Supporting Information

A Low-temperature Formation Process to high efficiency Se-free Cu_2ZnSnS_4 solar cells fabricated through sputtering and sulfurization

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Figure S1. The comparison of device statistics between the slow ramping and the Low-temperature Formation Process. The Low-temperature Formation Process clearly produces a jump in the performance, giving average PCE of 8%, as compared to 6% in the ramping method.

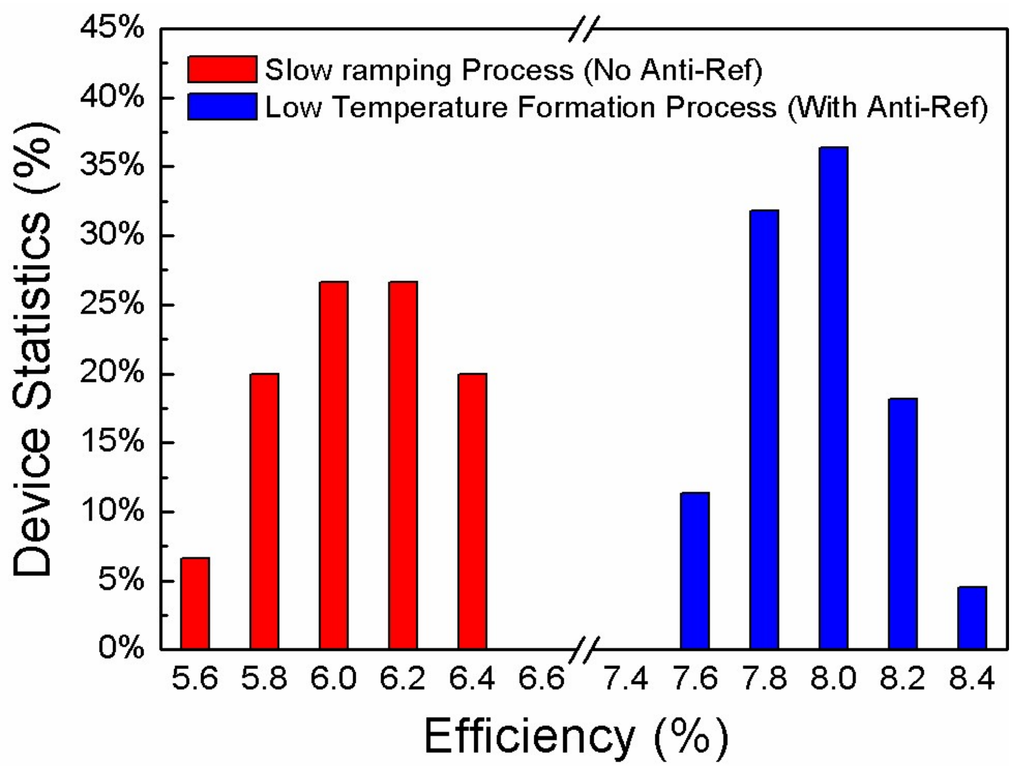
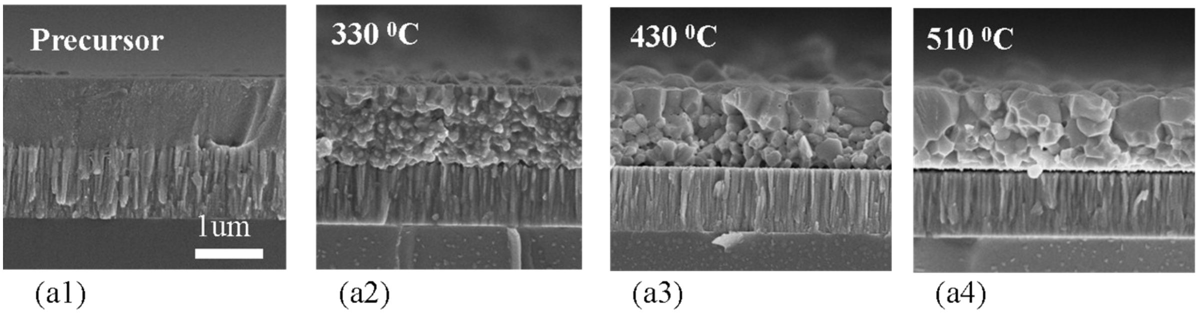


Figure S2. The cross section SEM figures of the CZTS thin films which are measured by terminating the temperature ramping process of P1 and P2 at the temperatures as indicated. (a1) is the image of the precursor. (a2)-(a4) show the evolution of the morphology of the thin films following P1 process. Large grains are observed to emerge from the surface layer. More and more small grains in the bottom layer are absorbed by the large grains when the temperature is increased for P1 process. (b1)-(b4) show the evolution of the morphology of the thin films following P2 process. No difference in crystal size for the bottom layer and the top layer is observed. Small grains are found to merge into larger ones when the temperature is higher or annealing time is longer.

