Impacts of proton irradiation on optical and electrical properties of $Cu(In,Ga)Se_2$ thin films and solar cells

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Abstract

The optical and electrical properties of proton irradiated $Cu(In,Ga)Se_2$ (CIGS) solar cells and the thin films that compose the CIGS solar cell structure were investigated. The transmittance and resistivity of transparent conducting oxide window layers remained constant for a fluence of up to 3×10^{15} cm^{-2}. For CIGS thin films, the number of non-radiative recombination center increases under proton irradiation. In CIGS solar cells, decreasing J_{SC} reflected the degradation of the depletion layer of the CdS/CIGS interface. These results constitute the first step in clarifying the degradation mechanism of CIGS solar cells.

1. Introduction

Thin film solar cells composed of polycrystalline $Cu(In,Ga)Se_2$ (CIGS) show high conversion efficiency and excellent radiation tolerance, making them very promising for space applications. In fact, irradiation-damage studies of CIGS solar cells in space have revealed that their electrical properties show relatively less degradation due to proton[1-6], electron[1,4,7-9], or gamma-ray[10] irradiation compared with Si solar cells in case of space satellite application for several years. However, despite many reports on CIGS solar cells[1-3,6-10], the degradation and/or resistance mechanism of these solar cells under irradiation remains to be clarified, because there have been few reports on irradiated CIGS thin films[4,5].

The mechanism of solar cell degradation, as researchers have gradually come to realize, is a complex phenomenon. Apart from irradiation effects, CIGS solar cell performance is known to be influenced by conditions such as damp heat[11,12] or light soaking. For example, Weinert *et al*. reported that the degradation properties of open-circuit voltage (V_{OC}) and short-circuit current density (J_{SC}) are different, and Pern *et al*. reported that the ZnO:Al layer degrades under damp-heat conditions of 85°C temperature and 85% humidity when it is used in CIGS solar cells[12]. Therefore, understanding the degradation properties of each semiconductor composing CIGS solar cells is necessary before using the cells in commercial or space applications.

Our group has previously studied the irradiation effects on transparent conducting oxide (TCO), CIGS, and $CuInSe_2$ (CIS) thin films and CIGS solar cells subjected to alpha rays (irradiation energy: 6.2 MeV, fluence: 1.4×10^5 cm^{-2}), protons (30–70 MeV, 4.9×10^9 cm^{-2}), gamma rays (0.66 MeV, 4.7×10^{14} cm^{-2})[13], and electrons (2 MeV, 1.0×10^{13}–1.5×10^{18} cm^{-2})[14]. We found that particle irradiation tended to degrade primarily around the interfaces of the several layers that compose CIGS solar cells. These results suggest that the interfaces play an important role in defining the solar cell performance. From a scientific viewpoint, an investigation of the irradiation effects on CIGS thin films is needed to reveal the degradation mechanism of CIGS-based solar cells. On the other hand, from an industrial viewpoint, this investigation is needed to clarify the radiation tolerance of a variety of irradiation sources for practical applications. Therefore, in this study, the effects of proton irradiation on the properties of TCO and CIGS thin films are demonstrated by irradiating each thin film that composes a CIGS solar cell structure.

2. Experiment

Single-phase polycrystalline CIGS ($CuIn_{0.8}Ga_{0.2}Se_2$) films with thicknesses of approximately 2.0 μm were prepared by a conventional three-stage process on Mo-coated soda-lime glass (SLG). These films were used to fabricate CIGS solar cells with an undoped ZnO (~100 nm)/CdS (~50 nm) layer by RF sputtering and chemical bath deposition. Finally, several types of window layers (1.0 μm) were deposited by RF sputtering using different TCOs, namely, ITO, IZO, ZnO:Al, and ZnO:Ga. The solar cells exhibited an efficiency of about 7% measured under simulated air mass (AM) 1.5 illumination. TCO thin films were also deposited on bare SLG to enable the investigation of their optical and electrical properties.

CIGS solar cells and the CIGS thin films were irradiated in vacuum without intentional heating. Previous reports show that degradation tends to start at 1×10^{12} cm^{-2} [1,15]; hence, in this study, we applied proton fluence ranging from 1×10^{11} to 3×10^{15} cm^{-2}. TCO and CIGS thin films were irradiated with 200 keV protons. The proton energy was estimated to investigate proton irradiation effect on each part of the CIGS solar cells; the estimation was carried out using Stopping and Range of Ions in Matter (SRIM) simulations[16]. Figure 1 shows the tracks of protons in CIGS solar cells at (a) 30, (b) 100, (c) 200, and (d) 380 keV, as simulated by SRIM. By adjusting the proton energy, selective regions in the solar cells could be irradiated: (a) only the surface of the window layer (30 keV), (b) from window layer to around *n*-layer (100 keV), (c) through the depletion layer (200 keV), and (d) through the bottom of the CIGS layer (380 keV).

