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Improved mobility of sol-gel method processed transparent tin sulfide thin films

Jian Sun [a,*], Yifei Huang [a], Sha Nie [a], Zequn Chen [a], Jianmei Xu [a,*], Ling Zhao [a], Wei Zhou [a], Qing Wang [a], Hao Gong [b]

[a] Faculty of Materials Science and Chemistry, China University of Geosciences, 430074, China
[b] Department of Material Science and Engineering, National University of Singapore, 21 Lower Kent Ridge Road, 117543, Singapore

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ABSTRACT

Transparent conductive tin sulfide is processed at room temperature by a cheap simple sol-gel method. The effect of annealing temperature on its crystal structures, morphologies, electrical and optical properties are systematically studied. High electron mobility of 48 $cm^2 V^{-1} s^{-1}$ and the average transmittance of 61% in 400–850 nm are achieved, indicating its potential application in invisible TFT.

1. Introduction

Owing to the demand in flexible and invisible electronics, there is an increasing interest in transparent conducting films (TCFs). One application of TCFs is as a channel layer of thin film transistors (TFTs). a-IGZO with Hall mobility larger than 10 cm^2/Vs is currently the most widely studied chanel layer material [1] and has already entered the stage of industrialization. It is interesting and important to develop new TCFs to replace a-IGZO due to the natural scarcity of indium, scientific curiosity and potential applications.

Tin sulfide is a potentially promising, yet relatively less studied, TCF comprised of earth abundant elements [2,3]. Tin sulfide thin films have been deposited using various methods, such as thermal and electron beam evaporation [4], RF-sputtering [5], chemical vapor deposition [6], spray pyrolysis [7], electrochemical deposition [8], chemical bath deposition [9] and spin-coating methods [10,11]. Compared with vacuum based methods, solution based methods have been a more attractive alternative due to the cost-effective production capabilities. It is very necessary to find a chemical method to fabricate n-type tin sulfide thin films with good performance. Spin-coating method is cheap, simple and compatible to roll-to-roll industry. To our knowledge, there are only the above two reports on spin-coating fabricated tin sulfide on the application of solar cells [10,11]. Tin sulfide in other kinds of applications has not been reported yet and the electrical and optical properties of tin sulfide are still not clear.

In this work, tin sulfide thin films are prepared via a cheap and simple sol-gel method. An improved Hall electron mobility of 48 cm^2/V s is achieved. The effect of annealing temperature on the tin sulfide thin films are studied.

2. Material and methods

For the synthesis of tin sulfide thin films, $SnCl_2 \cdot 2H_2O$ and SC $(NH_2)_2$ were precursors, 2-methoxyethanol and monoethanolamine were served as solvent and stabilizer, respectively. Sn/S molar ratio in the mixed solution was kept at 1:1.5. Ar was first blowed in the flask and then the mixed solution was loaded into the flask and the flask was put on the hotplate at 80 °C under vigorous magnetic stirring for 2 h. After full reaction, the solution became light yellow. Tin sulfide thin films were synthesized via a spin-coating method on glass substrates. Then the samples were dried at 200 °C in the tuber furnace with flowing argon for 30 min. The samples were repeated another 4 times for spin-coating and drying. Finally, the samples were annealed at 300 °C, 350 °C, 400 °C, 450 °C in the furnace with a flowing argon.

X-ray diffraction (XRD) patterns of the samples were obtained by BRUKER AXS (model D8 ADVANCE, $\lambda_{CuK\alpha}$=0.154056 nm). The conductivities, sheet resistances, mobilities and carrier concentrations of the samples were measured with the Hall system in a Van der Pauw configuration at room temperature. The surface morphology of the samples was examined by field emission

* Corresponding authors.
E-mail addresses: sunjian516@gmail.com (J. Sun), xjmcjh@163.com (J. Xu).

