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Effect of Annealing on the Electrical Properties of Cu_xS Thin Films

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Abstract

Copper sulphide CuS was deposited on three substrates; glass, Indium Tin Oxide (ITO) and Ti by using spray pyrolysis deposition (SPD). After depositing CuS thin films on the substrates at 200°C, they were annealed at 50, 100, 150, and 200°C for 1 hour. Structural measurements revealed covellite CuS and chalcocite Cu_2S phases for thin films before and after annealing at 200°C with changes in intensities, and only covellite CuS phase for thin films after annealing at 50, 100, and 150°C. Morphological characteristics show hexagonal-cubic crystals for the CuS thin film deposited on glass substrate and plates structures for films deposited on ITO and Ti substrates before annealing, these crystals became bigger in size and there were be oxidation and some agglomerations in some regions with formation of plates for CuS on glass substrate after annealing at 200 °C. For Hall Effect measurements, thin films sheet resistivity and mobility increased after annealing while the carrier concentration decreased. Generally, the thin film deposited on ITO substrate had the lowest resistivity and the highest carrier concentration before and after annealing. The thin film deposited on Ti substrate had the highest mobility before and after annealing, which makes it the best thin film for device performance. The objective of this research is to show the improvement of thin films electrical properties especially the mobility after annealing those thin films.

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Keywords: CuS; annealing; p—type thin film.

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1. Introduction

In the past few decades, there has been an increasing investigation in semiconducting chalcogenide thin films, which have been due to their wide applications in various fields of science and technology[1]. Among them, copper sulfide is the most commonly used material. The copper sulfide thin film attracts the attention of many researchers mostly due to its semiconducting properties. Also, the constituent elements of this material (Cu and S) are non-toxic and abundant in nature[2].

The copper sulphide is an important material from the point of basic research because it is known to exist in several crystallographic and stoichiometric forms. They are copper-rich phases exist as chalcocite (Cu_2S), djurlite ($Cu_{1.96}S$), digenite ($Cu_{1.8}S$) and anilite ($Cu_{1.75}S$). And sulphur rich phase exists as covellite (CuS). It is well known that Cu_xS ($1<x<2$) has a distinct composition because of the variation in x, with different stoichiometry and oxidation and temperature is responsible for a change from one composition to another[3].

Chemical methods have been used to grow CuS films. The shape, phase and the size of inorganic nano-crystals and micro-crystals are the determinant elements in varying their electrical, optical and other properties[4].

The chemical methods are economical and desirable structure can be obtained with them. Therefore, to grow CuS thin films with desirable shape and structure, the chemical method was employed[4]. Several techniques have been used: Chemical Bath Deposition (CBD)[2,5,6], Successive Ionic Layer Adsorption and Reaction (SILAR)[4,7], and Spray Pyrolysis Deposition (SPD)[8-10].

As an important semiconductor with unique electronic, optical and chemical properties, CuS thin films are of great concern due to its wide range of application in optical and electrical devices, such as photothermal conversion of solar energy, electro-conductive electrodes, microwave shielding coatings, solar control coatings, dye-sensitized solar cell, potential nanometer-scale switch, selective radiation filter on architectural windows, solar cells, cathode material in lithium rechargeable batteries and some chemical sensing applications[4], photoconductor, antireflection coatings, interference items, polarizers, narrow band filters, waveguide coatings, IR detectors, temperature control of satellites[7], light emitting diodes and other optoelectronics[5].

In this paper, CuS thin films were deposited on three different substrates by spray pyrolysis deposition. Then these thin films were annealed at 50, 100, 150 and 200 °C for 1 h. X-Ray Diffraction (XRD), Field Emission Scanning Electron Microscopy **(**FESEM) and Hall effect measurements were obtained for thin films before and after annealing to find which thin film and at which temperature has better contact and can perform as a device. To the best knowledge of the authors the results presented in this paper are not reported so far in the literature, only deposition CuS on different substrate or annealing for one substrate were reported. This search studies the effects of four different temperatures for thin films deposited on three different substrates to find which one will be more suitable as a device according to its electrical properties.

2. Experimental

Copper chloride and sodium thiosulfate were used to prepare the solution for depositing CuS thin film by spray pyrolysis deposition. The deposition by this method is simple, cover a large area, low cost and widely used. The solution was sprayed on glass, ITO and Ti substrates at 50, 100, 150 and 200 °C which is suitable for deposition CuS because at higher temperature the film will peel and the lower temperature is not enough for composition CuS thin film on the substrate as there is no catalyst in this work so the temperature will take place the catalyst. The best results of CuS thin films were obtained at 200-210˚C, this range of temperature was chosen because CuS is reported to decompose at ≈220˚C[10]. The distance between the nozzle and the substrates was 30 cm, which is the best spray distance because more than this will cause spray flight out of the substrate, less than this will cause the combined solution drops in one spot. Then after completing the deposition, these thin films were annealed at 50, 100, 150 and 200°C for 1 h. Annealing can be defined as heat treatment of materials at elevated temperatures aimed at investigating or improving their properties. Material annealing can lead to the phase transition, recrystallisation, homogenization and relaxation of internal stresses. Structural and electrical properties were measured for thin films before and after annealing using XRD, FESEM and Hall effect measurements.