# **Supporting Information**

# Improving the open circuit voltage through surface oxygen plasma treatment and 11.7% efficient Cu<sub>2</sub>ZnSnSe<sub>4</sub> solar cell

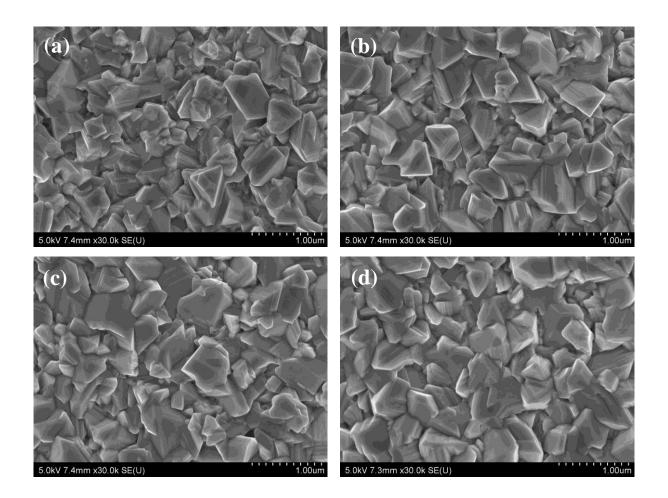
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## The time dependence of the surface oxygen plasma treatment



**Figure S1.** SEM images of CZTSe surface after the surface oxygen treatment as a function of treatment time, 1 min (a), 3 min (b), 10 min (c), 30 min (d).

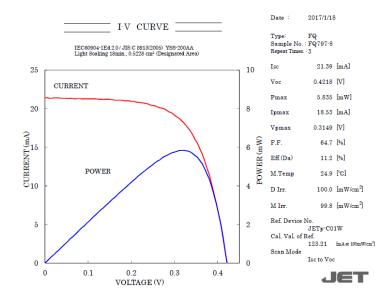
Figure S1 shows SEM images of CZTSe surface after the surface oxygen treatment as a function of treatment time, 1 min (a), 3 min (b), 10 min (c), 30 min (d). It was found that the morphology and contrast of the surface were almost identical. This implies that the CZTSe surface was homogeneously oxidized both in-plane and perpendicular to the surface plane.

#### EQE spectra calculation using e-ARC software

The EQE spectra were simulated using the e-ARC software. In this study, two structures were used as follows, ZnO:Ga (350 nm n=1.1×10<sup>20</sup> cm<sup>-3</sup>)/i-ZnO (50 nm)/CdS (50 nm)/CZTSe (1.8  $\mu$ m)/ MoSe (500 nm)/Mo, and MgF<sub>2</sub> (130 nm)/ZnO:Ga (350 nm n=1.1×10<sup>20</sup> cm<sup>-3</sup>)/i-ZnO (50 nm)/CdS (25 nm)/CZTSe (1.8  $\mu$ m)/MoSe<sub>x</sub> (500 nm)/Mo, for the comparison of treatment time, and the best performing cell, respectively. ZnO:Al was used in this work, although ZnO:Ga, whose optical properties are prepared in the software, was used in the simulations for a better agreement with the experimental results.

### Certified J-V curve of the best-performing cell in JET

The certified *I-V* curve by Japan Electrical Safety & Environment Technology Laboratories (JET) is shown in Figure S2. The conversion efficiency was 11.2%, and which is slightly smaller compared to the reporting 11.7% measured in-house. The efficiency measured in-house just before the measurement in JET was same as 11.2%, the deviation is considered to be due to the degradation by aging of the sample.



**Figure S2.** the certified I-V curve by JET. 11.2% was obtained, which was same as measured in-house with 11.2% just before the measurement in JET.