Impacts of Ga evaporation rate on $Cu(In,Ga)Se_2$ growth at low temperatures: from a three-stage to a multi-stage process

V. Achard[1,3,4], J. Posada[1,3,4], M. Balestrieri[4], L. Lombez[2,3,4], M. Jubault[1,3,4], D. Lincot[2,3,4] & F. Donsanti[1,3,4]

[1] *EDF R&D, 6 quai Watier, 78400 CHATOU Cedex, FRANCE*
[2] *CNRS, 6 quai Watier, 78400 CHATOU Cedex, FRANCE*
[3] *IRDEP, 6 quai Watier, 78400 CHATOU Cedex, FRANCE*
[4] *IPVF, Institut Photovoltaïque d'Ile de France, 30 RD 128, 91120 PALAISEAU, France*

*e-mail : valentin-va.achard@edf.fr

$Cu(In,Ga)Se_2$-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 Dischage 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 $CuIn_{0.4}Ga_{0.6}Se_2$. Moreover, small grains close to the Mo/CIGS interface were observed by SEM measurements. We obtained higher V_{OC} and J_{SC} 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 V_{OC} and therefore a lower J_{SC}.

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 V_{OC} (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 V_{OC} and +1.8 mA on the J_{SC} compared to a non-optimized Ga process.

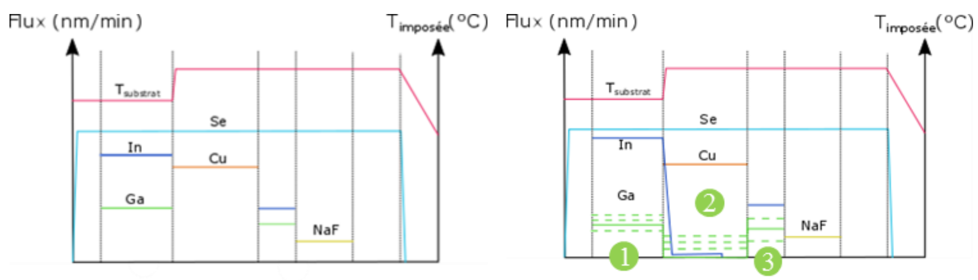

Figure 1 Standard three-stage process (left) and new multistage process (right) with the different Ga evoporation rates tested

[1] P. Jackson, R. Wuerz, D. Hariskos, E. Lotter, W. Witte, et M. Powalla, « Effects of heavy alkali elements in Cu(In,Ga)Se2 solar cells with efficiencies up to 22.6% », *Phys. Status Solidi RRL – Rapid Res. Lett.*, vol. 10, nº 8, p. 583-586, 2016.

[2] A. Chirilă *et al.*, « Potassium-induced surface modification of Cu(In,Ga)Se2 thin films for high-efficiency solar cells. », *Nat. Mater.*, vol. 12, nº 12, p. 1107-1111, déc. 2013.