

Vapor-Phase Incorporation of Ge in CZTSe Absorbers for Improved Stability of High-Efficiency Kesterite Solar Cells

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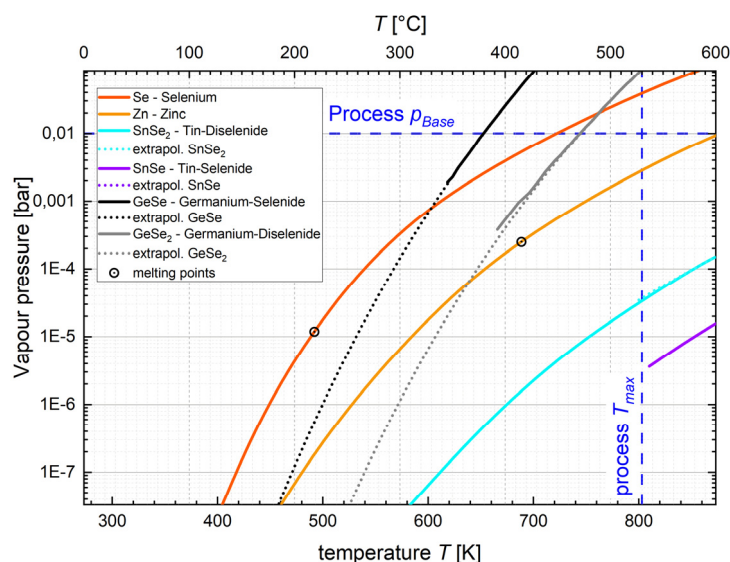


Figure S1. Vapor pressure curves for relevant compounds. Data source from [1]–[4].

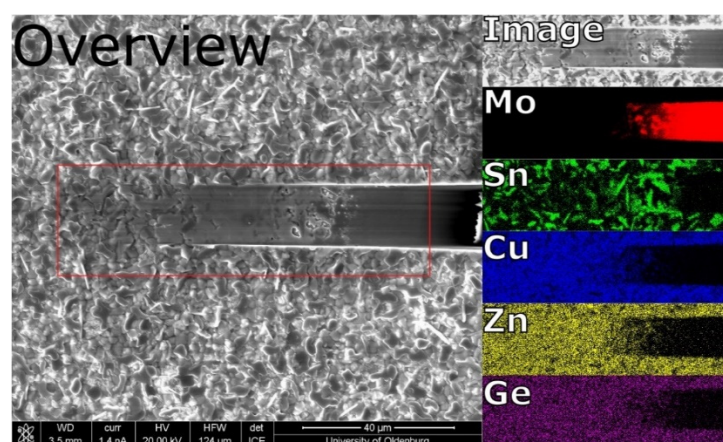


Figure S2. Shallow angle FIB cut performed on an inhomogeneous sample to perform a depth resolved EDX mapping for Sn and Ge distribution analysis.

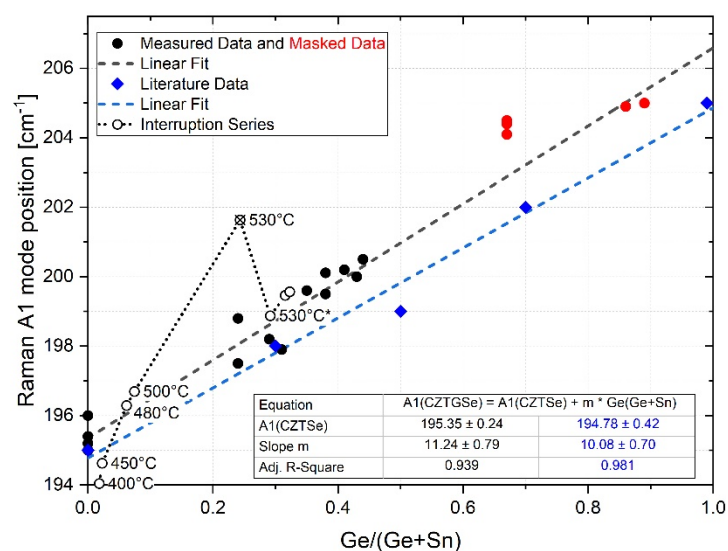


Figure S3. Raman A1 peak position vs. Ge/(Ge + Sn) ratio for the produced samples and a comparison to literature values from [5]. The sample labelled with 530 °C from the interruption series does not follow the trend and is therefore expected to have a gradient.

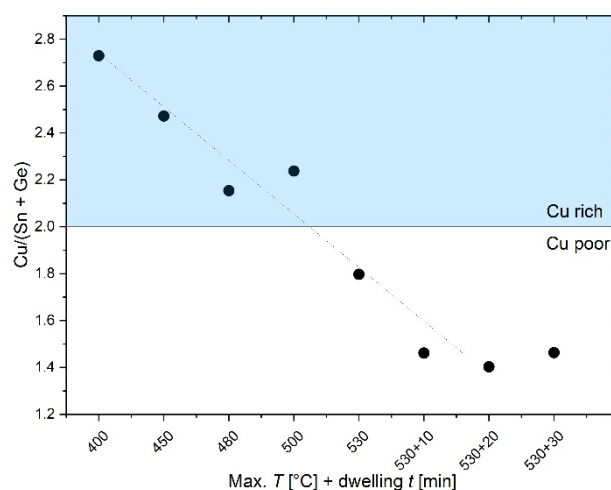


Figure S4. Cu/(Ge + Sn) ratio development for the performed interruption series. The switch from Cu/(Sn + Ge) > 2 to < 2 corresponds to the in-process composition shift from Cu-rich to poor via GeSe incorporation.

