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Flexible Cu(In,Ga)Se_2 Thin Film Solar Cells and Challenges for Low Temperature Growth

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1. Introduction

Among the available thin film solar cell technologies, devices based on Cu(In,Ga)Se_2 (CIGSe) are the most efficient. Recently a conversion efficiency of 20.1% has been demonstrated on rigid glass substrates [1]. Process temperatures of ~600° C and higher are needed for the deposition of such high quality CIGSe thin films [2]. In order to transfer these high standard processes to flexible substrates two main issues have to be dealt with: the need for an external supply of Na to the growing - or grown - CIGSe absorber layer and, in many cases, the use of reduced process temperatures. It is an accepted fact, that the presence of Na enhances the performance of CIGSe thin films when used as absorbers in thin film solar cells [3]. Most commonly either metal or polyimide (PI) foils are used as flexible substrate materials. While a glass substrate can inherently supply Na by diffusion through the Mo back contact during the high temperature CIGSe deposition process, sodium has to be added extrinsically when using metal or PI foils as substrates. In addition the low tolerance to thermal stress of PI so far only allows the use of process temperatures of up to ~420° C. However, the fact that PI is a good insulator makes monolithic integration a feasible option for module integration. Of course metal foils will generally tolerate high process temperatures, but for monolithic integration to be applied an insulating barrier layer needs to be deposited prior to the molybdenum back contact [4]. The best reported conversion efficiencies for flexible substrates so far are 17.4% on a metallic substrate [5] and 15.9% on PI foil [6].

From the available deposition methods for CIGSe two approaches seem to be most common: the sequential process, which relies on the selenization of metallic precursors [7], and CIGSe co-evaporation, a process that relies on the simultaneous evaporation of Cu, In and Ga in a Se atmosphere [8] and, so far, results in absorber layers, which produce solar cells with higher conversion efficiencies than those from sequential processes. Regarding the fabrication of flexible devices both approaches - along with ink based printing or paste-coating techniques - are currently being developed in view of industrial roll-to-roll processing. High and low process temperature approaches on metal and PI foils are being followed. Hence the main challenge for low temperature CIGSe deposition processes is to match the absorber material quality in terms of resulting device efficiency to that of high temperature processes. Consequently the use of co-evaporation as fabrication method seems to be the logical choice in order to develop a high η, low temperature CIGSe process. It should be added that the availability of a low temperature process is not only relevant for the deposition on PI substrates, but also with respect to applications such as the realization of an efficient superstrate configuration or, eventually, the fabrication of chalcopyrite based tandem device structures, i.e. in all the cases where a less temperature stable substrate is being used.

2. Low Temperature CIGSe Co-Evaporation

The work, that is described here, relies on CIGSe thin films that are fabricated by a multi-stage co-evaporation process [9], utilizing laser light scattering and pyrometry as in-situ process controls [10]. The basic flux scheme and substrate temperature profile for such a process are depicted in Figure 1. Initially a layered $(In,Ga)_2Se_3$ precursor layer is deposited onto a Mo coated substrate at a nominal substrate temperature T of 330° C. Then Cu and Se are evaporated onto the substrate while the temperature is increased to T_{MAX}. Once the thin film is Cu-rich, i.e. the [Cu]/([In]+[Ga])-ratio (Cu/III) reaches 1.15, the final stage of the absorber deposition process ensures an overall Cu-poor layer composition. Devices are fabricated by the application of a CBD CdS buffer layer, a transparent i-ZnO/ZnO:Al front contact and a Ni/Al front contact grid. Characterization of thin film properties and electronic device performance is described elsewhere.

