

Supporting Information for Wide-Eg $\text{CuGa}(\text{S},\text{Se})_2$ Thin Films on Transparent Conductive Fluorinated Tin Oxide Substrates as Photocathode Candidates for Tandem Water Splitting Devices

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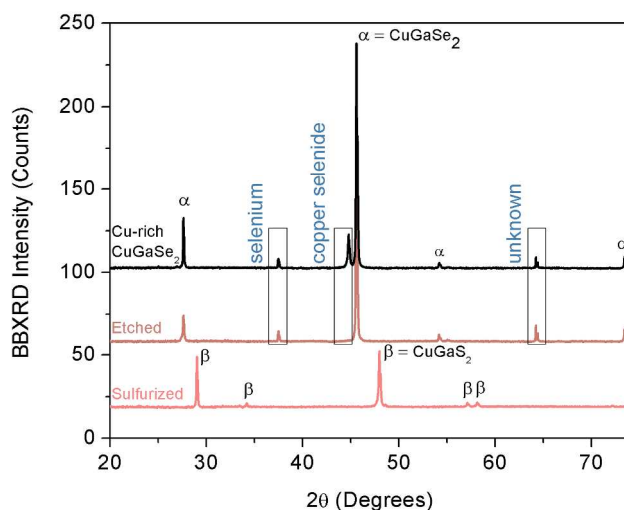


Figure S1) Bragg-Brentano XRD scans probing the bulk of a Cu-rich CuGaSe_2 sample before etching (top), after etching (middle), and after sulfurization (bottom). All phases closely matched to either CuGaSe_2 (Powder Diffraction File, 035-1100), selenium (PDF 00-054-0500), copper selenide (PDF 027-1131), or CuGaS_2 (PDF 025-0279) except for two unknown peaks at 64.24 and 64.40 degrees, which did not match any combination of copper, gallium, selenium, oxygen, and sodium.

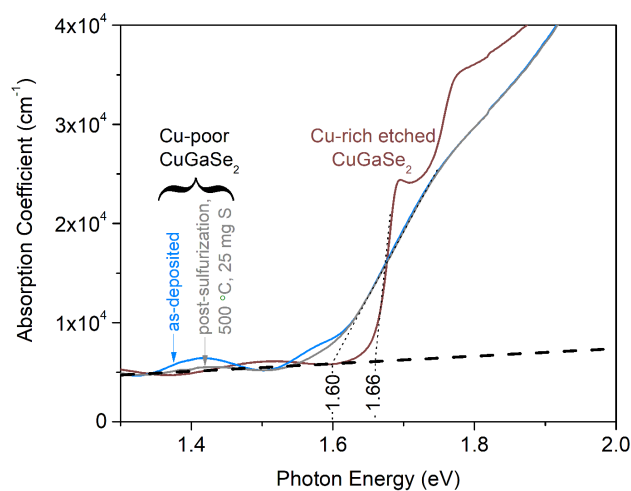


Figure S2) Plot of absorption coefficient showing that the one-step sulfurization does not increase the bandgap of Cu-poor CuGaSe_2 . As-deposited CuGaSe_2 is plotted to show the slight differences in bandgap between Cu-rich and Cu-poor CuGaSe_2 as measured by UV-vis.

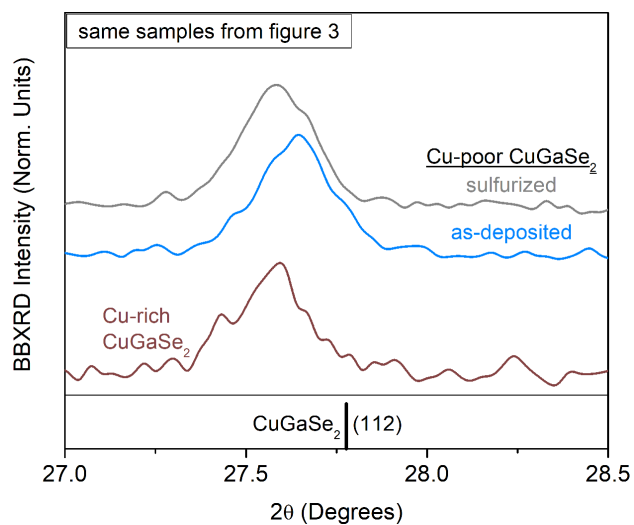


Figure S3) Plot of XRD data for the same samples of figure S1 showing that Cu-poor CuGaSe_2 (112) peak did not shift as a result of the sulfoselenization. As-deposited Cu-rich CuGaSe_2 was plotted as well, showing little difference from the Cu-poor sample.

