ASIA-PACIFIC SOLAR RESEARCH CONFERENCE

Defect and heterojunction engineering in CZTS solar cells by nanoscale atomic layer deposited aluminium oxide

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Kesterite Cu_2ZnSnS_4 (CZTS) solar cells offer a compelling low-cost alternative to conventional thin-film solar cells, mainly due to its outstanding performance compatibility with $CuInGaSe_2$ (CIGS) and CdTe solar cells manufacturing while only consisting of earth-abundant and non-toxic constituents. To date, the record efficiency of Se-rich $Cu_2ZnSn(S_xSe_{1-x})_4$ (CZTSSe) cell is 12.6%[1], which is still substantially lower than its CIGS counterpart with a record efficiency of 22.9 % [2]. This performance gap can mainly be attributed to the large open circuit voltage (V_{oc}) deficit ($E_g/q - V_{oc}$, E_g is the band gap, q is the electron charge)[3], with a huge gap between the experimentally obtained and the theoretical maximum value. Several factors have been suggested to contribute to this V_{oc} deficit including bulk defects and band tailing in the CZTS bulk [4,5] and recombination at interface defects[3,6]. We have reported 9.3% efficiency CZTS solar cells by using Cd-free ZnSnO buffer and found that the interface recombination at CZTS/ZnSnO heterojunction is a key issue for the further performance improvement of Cd-free CZTS solar cells. Atomic layer deposition (ALD) could provide precise modification at the interface due to its self-terminating process, which enables atomic-level control over the coating thicknesses and conformal coverage over large area substrates. Aluminium oxide (Al_2O_3) deposited by ALD has been well accepted as a very effective way to passivate *p*- and *n*-type silicon through chemical and field-effect passivation [7]. The combination of these effects has led to low recombination velocities and high-efficiency devices in the silicon community. Despite the previous work with ALD-Al_2O_3 to improve the interface passivation in Cu(In, Ga)Se_2, CdTe, and CZTS solar cells, the application and mechanism of the heterojunction passivation via ALD-Al_2O_3 in CZTS/ZnSnO devices is still not investigated yet.

In this work, we demonstrated that ultrathin Al_2O_3 films deposited by ALD can effectively improve the performance of Cd-free CZTS devices with the structure of Mo/CZTS/ZnSnO/i-ZnO/ITO through heterojunction modification. With a significant gain of V_{oc} by 40 mV, we achieved a device efficiency approaching 9.8 %, the highest efficiency ever reported for Cd-free CZTS solar cells. The ultrathin Al_2O_3 layers were deposited using trimethylaluminum (TMA) and water as precursors via thermal ALD. To understand the underlying reasons for this performance improvement, we investigated the effects of each step in the ALD-Al_2O_3 process separately, i.e. we studied the effect of the TMA dosing, H_2O dosing, and vaccum annealing at the ALD process temperatures. Gains in open-circuit voltage and concomitant increases in efficiency were observed in all three cases, although the vaccum annealing benefit was not that significant. The panchromatic photoluminescence (PL) intensity increased after all the treatments of CZTS absorber, indicating reduced recombination resulting from defect passivation, which resulted in the V_{oc} improvement of the devices. This result indicates that ultra-thin ALD Al_2O_3 films improve the performance of CZTS solar cells through heterojunction passivation, and further detailed investigation is ongoing to elucidate the mechanisms behind the passivation effect.

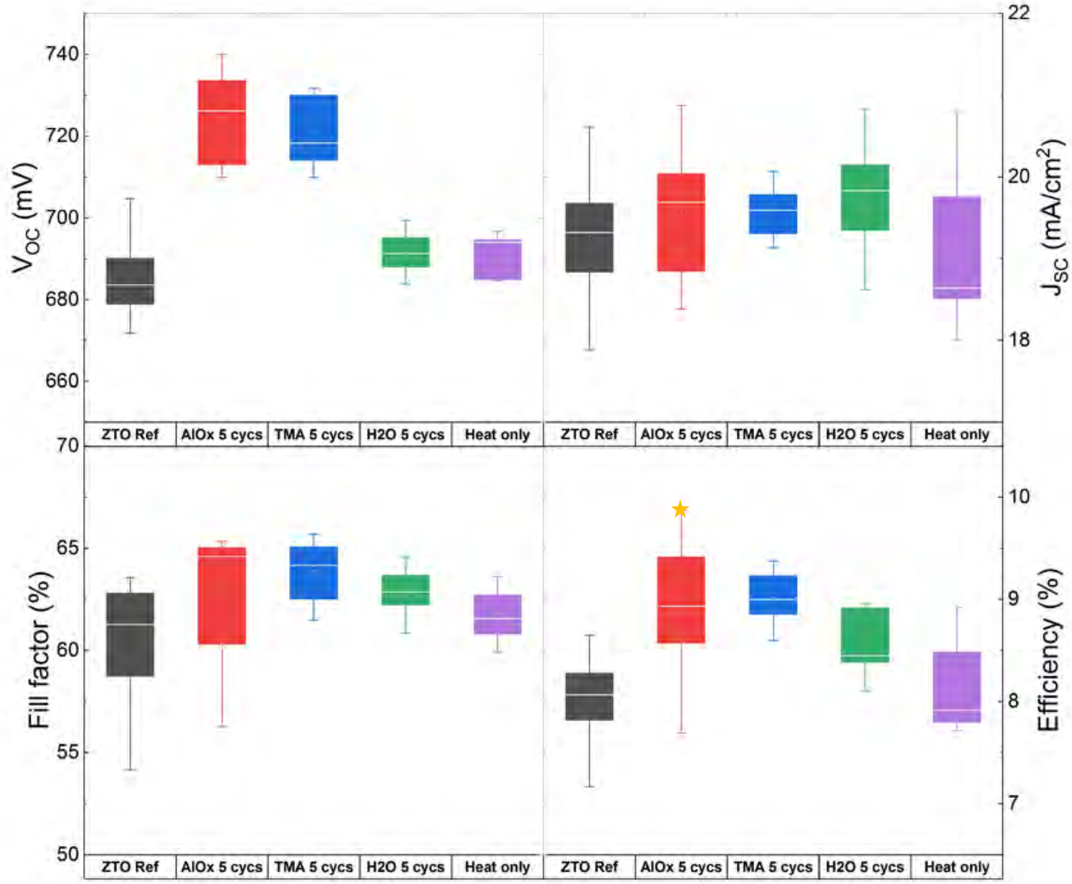

Figure 1. Box-plot diagram of the one-sun solar cell performance parameters of Mo/CZTS/ZnSnO/i-ZnO/ITO solar cells made with the heterojunction treatment of 5 cycles of Al_2O_3, TMA, H_2O via ALD, and vaccum annealing at 200 °C. Box, horizontal bars, point symbols and indicate 25/75 percentile, min/max and mean values, respectively. Results obtained for a CZTS device without any treatment are included as a reference as well.

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