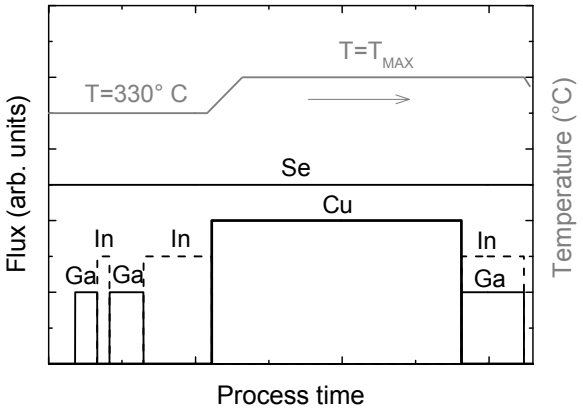

Figure 1: Element fluxes and substrate temperature for the HZB low temperature multi-stage CIGSe co-evaporation process.

3. Challenges for Low Temperature Growth

Initially the influence of T_{MAX} on the properties of the CIGSe thin film and the resulting solar cell had been studied on float glass substrates [11]. A clear impact of the process temperature on the final distribution of In and Ga along the depth of the thin film was observed. However, for a nominal T_{MAX} as low as 330° C a conversion efficiency of 11.8% was reported.

Once a Na-free substrate was being used the effect of the amount of Na present during the CIGSe thin film deposition could be explored. For these studies NaF precursor layers of different nominal thicknesses have been evaporated onto the Mo back contact prior to the chalcopyrite deposition. The general truths that Na affects the electronic properties of the resulting CIGSe thin film, the interdiffusion of the In and Ga within the growing layer and also the morphology of the final film could be affirmed. However, it became clear, that a delicate interplay exists between the added amount of Na and individual process parameters such as T_{MAX} and the final Cu/III ratio regarding the overall quality of the resulting CIGSe absorber [3,6,12]. The interplay of Cu-deficiency and Na content in particular seems to play a relevant role regarding the CIGSe thin film morphology [13].

A further effect of the Na - at least when added via deposition of a NaF precursor - was detected at the CIGSe/Mo back interface of the device, where, depending on the amount of NaF deposited, the formation of $MoSe_2$ is affected [14].

All in all it seems evident that at low process temperatures the process window for the deposition of high quality CIGSe absorber layers is potentially smaller and process control therefore required to be more refined than for high temperature processes. So far the approaches used here, led to a maximum certified efficiency of 15.9% for a 0.95 cm² total area device [6].

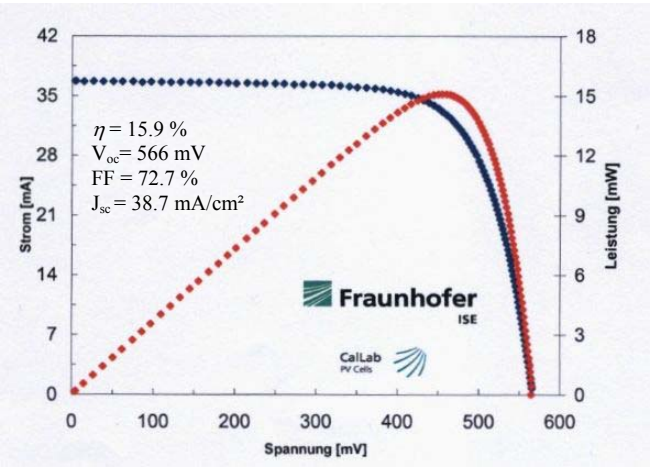

Figure 2: Externally certified performance characterization of a low temperature CIGSe thin film solar cell on PI foil (cell area 0.95 cm², with MgF_2 antireflective coating) [6].

4. Further Topics

The efficiency of devices based on CIGSe absorbers deposited by low temperature processes needs to be further improved. Aspects needing more thorough investigation and optimization include the method of Na supply and the interplay of the effect listed here with the Ga-content of the CIGSe absorber. The role of the Se-flux during film growth at low process temperatures is not verified. And in terms of future large scale application of flexible CIGSe thin film devices it may also be beneficial to identify a Cd-free buffer layer technology suitable for low temperature grown absorber thin films.

5. Acknowledgements

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