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Effect of Post-deposition Se-annealing on the defect distribution in $CuGaSe_2$ Thin-films and Solar-cells

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Chalcopyrite Cu(In,Ga)Se_2 (CIGS) being a pseudo-binary alloy of ternary $CuInSe_2$ and $CuGaSe_2$ is one of the most promising materials to realize high-efficiency thin-film solar cell. Band-gap of the $CuInSe_2$ is 1.04 eV, while it can be widened by inserting Ga and approached to 1.68 eV for the $CuGaSe_2$ counterpart. The value is close to the ideal band-gap (1.4 eV under AM 1.5 sunlight) of the absorber-layer to achieve the highest possible efficiency. Moreover, larger band-gap makes the $CuGaSe_2$ suitable for the top-cell in the tandem structure together with $CuInSe_2$ as the bottom-cell. Nevertheless, so far, $CuGaSe_2$ solar cells with a CdS buffer have achieved efficiency of around 11 % for the thin film [1]. Therefore, to achieve the optimized material quality of the $CuGaSe_2$ material which is compatible for highest possible efficiency, an extensive study of this material with various compositions is indispensable. In this paper, we have changed the post-growth Se-annealing time to study the effect of Se-annealing on the properties of $CuGaSe_2$ film.

Polycrystalline $CuGaSe_2$ thin films with the typical thickness of 2 μm were grown over Mo-coated soda lime glass (SLG) substrates through a three-stage co-evaporation process. After the third stage of growth, sample-temperature was decreased at a rate of 9°c/min down to 250°c in a Se atmosphere that we defined as Se-annealing. During Se-annealing Cu, and Ga shutter remained closed Several $CuGaSe_2$ samples with different Se-annealing time were grown. Doppler broadening spectra of the annihilation radiation were measured as a function of the incident positron energy *E* using a continuous-current-type positron beam located at the University of Tsukuba. The spectra were characterized by the *S* parameter, defined as the fraction of annihilation events in the energy range of 510.24 - 511.76 keV. The relationship between *S* and *E* was analyzed by VEPFIT, a computer program. In general, increased *S* value suggests increased amount of vacancy type defects in the material.

Figure 1 shows the *S*-*E* curve of the $CuGaSe_2$ samples with different Se- annealing time of 5, 40, and 60 min. Depth distributions of the *S* parameter were shown at the inset of the figure. A general decrease in the *S* value at the near-surface region with an increase of the annealing time suggests reduction of vacancy type defects with annealing. Volume of the near-surface defect-rich zone also decreased with increasing annealing time. However, for the samples with 40 min annealing time, vacancy type defect was further decreased in the bulk region, while for the sample with 60 min annealing time defect in the bulk region is higher than the sample with 40 min annealing time. This observation is consistent with the improved electrical properties (reduced resistivity, and increased hole carrier concentration) of the $CuGaSe_2$ film with increased annealing time up to 40 min.

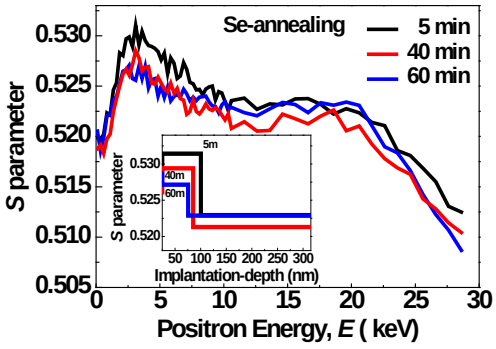

Fig. 1 *S*-*E* curve of $CuGaSe_2$ samples with different annealing time. Inset shows depth distributions of *S* parameter obtained from analysis of the *S*-*E* curves

Usually, at the end of the third stage of $CuGaSe_2$ growth, a defect-rich region mainly composed of vacancy-type defects has been formed at the surface of the film. It can be hypothesized from our results of the positron annihilation technique that it is possible to control the diffusion of these defect clusters to the bulk of the material by post deposition Se-annealing. Thus, post-deposition Se-annealing has been found to affect both the surface and also bulk of the film by controlling the defect-distribution along the depth of the film [2]. Mechanism of the defect distribution has been investigated. Performance of the fabricated solar cells has been studied in correlation to the Se-annealing time.

Ref: [1] Ishizuka et al. Appl. Phys. Lett.103 (2013) 143903.
[2] Uedono et al. Thin Solid Films 603 (2016) 418.