Optical transmittance spectra were measured in the ultraviolet region by using a deuterium lamp and in the visible and near-infrared regions by using a halogen lamp. The transmittance values of the films were deduced by using irradiated bare SLG substrates as references. The resistivity ρ, Hall mobility μ, and carrier density n at room temperature (RT) were measured by the van der Pauw technique. For CIGS thin films and solar cells, photoluminescence (PL) was carried out at 77 K using the 532.0 nm line of a frequency-doubled quasi-cw Nd:YAG laser (60 mW) as an excitation source. Phase-sensitive detection was carried out from 950 to 1550 nm using a 50-cm focal length grating monochromator and liquid-N_2-cooled Ge photodetector. The performance of the solar cells was determined on the basis of the current density–voltage (J–V) curves measured at RT under AM 1.5 and 100 mW/cm^2 illumination.

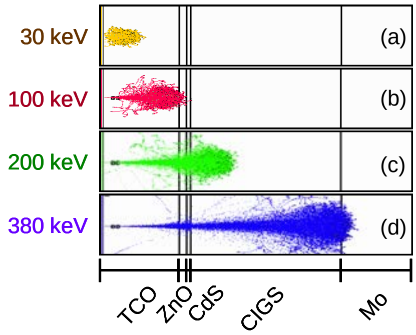

Fig. 1. The tracks of proton in CIGS solar cells with (a) 30, (b) 100, (c) 200, and (d) 380 keV, simulated by SRIM.

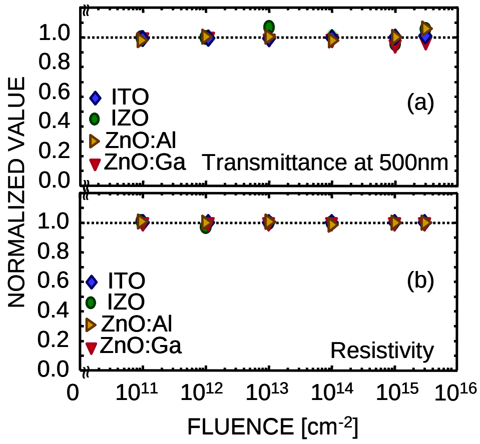

Fig. 2. The normalized (a) transmittance at 500 nm and (b) resistivity of ITO, IZO, ZnO:Al and ZnO:Ga as a function of irradiation fluence.

3. Results and Discussion

First, the proton irradiation effects on thin films composing the window layer and absorber in CIGS solar cells were investigated. Figure 2 shows the normalized (a) transmittance at 500 nm and (b) resistivity of several TCO thin films, namely, ITO, IZO, ZnO:Al and ZnO:Ga, as a function of irradiation fluence. It should be noted that the normalized values were defined by the values of the transmittance at 500 nm or resistivity after/before irradiation. These transmittance and resistivity values of TCO thin films did not change with respect to proton irradiation. Our group previously reported that the transmittance at 500 nm and resistivity of ITO and IZO did not change for less than 1 × 10^{18} cm^{-2} electrons and those of ZnO:Al and ZnO:Ga did not change for less than 1 × 10^{16} cm^{-2} electrons[14]. Moreover, K. S. Chan *et al.* reported that the structure of ZnO did not change at room temperature under 3 × 10^{16} cm^{-2}. These results indicate that ITO, IZO, ZnO:Al, and ZnO:Ga thin films have a good tolerance to particle irradiation[17], such as proton and electron irradiation at less than 3 × 10^{15} cm^{-2}.

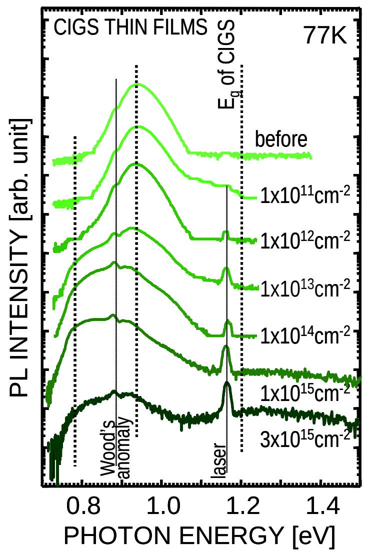

Fig. 3. PL spectra of CIGS thin films at 77 K as a function of irradiation fluence.

The PL spectra of CIGS thin films irradiated with 3 × 10^{15} cm^{-2} proton fluence are shown in Fig. 3 as a function of proton energy. With increasing irradiation fluence, the PL peak at 0.94 eV tends to disappear and a PL peak at 0.8 eV tends to appear. A previous report also showed that PL peak intensities due to deep level defects of CIGS were decreased under electron[14] and proton[6] irradiation. Note that XRD patterns and surface SEM images did not change (data not shown). Therefore, this result may indicate that the number of non-radiative recombination center increases under proton irradiation.

