The power of the crowd? Does a 25'000 kesterite samples database bring us to a new place?

Leo Choubrac, Ässia Boukhtami, Taiseer Jokadar, Lars Steinkopf, Thomas Unold

Helmholtz Zentrum Berlin – Department Structure and Dynamics of Energy Materials

Despite important efforts to understand kesterite solar cells performances limit and to overcome it (via alloying, doping, revisiting synthesis route as well as developing alternative buffer/back contact material), IBM 12.6% record efficiency remains unbeaten for not less than 8 years[Wang, 2014]. It makes consensus that the limiting factor is a short carriers lifetime resulting in high Non-Radiative recombination losses (NR-losses) and ultimately poor open-circuit voltage (V_{OC}), with respect to the Schockley-Queisser limit. But the underlying root for these high non-Radiative losses remains unclear, and a vast majority of the literature focuses on improving devices V_{OC} or efficiency rather than reducing NR-losses. Consequently, some efficient strategies to improve the core-problem may have been already experienced but filtered out due to deleterious side-effect at the device scale.

Factors demonstrated or suggested to influence V_{OC}, NR losses or lifetime includes composition, structural disorder, doping (through alkali), presence of secondary phases and selenisation condition (nature of atmosphere, temperature, duration, partial pressures).

The fact that changing one of those factors usually affect others makes it extremely difficult to decorrelate the effect of each of them. Moreover, optimum composition may shift depending on those parameters [Haass,2017]. Finally, clear conclusions require variations higher than the process reproducibility fluctuations, which is possibly important for kesterite synthesis.

In order to clarify on the role of those factors while avoiding device-making related issues, reproducibility concerns and circumvent composition drift issues, we embarked on a combinatorial analysis of kesterite material.

CZTSe thin-films were synthesised on transparent substrate (SLG/Na-Barrier coated by NaF) by the simultaneous deposition of Cu,Sn (by ebeam) and ZnSe(thermal evaporation) precursors, followed by selenisation in a graphite box. The precursors sources are placed in a way leading to lateral compositional gradient over the substrate. The film is then (virtually) separated in 575 areas, corresponding to 575 samples synthetised together -limiting the questions related to process fluctuation- but having compositions spreading over a large part of the phase diagram. Over 40 of thus films have been prepared, resulting in a database of more than 25'000 samples.

Routine characterisations set starts with composition measurement by XRF, secondary phases detection by Raman spectroscopy and Near-Infrared Imaging. Transmission/Reflection mapping and calibrated photoluminescence imaging permit to determine the bandgap, calculate the NR losses, and the QFLS (quasi Fermi Level splitting). Those optoelectronical properties are measured twice (after 2 different controlled low-temperature annealing procedures) in order to take into consideration the role of the structural disorder.

The data analysis allows us to clarify the role of composition, doping and selenization conditions on NR losses are QFLS, which are the intrinsic properties of kesterite limiting solar cells efficiency.

The focus of the presentation is not to present extensive results on the effect of each particular process parameter, but rather to select few critical examples showing the advantages and the limits of such approach (important number of samples, limited number of characterization tools, need to develop a methodology and to analyse large dataset)