PROGRESS TOWARDS THE DEVELOPMENT OF 18% EFFICIENCY FLEXIBLE CIGS SOLAR CELLS ON POLYMER FILM

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ABSTRACT: Development of lightweight and flexible solar cells using roll-to-roll manufacturing and monolithic interconnection technology can lead to a significant reduction in the manufacturing cost of solar modules. We have been conducting research and development work for improving the efficiency of flexible CIGS solar cells and implementing monolithically interconnected solar modules. A vacuum evaporation process for the growth of high quality CIGS absorber layers at sufficiently low temperature of about 450°C and a method of Na incorporation for improving the electronic properties have been developed. Flexible solar cells with a record efficiency of 17.6% and monolithically interconnected mini-modules with 12.1% efficiency have been realized. Analyses of photovoltaic properties suggest that efficiencies above 18% are achievable.

1 INTRODUCTION

High efficiency solar cells on are commonly processed by growing CIGS absorber layers on soda lime glass substrates at temperatures of around 600°C [1]. Processing of highly efficient solar cells on polymer films is challenging because most of the polymer films used as substrate lack thermal stability for the growth of high electronic and structural quality CIGS absorber layers. High thermal expansion coefficient of polymer causes a large stress in the layers deposited at high substrate temperature resulting in cracks and delamination of the solar cells from the substrate.

We have developed a vacuum evaporation process for the growth of high quality CIGS absorber layers at sufficiently low temperatures below 500°C and a method of Na incorporation for improving the electronic properties. We present a CIGS growth process at low temperature suitable for flexible solar cells on polymer substrates with an objective towards 18% efficiency.

A large improvement in the efficiency of flexible solar cells, from a previous record value of 14.1% [2] to a new record of 17.6%, was achieved by reducing the optical and electronic losses in the CIGS solar cell structure. The most important factor was the optimization of the composition gradient of Ga across the CIGS layer thickness and an appropriate incorporation of Na during the final stage of the growth process. Consequently, an optimum band gap grading and larger grain size in CIGS layer resulted in a substantial increase in the efficiency of flexible solar cells. Based on the results of 17.6% efficiency cells we propose the prospects for flexible CIGS solar cells on polymer films with efficiencies exceeding 18%.

2 EXPERIMENTAL

The device structure presented in this work is as follows. First, a 600 nm thick Mo bilayer was deposited by dc-sputtering onto commercially available polyimide film. The CIGS absorber layer was deposited by elemental co-evaporation technique in a high vacuum chamber (base pressure $\sim 10^{-8}\,\mathrm{mbar}$). Typical pressure

during processing was in the range of 10^{-7} - 10^{-6} mbar. A 40 nm thick CdS buffer layer was deposited by chemical bath deposition in two cycles. The i-ZnO/ZnO:Al(2%) window layers were grown by rf-sputtering with thicknesses of approximately 70 nm/210 nm, respectively, resulting in a sheet resistance of about $50~\Omega_{\square}$. Collection grids comprised of e-beam evaporated 50~nm/4000~nm/50~nm thick Ni/Al/Ni layers. The devices were antireflection coated with $100~\text{nm}~\text{MgF}_2$, deposited as well by e-beam evaporation. Individual solar cells were defined by means of mechanical scribing.

Figure 1 shows the evaporation profile of the process that led to the 17.6% record efficiency cells. The In/Ga ratio was held constant throughout the process. The maximum Cu content reached during processing was Cu/(In+Ga) ~ 1.15-1.20. About 20 nm of NaF were provided in a post-deposition treatment [3]. The substrate temperature shown was measured with a thermocouple behind the substrate in non-contact mode. Therefore, we assume that the real substrate temperature is significantly lower. The transition between Cu-rich to Cu-poor and vice versa was monitored by end-point detection [4].

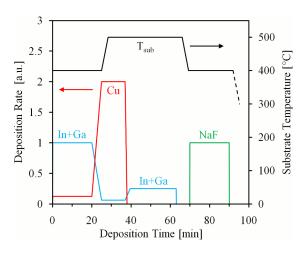


Figure 1: Schematic of the CIGS evaporation process and sodium incorporation. Selenium (not shown) is provided in constant overpressure until the deposition of the NaF compound.

