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Study of Heterojunction Interface for High-efficient Cu(In,Ga)Se₂ solar cells

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Photovoltaic power generation is expected as one of clean renewable energy sources to solve environmental and energy issues. Cu(In,Ga)Se₂ (CIGS) is promising materials for absorber of solar cells because of its unique characteristics. CIGS which has high absorption coefficient enable to be applied for absorber of flexible lightweight thin-film solar cells, and therefore it is possible to realize reduction of mass production cost by roll-to-roll deposition in continuous high-speed manufacturing process. The highest conversion efficiency of 22.9% has been achieved in CIGS solar cells. In this thesis, to further enhance conversion efficiency in CIGS solar cells, required condition for suppression of recombination at CdS/CIGS hetero junction interface in CIGS solar cells was theoretically analyzed, and the effectiveness of Cu-deficient layer (CDL) on CIGS was experimentally suggested, finally establishing the unique control methods of CDL.

A guideline for improvement of CdS/CIGS hetero interface in CIGS solar cells was suggested using one-dimensional device simulation. Carrier concentration and energy bandgap in surface layer (SL) at the hetero interface were changed to reproduce effects of donor doping and valence band offset (ΔE_V), respectively, on CIGS surface region. High conversion efficiency of 22.1% and 21.6% was obtained by suppressing interfacial recombination when SL had high donor concentration (N_D) of $1.0 \times 10^{18} \text{ cm}^{-3}$ and high ΔE_V of 0.2 eV, respectively, although the efficiency was 19.6% in SL with low N_D of $1.0 \times 10^{15} \text{ cm}^{-3}$ and with ΔE_V of 0 eV. It was revealed that the required condition for SL to boost conversion efficiency is $N_D > |N_{A, \text{CIGS}}|$ ($|N_{A, \text{CIGS}}|$: absolute acceptor concentration in CIGS) or $\Delta E_V \geq 0.15 \text{ eV}$.

CIGS and Cu(In,Ga)₃Se₅ (Cu-deficient material with Cu/(Ga+In) of 0.33) films were compared to discuss the effects of CDL formed on CIGS surface. The higher Cd content at the surface region in Cu(In,Ga)₃Se₅ than CIGS was observed when CdS layer was deposited on the top of the films. The bandgap of 1.51 eV in Cu(In,Ga)₃Se₅ was wider than that of 1.22 eV in CIGS. These results suggest a promotion of Cd-diffusion and a formation of $\Delta E_V \sim 0.29 \text{ eV}$ between CIGS and Cu(In,Ga)₃Se₅ owing to Cu vacancies in Cu(In,Ga)₃Se₅. The same cross-section of a CIGS solar cell with an efficiency of 18.5% fabricated by three-stage process was compositionally, electrically, and structurally evaluated. CDL (Cu/(Ga+In) of 0.31) with high Cd contents of 3.4at% unintentionally formed on the CIGS surface had lower carrier concentration of $4.8 \times 10^{10} \text{ cm}^{-3}$ than that of 10^{14} – 10^{16} cm^{-3} in CIGS grain interior. This implies difficulty of Cd-doping into the CIGS surface to satisfy the condition of $N_D > |N_{A, \text{CIGS}}|$ owing to insufficient activation of Cd atoms. In contrast, the formation of ΔE_V is effective to satisfy the condition of $\Delta E_V \geq 0.15 \text{ eV}$ owing to low Cu/(Ga+In) ratio of 0.31.

Suppression effect of recombination at CdS/CIGS interface by introducing Cu(In,Ga)₃Se₅ was investigated in CIGS solar cells with energy bandgap of 1.2 and 1.4 eV. Open-circuit voltage (V_{OC}) in both solar cells was clearly improved from 0.62 to 0.64 V and from 0.66 to 0.75 V, respectively, with increasing Cu(In,Ga)₃Se₅ thicknesses from 0 to 5 nm. This result suggests the effectiveness of ΔE_V at the CdS/CIGS interface by CDL formation to suppress interfacial recombination.

Growth mechanism of CDL during CIGS deposition by three-stage process was revealed, and Se irradiation process was proposed for control of CDL. Se irradiation process leads to follow effects; (i) thin and homogeneous Cu_2Se layer was formed in the second stage; (ii) Cu_2Se changes to coexistence condition of Cu-deficient Cu_{2-x}Se solid and Cu–Se liquid. A uniform CDL with a thickness of 150–200 nm was formed via control of Cu_{2-x}Se on CIGS with irradiation time of 5 min, whereas the CDL formed at an irradiation time of 10 min was rough and non-uniform.

The effect of CDL formation on performance in CIGS solar cells was investigated. V_{OC} and fill factor (FF) were successfully increased up to 0.672 V and 76.2% in a CIGS solar cell with an irradiation time of 5 min, and a maximum efficiency of 19.8% was achieved. These results suggest that the uniform CDL on CIGS leads to efficiency enhancement in CIGS solar cells. At an irradiation time of 10 min, V_{OC} and FF decreased to 0.633 V and 72.9% owing to rough and non-uniform CDL. Finally, the Se irradiation step is a simple, unique process that can boost the performance of CIGS solar cells.