Study of the electrical properties of CuGaSe₂ thin-film solar-cells using admittance spectroscopy

Tsukuba Univ. ¹, AIST², Muhammad Monirul Islam¹, Shogo Ishizuka², Hajime Shibata², Shigeru Niki², Katsuhiko Akimoto¹, and Takeaki Sakurai¹

E-mail: islam.monir.ke@u.tsukuba.ac.jp

Chalcopyrite Cu(In,Ga)Se₂ (CIGS), a binary alloy of CuInSe₂ and CuGaSe₂, is a promising material for high-efficiency thin-film solar-cells. Band-gap of the CuInSe₂ is 1.04 eV, while it becomes 1.68 eV for the CuGaSe₂. Thus, band-gap of CuGaSe₂ is close to the ideal band-gap of the absorber-layer to achieve highest possible efficiency. Moreover, larger band-gap makes CuGaSe₂ suitable for the top-cell in the tandem solar-cell structure together with CuInSe₂ as a bottom-cell. Nevertheless, so far, CuGaSe₂ solar cells with a CdS buffer have achieved efficiency of around 11%. Therefore, to achieve the efficiency beyond current limit, an extensive study of the CuGaSe₂-material including defect-study with various compositions is indispensable. In this paper, we have used admittance spectroscopy to study the defect properties of CuGaSe₂ thin-film solar cells, where CuGaSe₂ thin-films were grown with various Se-flux (Pₜₜₑ) conditions.

Polycrystalline CuGaSe₂ thin-films with 2 μm of thickness were grown over Mo-coated soda lime glass (SLG) substrates through three-stage co-evaporation process. Several samples were deposited by changing the Pₜₜₑ-conditions. Admittance spectroscopy (AS) was carried out in the dark within the temperature range, 50 ~ 380 K using Agilent 4284A LCR meter. Modulation frequency was varied from 1 kHz to 1 MHz. For AS measurement, we considered the depletion region as a parallel combination of capacitance, Cₚ and conductance, Gₚ, thus admittance becomes, \( Y = Gₚ + iωCₚ \). Since, imaginary part Cₚ and real part Gₚ in the admittance term are related explicitly by Kramers-Kronig relations, similar information can be obtained from both Cₚ and Gₚ. Here, we used both parameters to investigate the nature of the trap signatures found in our measurement.

Shown in Fig. 1(a), is the temperature dependence admittance spectra of a CuGaSe₂-based solar cells structure (ZnO/CdS/CuGaSe₂/Mo/SLG) at various frequencies; (b) CuGaSe₂ solar cells with various Pₜₜₑ conditions during CuGaSe₂ growth.

In Fig. 1(a), the temperature dependence admittance spectra of a CuGaSe₂ based solar cells at various frequencies, where CuGaSe₂ films were deposited with low Se-flux condition of 2.39 ×10⁻³ Pa. AS spectra shows two major peaks, α and ζ. Peak-α is usually attributed to the Cu-vacancy in the CuGaSe₂ material, while peak-ζ corresponds to the electrical response of a defect-level within the band-gap of the CuGaSe₂. Increase in the temperature shifts the AS-peaks (i.e., characteristic frequency, \( ω₀ \)) to the higher value. Thus, activation energy, \( Eₐ \) of a particular defect could be calculated from the slope of the corresponding Arrhenius plots, ln(\( ω₀/T^2 \)) versus 1/T. Here, T is the temperature of the AS-peak. \( Eₐ \) of the Peak-α and ζ was calculated as ~ 50 meV and 350 meV above valence band (\( Eᵥ \)) of the CuGaSe₂, respectively. As seen from the Fig. 1(b), in general, intensity of the peak-ζ increases with a decrease in the Pₜₜₑ-condition during growth of the CuGaSe₂ films.

Defect-parameters including defect-density, capture cross-section etc. were calculated, and compared with the Pₜₜₑ-condition during growth. In general, CuGaSe₂ samples grown with higher Pₜₜₑ show lower defect density (peak- ζ), and improved solar-cell performances. Origin of the peak- ζ has been discussed. Temperature-dependent AC-conductance (G), and capacitance-voltage (C-V) measurement was performed to obtain transport properties, charge-density etc., and discussed in relation to the Se-flux condition during deposition of the CuGaSe₂ films.