3. RESULTS AND DISCUSSION

A cross-section of the finished device was investigated with scanning electron microscopy (SEM), and Figure 2 shows a representative image. The grain size is rather large for the low substrate temperature with some grains extending throughout the whole layer. The CIGS surface is relatively rough compared to previous investigations [5], which is attributed to a higher amount of Cu-excess reached at the end of the Cu evaporation. The apparent CIGS layer thickness is about 2.2 µm. Due to the preparation method used for SEM investigation the Mo back contact is not visible as a cross-section and only the surface is visible.

The bulk composition of the CIGS layer was determined by x-ray fluorescence measurement and yielded values in the range of x=Ga/(In+Ga)=0.34-0.36 and y=Cu/(In+Ga)=0.88-0.90.

The preferential grain orientation of the CIGS layer was investigated with x-ray diffraction in standard Bragg-Brentano configuration. Figure 3 shows the resulting spectra of the two commonly observed main peaks of CIGS, revealing a strong preferential (220)/(204) orientation of grains in the layer. The shape of the peaks suggests a broad Ga grading profile with a rather large portion of the CIGS layer being Ga-poor as indicated by a broad shoulder towards lower diffraction angles. However, depth profiling methods such as secondary ion mass spectroscopy are necessary to confirm this observation.

Two solar cells were measured independently at the Fraunhofer Institute for Solar Energy Systems (ISE) in Freiburg, Germany. Figures 4-7 show the corresponding J-V curves with photovoltaic parameters and the external quantum efficiency measurements.

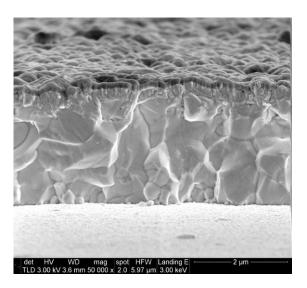


Figure 2: SEM cross-section of a 17.6% record efficiency solar cell grown on polyimide film.

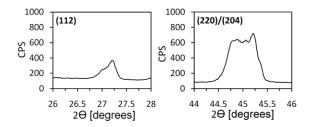


Figure 3: XRD scans of the record cell with the (112) peak on the left and the (220)/(204) doublet peak on the right. The scan duration was the same in both cases so that the intensities can be compared directly.

Compared to the previous 14.1% efficiency record device, improvements in all parameters resulted in the new record. Beside more optimized window and buffer layer deposition conditions, the significant improvement is attributed mainly to the CIGS deposition process. A reduction in growth rate in the 3rd stage is believed to enhance the grain size at the low substrate temperature applied here, especially towards the back contact.

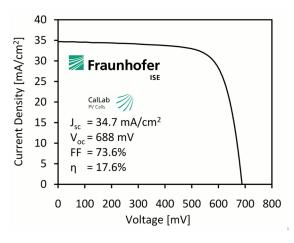


Figure 4: J-V curve and photovoltaic parameters of a 17.6% efficiency record cell on polyimide, which was independently certified by the Fraunhofer Institute for Solar Energy Systems (ISE) in Freiburg, Germany.

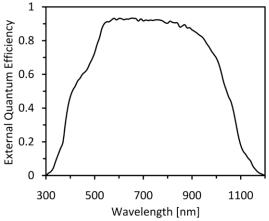


Figure 5: External quantum efficiency measurement of the corresponding 17.6% efficiency record cell.

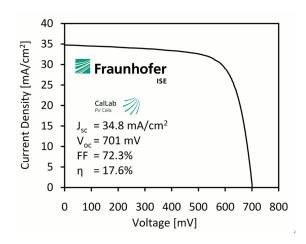


Figure 6: J-V characteristics of a second 17.6% efficiency solar cell grown on polyimide film.

