Impacts of Ga evaporation rate on Cu(In,Ga)Se2 growth at low temperatures: from a three-stage to a multi-stage process

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Cu(In,Ga)Se2-based (CIGS) solar cells have achieved the highest efficiencies (up to 22.6% on soda lime substrate[1] and 20.4% on polyimide substrates[2]) among the thin film solar technologies. The use of polyimide substrates broaden the scope of applications for which the weight of the solar panel remains an issue such as Building Integrated PV. Polyimide substrates cannot stand high deposition temperature (<450°C). Because of lower process temperature, lower diffusion of copper, indium and gallium are observed. It implies a strong composition gradient with a classic coevaporation three-stage process. An optimization of this process is necessary to have a better control of the Ga gradient.

In this study, we focus on the Ga distribution in the CIGSe layer and its impacts on the cell properties. Three main sets of experiments were realized (referenced as 1, 2 and 3 in the figure). Gallium profiles were systematically characterized by Glow Discharge Optical Emission Spectroscopy (GDOES).

In a first part (1), we varied the Ga flux during the first stage of the deposition process. According to XRD measurements, with an higher Ga rate, an additional peak was found for the (220)/(204) orientation corresponding to CuIn0.75Ga0.25Se2. Moreover, small grains close to the Mo/CIGS interface were observed by SEM measurements. We obtained higher VOC and JSC for a higher Ga rate due to the back surface field effect.

Then (2), Ga was added during the second stage changing the growth of the material compared to a standard three stage process. In order to understand the new growth mechanisms, samples were pulled out of the deposition chamber at different moments of the deposition process. Raman spectroscopy as well as XRD were performed to identify the intermediate phases. In situ substrates temperature measurements (thermocouples) do not exhibit identical temperature variations between the processes with and without Ga. This suggests the growth of new phases during the process. The morphology of the different samples was also analyzed by SEM. On the whole, Ga adding increased the band gap energy (calculated from External Quantum Efficiency - EQE measurements) leading to an increased VOC and therefore a lower JSC.

Finally (3), the impact of Ga rate in the last stage of the deposition process was investigated. Grazing Incidence XRD and Raman spectroscopy were used to estimate the rate of Ga at the top of the CIGSe layer. Surprisingly, a stronger gradient at the front interface exhibits the highest solar cells performances because of a higher VOC (up to 710 mV) and FF (72.2%).

As a result of this study, we achieved a 16.8% solar cell efficiency (without an anti-reflective coating). It represents +2.3% on the FF, +60 mV on the VOC and +1.8 mA on the JSC compared to a non-optimized Ga process.

![Standard three-stage process (left) and new multistage process (right) with the different Ga evaporation rates tested](image)