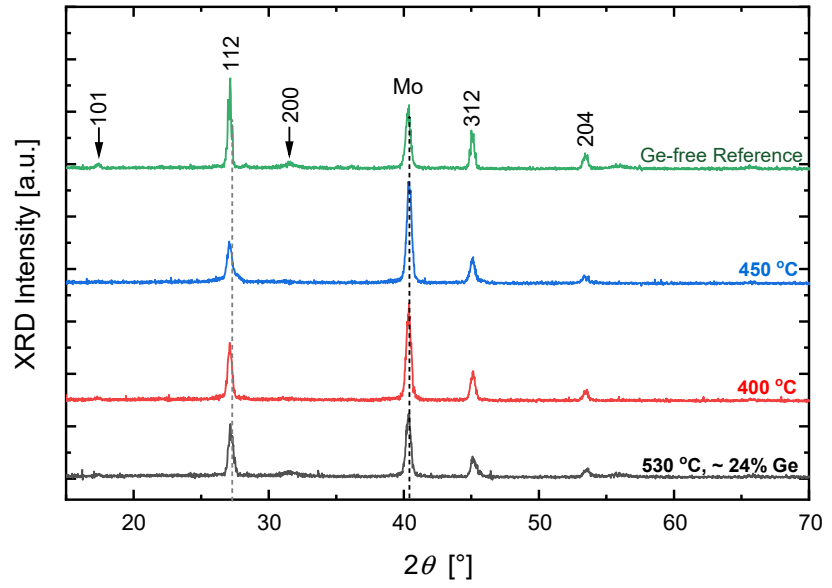


Figure S5. X-ray diffractogram of the samples processed: reference (no Ge) and Ge containing samples interrupted at different temperatures.

X-ray diffraction patterns of the reference and Ge incorporated samples are shown in Figure S7. The reference sample shows the main peak intensity corresponding to 112 plane (2θ centered around 27.14°) corresponding to the $\text{Cu}_2\text{ZnSnSe}_4$ phase. Samples annealed at 400°C and 450°C show a very slight peak shifting (i.e., 2θ at 27.16°) indicating a similar Ge content, and the sample annealed at 530°C shows higher Ge content (i.e., higher peak shifting, 2θ at 27.25°), corroborating the observation from Raman and EDX measurements. All the samples show the peaks corresponding to kesterite phase with no signatures of SnSe_{2-x} and GeSe_{2-x} phases.

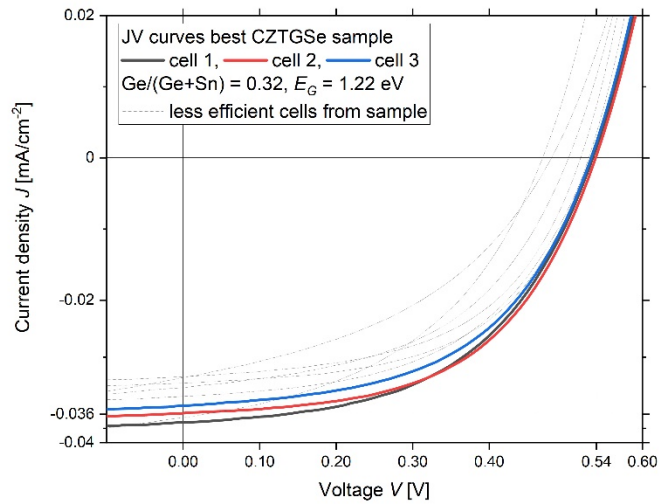


Figure S6. Highest PCE IV curves for sample with the best cell from the study.

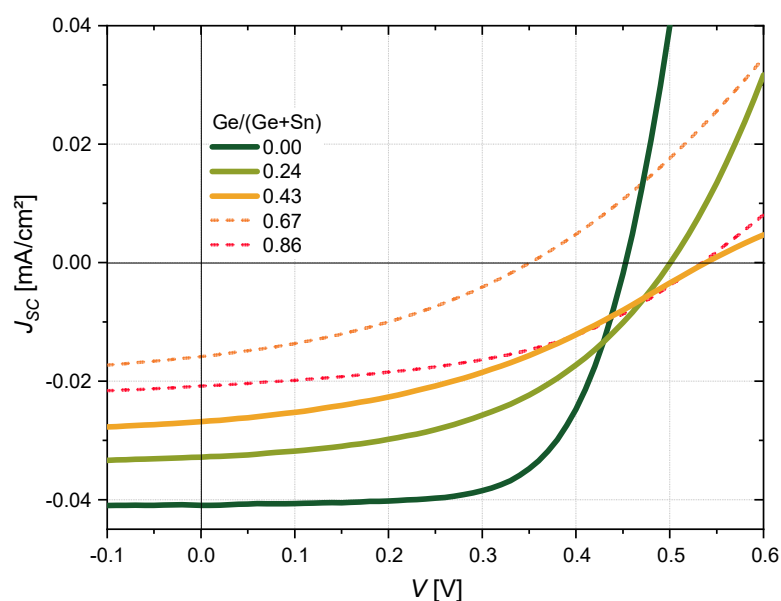


Figure S7: IV curves of the best samples for Ge/(Ge+Sn) groups shown in the main text in Figure 3. Dashed lines correspond to the masked data mentioned in the main text.

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