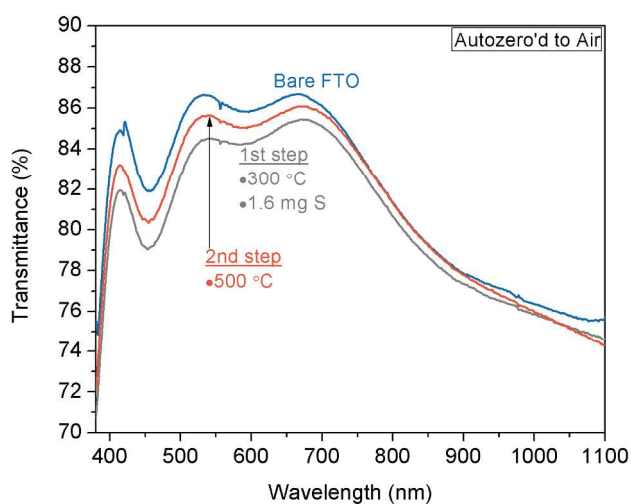


Figure S4) UV-Vis transmittance data of the bare FTO (control) before the 1st step sulfurization, after the 1st step, and after the 2nd step annealing in nitrogen. The FTO dropped approximately 2 percentage points in transmittance after the 1st step, then increased approximately 1 percentage point, showing that the two-step annealing had little effect on the optical transmittance of FTO.

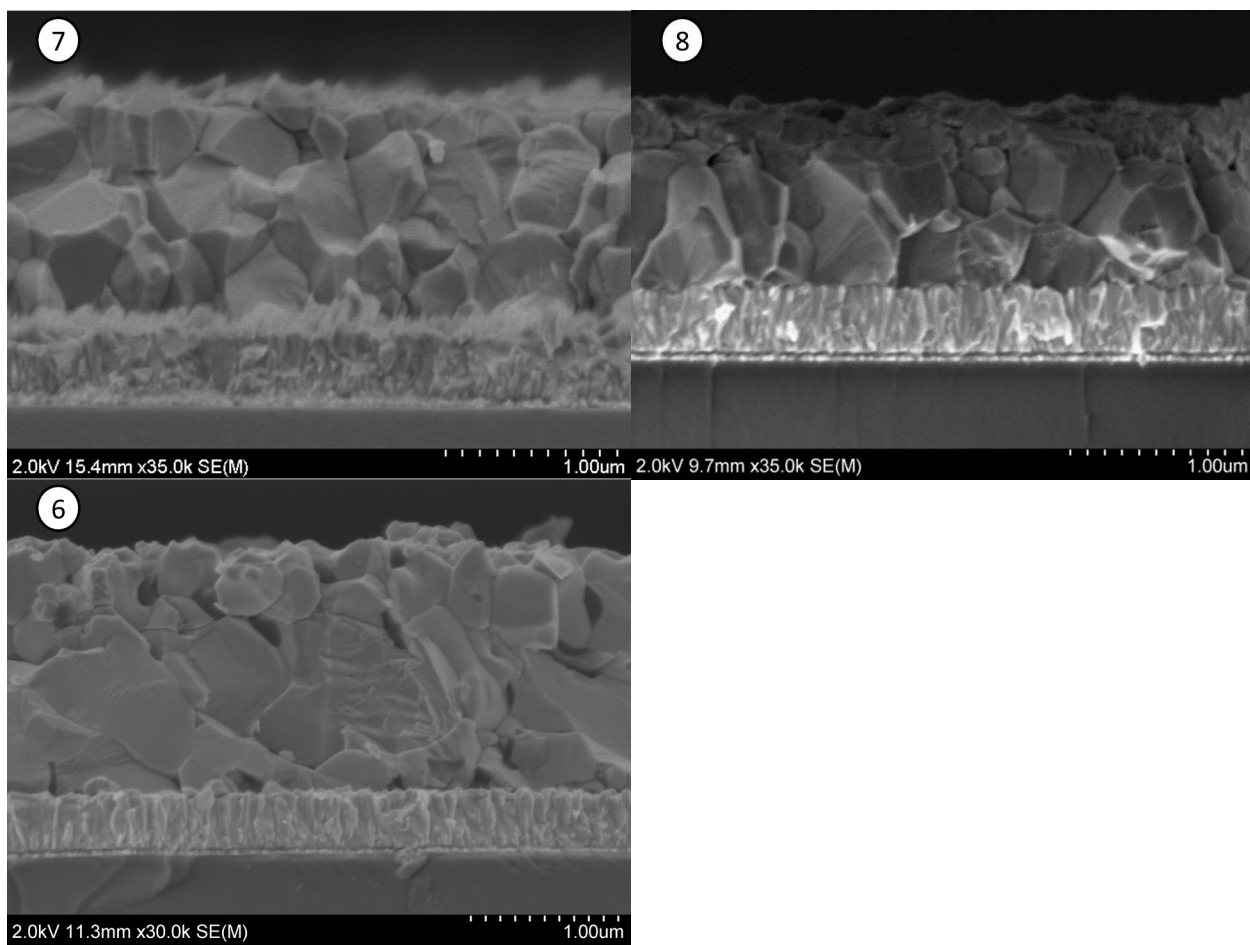


Figure S5) Cross-sectional SEM images showing that samples 6, 7, and 8 have similar microstructures.