Next, the effects on proton irradiation on CIGS solar cells were investigated. The PL spectra of CIGS solar cells are shown in Fig. 4 as a function of proton irradiation energy at 3

$\times 10^{15}$ cm^{-2} fluence. The PL peak at 0.94 eV disappears under irradiation with 200 and 380 keV proton energy. This result taken together with the simulation results in Fig. 1 indicates that the region around the CdS/CIGS interface, which forms the depletion region of CIGS solar cells, degraded under 200 and 380 keV proton energy.

Finally, the effects of the degraded depletion layer on the electrical properties were investigated. Two normalized electrical performance parameters of the conventional CIGS solar cell, namely, V_{OC} and J_{SC}, were plotted as a function of irradiation fluence for each irradiation energy, as shown in Fig. 5. The proton irradiation energy indicates the degraded position, as mentioned earlier. In the case of 30 keV proton energy, which only degraded the surface of the window layer, V_{OC} and J_{SC} did not change when the fluence was around the order of 10^{15} cm^{-2}. Since the transmittance and resistivity of several TCO thin films did not change, this result indicates that the degradation of the window layer did not affect the solar cells properties. On the other hand, in case of 200 and 380 keV proton energy, which degraded through the depletion layer and through bottom of the CIGS layer, V_{OC} and J_{SC} tend to decrease with increasing irradiation fluence for fluence values greater than around the order of 10^{13} cm^{-2}. These results indicate that the degradation of the depletion layer of CIGS affects CIGS solar cell properties. In the case of 100 keV proton energy, intermediate results between only window layer degradation and through the CIGS layer degradation were obtained. These results taken together with those in Fig. 1 indicate that precisely controlling degraded position around the CdS/CIGS interface is difficult. A further in-depth investigation is needed to reveal the degradation mechanism of CIGS solar cells. Our study is the first step toward realizing practical applications of CIGS solar cells in space and clarifying their degradation mechanism.

4. Conclusion

The effects of proton irradiation on the optical and electrical properties of CIGS solar cells and the layers that compose the solar cell structure, namely, CIGS, ITO, IZO, ZnO:Al, and ZnO:Ga, were measured. The transmittance and resistivity of TCO thin films did not change after irradiation. PL peak intensity of CIGS thin films decreased with increasing proton fluence. The experimental *J–V* properties of CIGS solar cells indicated that the interface of the depletion layer might be weaker than that of the other layers in the bulk. This study partially clarifies the degradation mechanism of CIGS solar cells.

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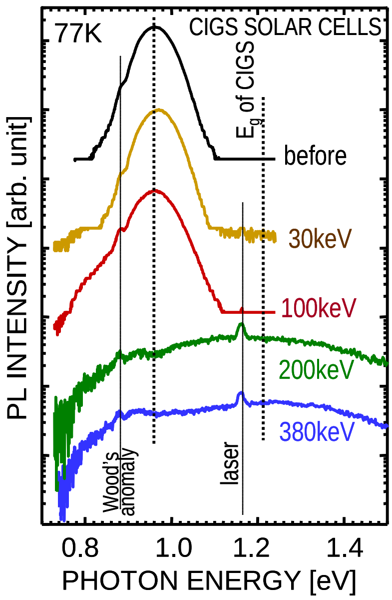

Fig. 4. PL spectra of CIGS solar cells at 77 K irradiated with 3×10^{15} cm^{-2} as a function of proton energy.

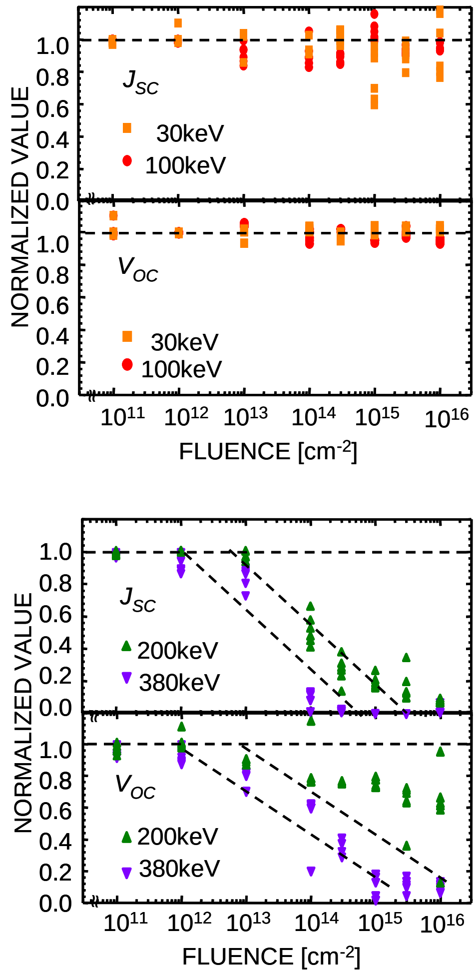

Fig. 5. The normalized (a) J_{SC} and (b) V_{OC} values of CIGS solar cells irradiated with 30 and 100 keV and (c) J_{SC} and (d) V_{OC} values of CIGS solar cells irradiated with 200 and 380 keV as a function of proton irradiation fluence.

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