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Cu_2SnS_3 thin films growth in controlled Sn-S vapor for grain boundary suppression

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Abstract

Cu_2SnS_3 (CTS) has been expected to be a next-generation solar cell material. However, the power conversion efficiency (PCE) of CTS solar cells is still low. One of the reasons for this low PCE is the recombination of carriers that occurs at the grain boundaries of CTS films. In this study, to suppress the grain boundary of the CTS, the effect of the sulfurization processes in mixed tin sulfide (Sn-S) and Sulfur (S) vapor on the grain size of CTS was investigated. The grain size of CTS deposited in Sn-S vapor was no obvious changed. On the other hand, although deposited CTS without Sn-S vapor contained an extra phase such as tetragonal CTS, the CTS films deposited in Sn-S vapor were dominantly attributed to monoclinic CTS. Thus, sulfurization in Sn-S and S vapor implies contribution to the growth of monoclinic CTS films.

1. Introduction

In recent years, absorption materials of solar cells with a high optical absorption co-efficient and the ability to be made into thin films have been the focus of much attention. In the field of thin films solar cells, several studies have focused on Cu_2SnS_3 (CTS), which are composed of non-toxic elements abundant in the earth's crust and have a theoretical power conversion efficiency (PCE) over 30%[1)]. However, their PCE is still as low as 5.24%[2)] and they have not yet reached practical application. One of the reasons for this is the recombination of carriers that occurs at grain boundaries in CTS thin films[1)]. Previous studies[3)] reported that sulfurization in tin sulfide (Sn-S) vapor for CTS could potentially lead to an increase in grain size of CTS crystals. This increase in grain size may suppress recombination of carriers by grain boundaries. In this study, the effect of the sulfurization process in mixed vapor Sn-S and sulfur (S) on the grain size of CTS was investigated using a sulfurization furnace (3-zone furnace)[4)] which the Sn-S and S vapor can be precisely controlled.

2. Experiment

Cupper (Cu)/Sn-S precursors were deposited on Mo-coated soda-lime glass using Radio Frequency sputtering equipment. A schematic diagram of the 3-zone furnace is shown in Fig. 1. The 3-zone furnace can independently control the temperature in each of the three locations. CTS was sulfurized by controlling the temperature of each part in this furnace. The temperatures of precursor and S powder were set at 550 and 200°C, respectively. Additionally, the temperature of the SnS_2 powder during the sulfurization process was changed from 550 to 850°C to study the effect of Sn-S vapor on the grain size of CTS. The CTS films were measured by X-ray diffraction (XRD), Raman spectroscopy (Raman),

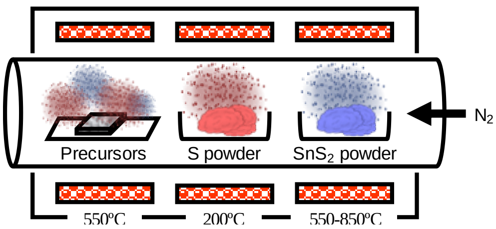

Fig.1 3-zone furnace

electron probe micro analyzer (EPMA) and scanning electron microscope (SEM).

3. Results and discussion

The Raman spectra of the obtained CTS films are shown in Fig. 2. CTS without (w/o) SnS_2 (S powder at 200ºC) is attributed to monoclinic and tetragonal CTS[4]. In contrast, the CTS thin films deposited in Sn-S vapor at 550-850ºC are only attributed to monoclinic CTS[5], which is a suitable for the absorber layer of solar cell[2,3]. Thus, sulfurization in Sn-S and S vapor implies contribution to the growth of monoclinic CTS films. On the other hand, no increase in the grain size of CTS deposited in Sn-S vapor was observed by SEM. This may be due to an insufficient supply of Sn-S vapor to the precursor during sulfurization. The Sn-S vapor cooled near the precursor despite the intentional generation of Sn-S vapor. The vapor pressure of Sn-S is low, which imposes limitations on the quantity of vapor generated at the sulfurization temperature of 550°C for the precursor. Consequently, the amount of vapor reaching the precursor may be reduced.

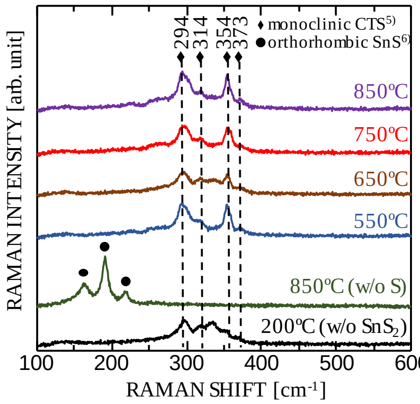

Fig.2 Raman spectra of growing CTS films in mixed Sn-S and S vapor.

4. Conclusions

The effect of the sulfurization process in mixed Sn-S and S vapor on the grain size of CTS was investigated using a 3-zone furnace. Deposition in mixed Sn-S and S vapor implies contribution to the growth of monoclinic CTS. On the other hand, grain size of CTS deposited in mixed Sn-S and S vapor was not increased. This may be due to an insufficient supply of Sn-S vapor to the precursor during sulfurization. Therefore, it is necessary to develop new methods to intentionally control Sn-S vapor.

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