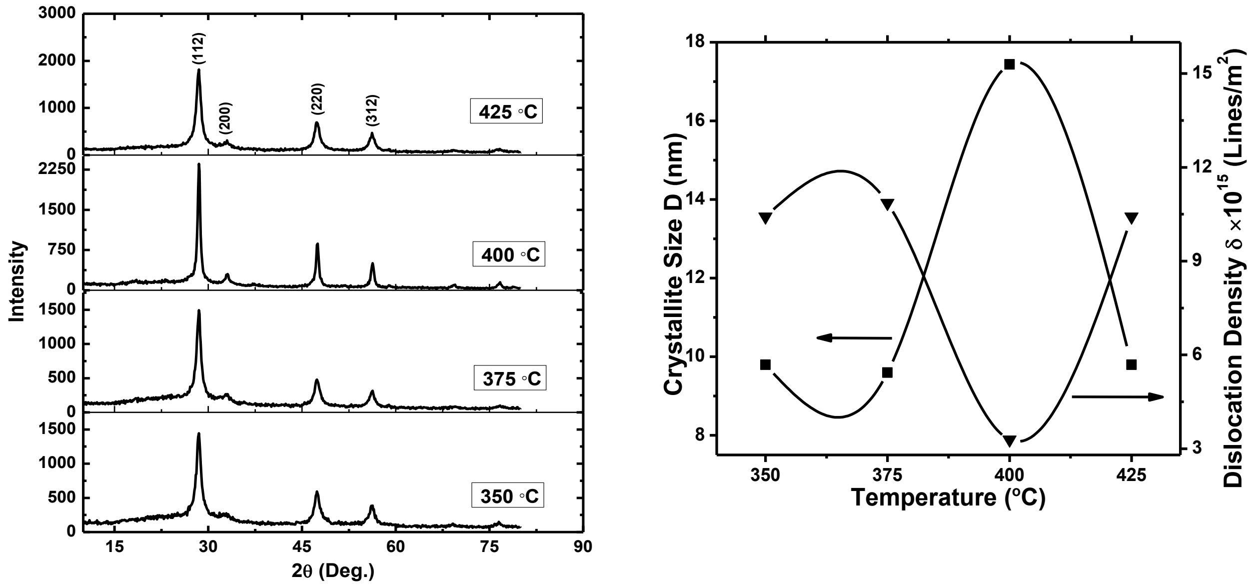
X-ray Diffraction analysis

The Fig. 1(a) gives the XRD patterns of the CZTS thin films using sulphate precursors onto glass substrates by spray pyrolysis deposition. The peaks can be accredited to (112), (200), (220) and (312) planes of kesterite CZTS structure observed at 2θ values of 28.5°, 33.09°, 47.31° and 56.23° respectively [4]. Formation of CZTS phase indicated by strong preferential orientation in (112) direction [5]. Over the range of substrate temperature (350 to 425 °C), no any impurity peak is observed. From Fig. 1(a), it is observed that, as substrate temperature increases from 350 °C to 400 °C, the peak intensity improved and simultaneously full width at half maximum (FWHM) values decreased attributing to the increase in crystallite size (D) and the decrease in dislocation density (δ) [6], shown in Fig. 1(b).

Also the microstructural properties are calculated; both the values of micro-strain and stacking fault probability are decreased at the substrate temperature 400 °C. This is due to the crystal size increment, hence the structure purity and perfections in the growth. As we compare the present data with the previous reported for the samples prepared by chloride precursors [7], sharpness of the peaks and the crystallite size (D) are enhanced by using the sulphate precursors.

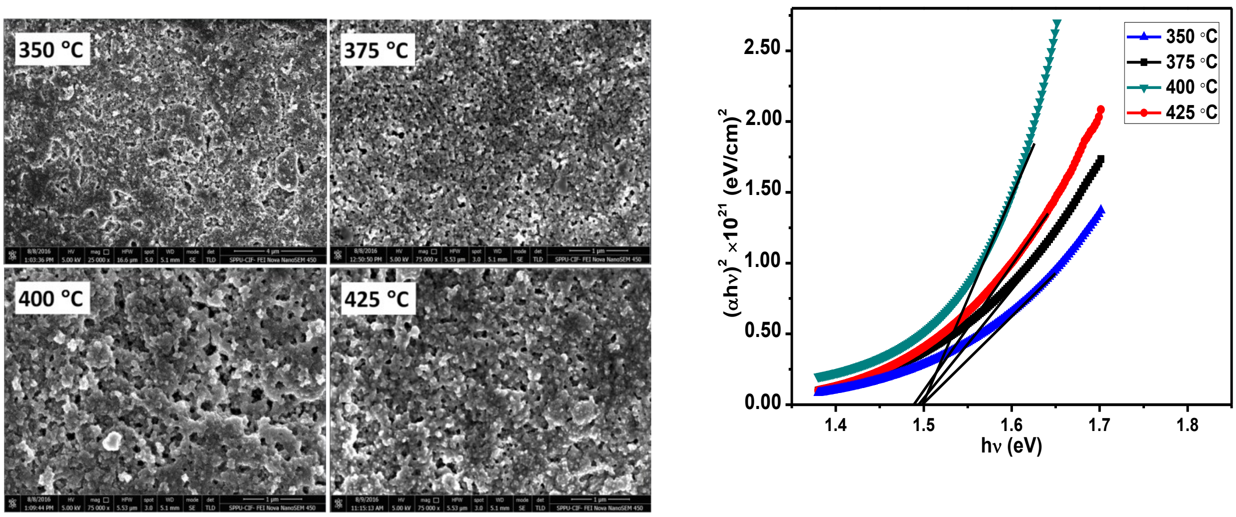
FE-SEM analysis

As shown in the Fig. 2(a), the effect of the synthesis temperature on the morphology and crystallite size is observed. At the lower substrate temperature, surface is dense with small crystallite size. As the substrate temperature increases to 400 °C, the sponginess properly increased. Also the crystallite size increases and matches with the as calculated from XRD patterns. The improvement in crystallinity of the film take place by the agglomeration of the adjacent islands motivated by the heat energy incorporated from the higher substrate temperature [19].



(a) (b)

FIGURE 1. Structural property analysis of CZTS thin films: (a) X-ray diffraction pattern of CZTS thin films at different substrate temperatures. (b) Variation of crystallite size and dislocation density with respect to the deposition temperature.



(a) (b)

FIGURE 2. (a) FE-SEM images of CZTS deposited at different temperatures. (b) Optical band gap variation with respect to temperature.

Optical analysis

The investigational value of absorption coefficient as a function of photon energy hν gives the band gap energy of the films. Extrapolating the straight line of $(\alpha h\nu)^2$ vs photo energy (hν) curve to the intercept on horizontal photon energy axis gives the band gap value, as shown in Fig. 2(b). It is observed that, band gap varies in the range of 1.489 eV to 1.499 eV. This energy gap range is fitting of energy gap for the efficient solar cells.

CONCLUSIONS

The quaternary kesterite Cu_2ZnSnS_4 (CZTS) thin films using sulphate precursors were successfully synthesized on SLG by chemical spray pyrolysis method at different substrate temperatures. The effect of substrate temperature on the structural, morphology and optical properties is studied. The observed phase of deposited CZTS films is kesterite. As the synthesis temperature rises to 400 °C, the porosity increases sufficiently. The optical band gap varies in the range of 1.489 eV to 1.499 eV. By taking into consideration of all the parametric study, the Cu_2ZnSnS_4 (CZTS) thin films prepared using sulphate precursors at 400 °C by spray pyrolysis will beneficial for the solar cells.

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