

## Title: Kinetic aspects of wide band gap CuIn<sub>1-x</sub>Ga<sub>x</sub>Se<sub>2</sub> thin-film solar cells

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Increasing the Ga content in the CuIn<sub>1-x</sub>Ga<sub>x</sub>Se<sub>2</sub> (CIGSe) alloy permits to adjust the device band gap energy to the theoretically optimum Eg=1.4eV and attain the maximum conversion efficiency for x~0.7. However, the experimental solar cell efficiencies do not follow the theoretical predictions, since many experiments have shown that the efficiency decreases for x>0.4.

Different scenarios concerning structural and chemical modifications of the CIGSe film have been proposed to explained the decreased efficiency at high x, but the gap between the theoretical and experimental performance for large-x CIGSe absorber layer is still under debate.

Our study on the thermodynamic and kinetic properties of the CIGSe alloy reveals that during the CIGSe co-evaporation process i) the alloy does not reach the thermodynamic equilibrium and ii) the slow kinetics at high Ga ratio leads to a detrimental accumulation of a Cu-Se compound at the inter- or intra-grain regions.

To further understand the limited performance for large x, we focus our study on In-free CuGaSe<sub>2</sub> layers and we examine two different co-evaporation methods a) a standard Cu-RO (Rich-Off) and b) a modified Cu-RO process, where a relaxation step of 60min is introduced during the co-evaporation process.

Using the micro-Raman spectroscopy at excitation wavelength (633nm) close to the band gap energy of CGSe, we investigate the difference in the local composition at the surface and at the interface between CGSe/Mo.

Coupling Raman, XRD, and EDX analyses, we demonstrate that the slow kinetics at high Ga ratios leads to a detrimental accumulation of a Cu-Se compound at the inter- or intra-grain regions.

Our work on the CGSe kinetic properties gives a clear evidence that i) a longer co-evaporation process and b) higher deposition temperature are needed to improve the solar cell efficiency when large x CIGSe absorbers are used.