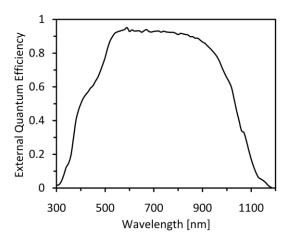


Figure 7: External quantum efficiency measurement of the corresponding 17.6% efficiency record cell.

Previous work revealed a significant decrease in grain size towards the surface as well as towards the back contact, and a much more pronounced Ga grading profile when the CIGS growth rate was increased drastically in the final stage of a low-temperature three stage process [6,7]. Further on, recent investigations revealed that optimization of the Ga grading profile is crucial for high efficiency solar cells on polyimide, and low temperature grown CIGS generally leads to a more pronounced Ga profile when three stage like processes are applied [5]. In the growth process applied in this work the main factors that appear to have smoothed the Ga grading profile beneficially are (i) post-deposition incorporation of Na, (ii) a higher amount of Cu-excess (15-20%), and (iii) a slow deposition rate of In and Ga at the end of the growth process.

4 CONCLUSIONS

A new record efficiency of 17.6% on flexible polyimide film has been presented, which is the highest reported value of any type of solar cell grown on polymer film. Main factors in achieving such high efficiencies lie

in systematic modifications of the CIGS growth process, which are required at the low substrate temperature applicable for polyimide films. Most importantly, a significant enhancement in grain size and optimization of the composition grading profile has been achieved.

Based on the best values of $J_{\rm sc}$, $V_{\rm oc}$ and FF of the two presented individual devices, it is feasible that efficiencies exceeding 18% can be reached on flexible substrates in the near future.

ACKNOWLEDGEMENTS

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REFERENCES

- [\1] I. Repins, M. A. Contreras, B. Egaas, C. DeHart, J. Scharf, C. L. Perkins, B. To, and R. Noufi, "19.9%-efficient ZnO/CdS/CuInGaSe₂ Solar Cell with 81.2% Fill Factor, Progress in Photovoltaics: Research and Applications, 2008, **16**, pp. 235–239.
- [2] D. Brémaud, D. Rudmann, G. Bilger, H. Zogg, and A. N. Tiwari, "Towards the development of flexible CIGS solar cells on polymer films with efficiency exceeding 15%", Conference Proceedings of the 31th IEEE Photovoltaic Specialists Conference, Orlando, 2005, pp. 223-226.
- [3] D. Rudmann, D. Brémaud, H. Zogg, and A. N. Tiwari, "Na incorporation for low-temperature Cu(In,Ga)Se₂ growth", 19th EU PVSEC, 2004, pp. 1710.
- [4] N. Kohara, T. Negami, M. Nishitani, and T. Wada, "Preparation of device-quality Cu(In,Ga)Se₂ thin films deposited by coevaporation with composition monitor", Japanese Journal of Applied Physics, 1995, **34**, pp. L1141–L1144.
- [5] A. Chirilă, D. Guettler, P. Bloesch, S. Nishiwaki, S. Seyrling, S. Buecheler, R. Verma, F. Pianezzi, A. Uhl, J. Perrenoud, Y. E. Romanyuk, R. Ziltener, D. Brémaud, G. Bilger, and A. N. Tiwari, "Optimization of Composition Grading in CIGS for Flexible Solar Cells and Modules", presented at the 35th IEEE Photovoltaic Specialists Conference, Honolulu, 2010.
- [6] A. Chirilă, D. Guettler, D. Brémaud, S. Buecheler, R. Verma, S. Seyrling, S. Nishiwaki, S. Hänni, G. Bilger, A.N. Tiwari, "CIGS solar cells grown by a three-stage process with different evaporation rates", Proceedings of the 34th IEEE Photovoltaic Specialist Conference, Philadelphia, 2009, pp. 812-816.
- [7] A. Chirilă, S. Seyrling, S. Buecheler, D. Guettler, S. Nishiwaki, Y. E. Romanyuk, G. Bilger, and A. N. Tiwari, "Influence of High Growth Rates on evaporated Cu(In,Ga)Se₂ layers and solar cells", submitted to Progress in Photovoltaics: Research and Applications.