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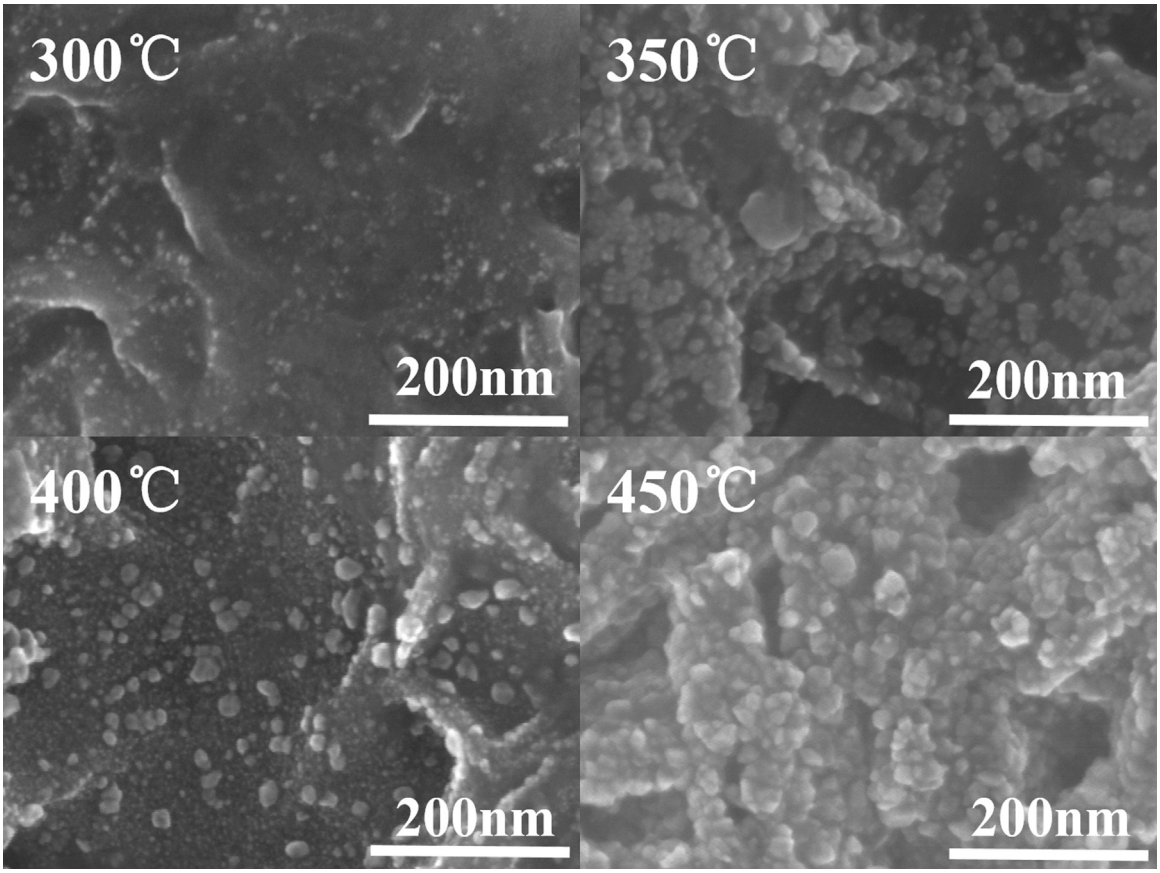

Fig. 2. Surface SEM images of the samples annealing from 350 °C to 450 °C.

scanning electron microscopy (FESEM) (Hitachi-SU8010). S/Sn molar ratio of the sample was tested by energy dispersive X-ray spectroscopy (EDS). The transmittance spectra of the samples were measured with a SHAIMADZU UV-2550 UV–vis spectrophotometer.

3. Results and discussion

Fig. 1 shows XRD patterns in the 2θ range of 10° – 60° for all the samples as functions of annealing temperature. For the samples annealed at 300 °C, the peaks at ~31.5° can be identified as (040) of SnS, according to the XRD PDF card No. 39-0354; while the peaks at 15.1° belong to (001) of SnS_2 according to PDF card No. 01-1010. For the samples annealed from 300 °C to 350 °C, the intensity of SnS (040) decreases and (001) of SnS_2 almost disappears. When the annealing temperature is increased from 350 °C to 400 °C, the intensity of the peak (040) of SnS becomes even weaker while the peaks (110) and (101) of SnO_2 appear, following PDF card No. 41-1445. With a further increase of the annealing temperature from 400 °C to 450 °C, the peak (040) of SnS disappears and only the peaks (110) and (101) of SnO_2 exist, indicating that tin sulfides are all oxidized. From XRD analyses, tin sulfide annealed at 350 °C can have a preferred orientation of SnS (040).

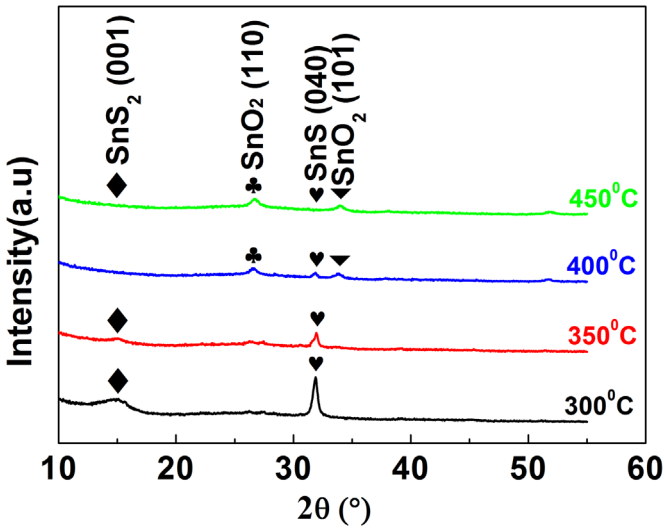

Fig. 1. XRD patterns of the samples as a function of annealing temperature.

Fig. 2 shows the SEM images of the samples annealing from 350 °C to 450 °C indicating their islands growth mode. From Fig. 2, it can be seen that as the annealing temperature increases from 300 °C to 350 °C, the size and the number of islands both increase, which is understandable since the island size and number of the tin sulfide will increase at an increased temperature according to the mechanism of the crystal growth in islands growth mode. As the temperature further increases from 350 °C to 400 °C, the size and the number of islands seem to decrease since tin dioxide starts to appear. When increasing from 400 °C to 450 °C, the size and the number of islands seems to increase again, which is due to the increased temperature for tin dioxide following the mechanism of islands growth mode. By energy dispersive X-ray spectroscopy (EDS), S/Sn molar ratio of the sample annealed at 350 °C is 0.82, close to 1:1 stoichiometry of the SnS, consistent with the XRD result of mainly SnS structure.

From the Hall measurements, all the samples show an n-type behavior. Fig. 3 reveals that the electron mobility increases from