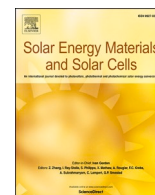




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## Chemical tailoring of sodium content for optimization of interfacial band bending and alignment in flexible kesterite solar cells

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## ABSTRACT

In this study, highly efficient and flexible Na-doped kesterite  $\text{Cu}_2\text{ZnSn}(\text{S},\text{Se})_4$  (CZTSSe) thin-film solar cells with an efficiency of over 8% were developed. A CZTSSe thin film was deposited on a flexible Mo foil using metal precursors with varying thickness of the NaF layers on the CZTSSe thin film. The sample maintained 92.2% of its performance after 1000 bending cycles with a bending radius of 12 mm. To investigate defect passivation effect of Na doping through the electrical properties of Na-passivated grain boundaries (GBs), the cells were examined by scanning probe microscopy. Downward band bending was observed at GBs in the Na-sufficient CZTSSe absorber layers; that is, Na enhanced the potential barrier at the CZTSSe GBs in the solar cells and the separation of carriers. The carrier separation increased under illuminated condition. Moreover, the surface photovoltage (SPV) of the cells was measured at laser wavelengths of 405, 532, and 640 nm using photo-assisted Kelvin probe force microscopy to investigate the photogenerated carrier transport in Na-passivated CZTSSe solar cells. The Na content strongly affected the photo-induced changes in the carrier behavior. A cell with a sufficiently Na-doped CZTSSe absorber layer exhibited a large SPV with a maximum value of 46 meV under illumination, whereas Na-deficient samples showed a lower SPV, indicating that Na passivates defects that act as recombination sites. A photo-induced defect passivation effect was observed in a cell with optimal Na content under 405 nm wavelength illuminated condition, which resulted in charge accumulation at the absorber surface.

## 1. Introduction

Kesterite ( $\text{Cu}_2\text{ZnSn}(\text{S},\text{Se})_4$ , CZTSSe) is a promising photovoltaic absorber material because of its suitable physical properties such as a direct energy band gap that is tunable from 1 to 1.5 eV by adjusting the ratio of S to Se, and high absorption coefficient [1–3]. However, the highest power conversion efficiency (PCE) achieved by CZTSSe thin-film solar cells to date is 12.6%, whereas that of  $\text{Cu}(\text{In},\text{Ga})\text{Se}_2$  (CIGS) thin-film solar cells is 23.35% with a CdS buffer layer and 22.9% without the CdS buffer layer [4–6]. However, despite the high efficiency of the CIGS thin-film solar cells, their use of In and Ga limit their wide application. By replacing In and Ga in CIGS with Zn and Sn, which are

relatively inexpensive and abundant, i.e., using CZTSSe instead of CIGS, the manufacturing cost of thin-film solar cells can be lowered [7,8]. Furthermore, attempts have been made to replace the rigid substrates of solar cells with thin, lightweight flexible substrates for wide application. For example, a CIGS solar cell on a flexible polyimide (PI) substrate with a PCE exceeding 20% has been reported [9].

Flexible glass, stainless-steel, PI, and metal foils, such as Mo, Ti, and Al foils, are considered as promising substrate materials for use in flexible kesterite thin-film solar cells. Among them, the metal foils possess high conductivity, which can lead to the development of a low-cost, roll-to-roll production. In particular, the Mo foil has attracted attention owing to the lack of metallic impurities and its compatible

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