**The $Cu_ySe$ behavior in the $CuIn_{1-x}Ga_xSe_2$ thin-film solar cells :**

**A possible reason of the limited performance for high x ?**

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Thin-film solar cells based on polycrystalline $CuIn_{1-x}Ga_xSe_2$ (CIGSe) absorber layers offer tunable optical band gap energy ($E_g$) by changing the Ga ratio (i.e. x=[Ga]/([In]+[Ga])) of the CIGSe thin film (from $E_g$=1eV to 1.7eV, for x=0 and 1 respectively)[1]. So far, best labscale energy conversion efficiencies (22.6%[2]) are achieved for x around 0.3, which corresponds to incident light absorption onsets around 1.15eV. This latter experimental result is in contrast with the theoretical value lying near x=0.75 ($E_g$=1.4eV) for a single junction under AMG1.5.

Our study seeks to adress the reason of the limited CIGSe performance when x exceed the threshold value of 0.4. Coupling computational and experimental approaches, we deduce two possible scenarii related to the different Cu behavior in the CIGSe internal interfaces, at low and high Ga ratios (x).

*Scenario A* concerns the role of the CIGSe internal interfaces and the segregated element at the vicinity of the grain boundaries (GBs). It seems that the nature of the predominant segregated species at the GBs varies with x. This is consistent with i) our theoretical-based model of segregation driving forces that combine ab initio and statistical thermodynamics[3], and ii) recent experimental results obtained by APT[4] (Atom Probe Tomography) showing Cu enriched GBs for large x and Cu poor GBs for low x.

*Scenario B* involves the detrimental copper-selenide ($Cu_ySe$) phases. This compound can be accumulated at the GBs or/and within the grains. Our XRD, RAMAN and EDS analyses demonstrate that the $Cu_ySe$ behavior differ, at low and high x. Furthermore, DFT calculations of a $Cu_2Se$ compound in a CISe and CGSe matrix, raise questions about the $Cu_ySe$ intermixing in the CIGSe environment.

The nature of the accumulated species or compound at the CIGSe internal interfaces can be detrimental or beneficial for the solar cell efficiency. Hence, in this study, we examine both experimentally and theoretically these two scenarii.

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