Ramping profile P1



Low-temperature Formation Profile P2

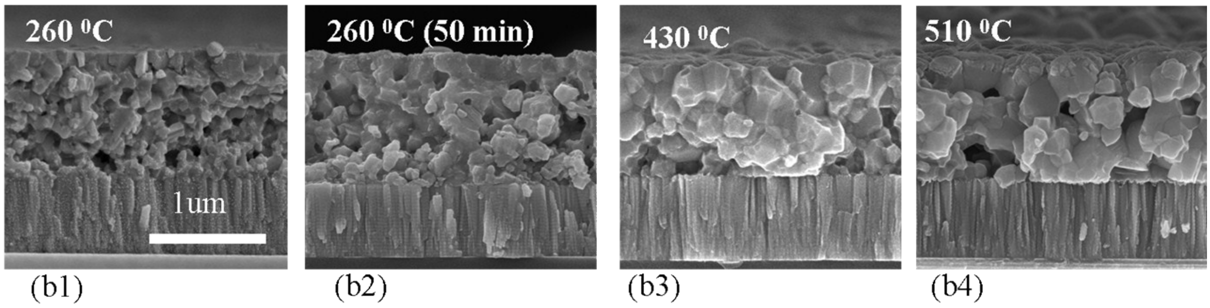


Figure S3. The intensity of CZTS (220) diffraction peak at q ~ 3.23Å^{-1} at different stages. Since ZnS and Cu_2SnS_3 (CTS) do not have diffractions at this q-value, the intensity could be attributed solely to the CZTS quaternary phase. The similarity between this figure to Figure 2b suggests that the evolution of the main peak at q ~ 1.97Å^{-1} mainly comes from the CZTS (112) reflection.

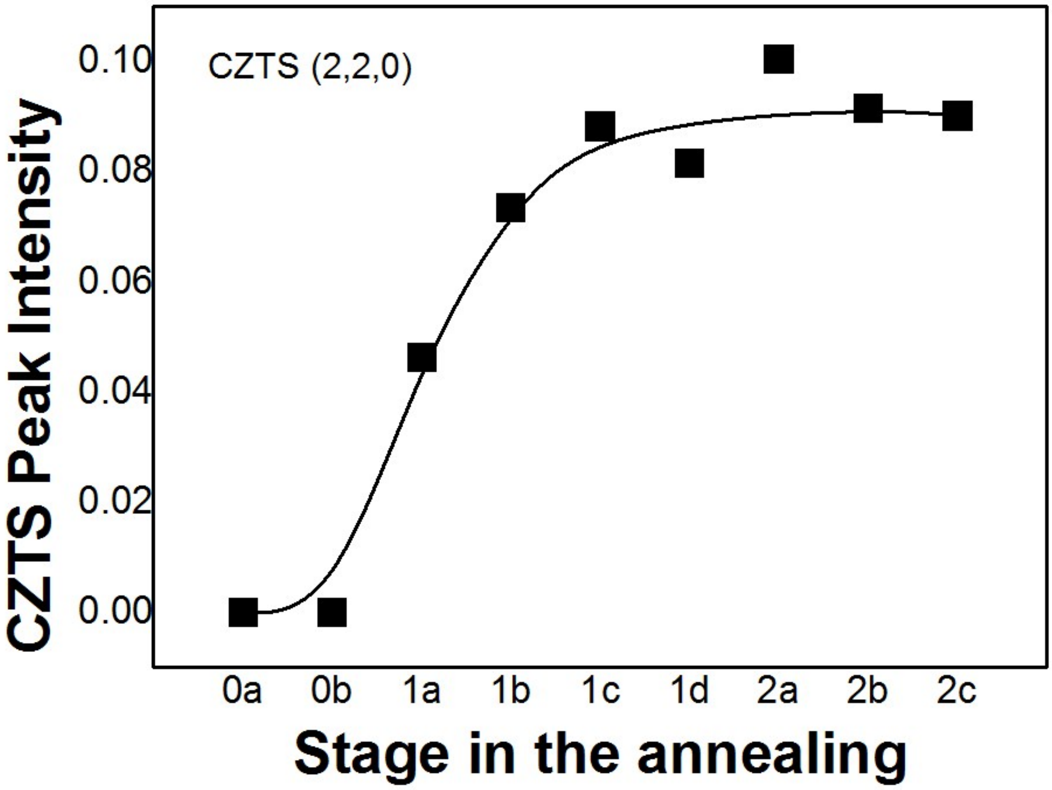


Figure S4. (a) A photo of the CZTS sample we made in our laboratory, (b) the picture of one of the devices on the sample. The white fingers are Al electrodes. The total area of each device defined by mechanical scribing is measured using a microscope with a digital camera.