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INORGANIC-ORGANIC HYBRID ENCAPSULATION OF FLEXIBLE $\text{Cu}(\text{In}, \text{Ga})\text{Se}_2$ MINI-MODULES

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ABSTRACT: An effective encapsulation solution is required to achieve market entry for flexible $\text{Cu}(\text{In}, \text{Ga})\text{Se}_2$ (CIGS) technology. In this work, we demonstrate the concept of inorganic-organic hybrid encapsulation by encapsulating $3 \times 10 \text{ cm}^2$ CIGS/polyimide mini-modules with 25 nm Al_2O_3 barrier layer grown by atomic layer deposition (ALD) in combination with back and front sheets. All mini-modules were encapsulated with a back sheet from Lenzing Plastics, while two different commercial front sheets from Feron and 3M were tested. When exposed to damp heat test, due to the presence of front encapsulation (ALD- Al_2O_3 and front sheet), the moisture ingress is slowed down during the first 150 h, but progressively penetrates into modules upon increasing exposure time, degrading mainly I_{sc} . The moisture ingress imposes stringent requirements in front encapsulation of flexible CIGS/polyimide mini-modules. Studies in employing thicker ALD- Al_2O_3 barrier layers and/or better H_2O -resistant front sheets will be carried out in the future.

Keywords: CIGS, Degradation, Encapsulation, Flexible Substrate, Stability

1 INTRODUCTION

$\text{Cu}(\text{In}, \text{Ga})\text{Se}_2$ (CIGS)-based photovoltaics (PV) technologies, being the thin film technology with one of the highest conversion efficiencies, are well positioned among the different flexible PV technologies already at industrial scale [1]. In order to achieve a cost-competitive market entry, effective encapsulation solutions are required to guarantee an operation time of 20-30 years. The European Solar-era.net project "Advanced Global Encapsulation Solutions for Long Term Stability in Industrial Flexible CIGS Photovoltaic Technology (DURACIS)" addresses this challenge, aiming to explore new and innovative low-cost encapsulation materials and concepts for flexible CIGS PV [2].

In order to maintain lightweight and flexibility, heavy and rigid encapsulation solutions such as glass - which are well adopted in the existing Si PV technology - are no longer suitable. Instead, organic encapsulants (such as polymeric front/back sheets) that are flexible and lightweight, are preferred. However, compared to inorganic materials, polymeric materials are generally more sensitive to moisture ingress [3]. As an example, EVA (ethylene-vinyl acetate) though being the most used encapsulant in PV field, is still sensitive to moisture and would decompose producing acetic acid upon exposure to moisture [4]. Therefore, inorganic thin films (such as Al_2O_3 , TiO_2 , SnO_2) grown by atomic layer deposition (ALD), which are widely used in OLED industry as efficient barrier layers thanks to their low water vapor transmission rate (WVTR) [5,6], have been proposed as promising barrier candidates for photovoltaics [7-9]. Among them, Al_2O_3 is one of the most studied barrier material [10,11]. However, despite a few studies discussing the application of ALD-grown Al_2O_3 as barrier layers in CIGS single cells with rigid glass as the substrate [7,12,13], reports are rarely seen in encapsulating flexible CIGS with ALD- Al_2O_3 as barrier layer, particularly on module level. Compared to rigid substrate counterpart, CIGS cells/modules made on flexible substrates (e.g. polyimide) are usually more fragile due to their sensitivity to mechanical stress (thus poor adhesion of the active

materials on the substrate), high susceptibility to moisture ingress (i.e. polymeric substrates are poorer water barrier than glass) etc.

We propose herein an inorganic-organic hybrid solution to encapsulate $3 \times 10 \text{ cm}^2$ mini-modules of flexible CIGS on polyimide substrates by combining ALD- Al_2O_3 barrier layer and front/back sheets. Our previous study on CIGS/glass mini-modules has shown that a thin ALD- Al_2O_3 layer $\geq 10 \text{ nm}$ is effective in preventing electrical degradation of mini-modules against damp heat test [14]. Damp heat test (DHT), *i.e.*, 1000 h at 85 °C and 85% R.H. (relative humidity), is one of the most severe tests for thin film modules according to the international standards IEC 61215-2 [15]. Here in this work, we have applied a 25 nm ALD- Al_2O_3 barrier layer on flexible CIGS/polyimide mini-modules, then further encapsulated the modules with a back sheet supplied by one of the industrial partners in the consortium Lenzing Plastics and with commercial front sheets purchased from Feron and 3M. The completely encapsulated flexible CIGS/polyimide mini-modules were then placed in a climate chamber to examine their damp heat stability.

2 EXPERIMENTAL DETAILS

2.1 Preparation of encapsulated CIGS/polyimide mini-modules

For the fabrication of flexible CIGS mini-modules, a 25 μm thick polyimide foil was used as the substrate. First, a 500 nm thick molybdenum back contact was DC sputtered inline, followed by laser scribing of the Mo back contact (P1), which defines the cell size. The CIGS absorber layer with a thickness of about 2.5 μm was then co-evaporated in a low-temperature 3-stage batch process. A CdS buffer layer (about 60 nm thick) was then deposited by chemical bath deposition (CBD). After the RF sputtering of a 80 nm thick undoped ZnO layer, the P2 scribe was performed via laser scribing. The subsequent Al:ZnO front contact was DC sputtered with a thickness of about 1000 nm. The final P3 patterning step was performed by a ps-laser. Module edge definition was done

by cutting the edges. Hence, 15 cells were interconnected to one module on a 3×10 cm² polyimide substrate with an active area of 24 cm². A total of eight mini-modules were prepared with an efficiency level of 8-11%.

Thin barrier layers of 25 nm Al₂O₃ were deposited by atomic layer deposition (ALD) in a BENEQ TFS-200 reactor at a deposition temperature of $T = 160$ °C, with trimethyl aluminum (Al(CH₃)₃, TMA, Optograde, Rohm & Haas) and deionized water as precursors [16]. The homogeneity of Al₂O₃ growth over 15.6×15.6 cm² (ALD reactor size) has been controlled. All the CIGS/polyimide modules with 25 nm ALD-Al₂O₃ barrier layer were further encapsulated with a back sheet supplied by Lenzing Plastics. Three modules were encapsulated with commercial front sheet (transluX Barrier²) from Feron and other three were encapsulated with commercial front sheet (3MTM Ultra Barrier Solar Film) from 3M. A polyolefin material (3MTM Solar Encapsulant Film PO8110) from 3M was used to laminate the front and back sheets to the CIGS/polyimide modules in combination with an edge sealant (HelioSealTM PVS 101). The lamination was carried out at 150 °C with 5 min for evacuation and 10 min for press. To avoid bending of the back sheet, the modules were cooled down to room temperature for 30 minutes before they were removed from the protecting foil of the laminator. The two modules without front encapsulation were annealed at 160 °C in the ALD reactor for 1 h, and encapsulated only with the back sheet from Lenzing Plastics at 170 °C using an encapsulant material (Solartex PV) from Lenzing Plastics (the same encapsulant material was also used on the front side to protect the modules during lamination process). For contacting, a tin coated Cu tape was fixed by a silver containing glue onto the contact bars of the modules.

2.2 Damp heat test and characterization of degraded mini-modules

Damp heat test (DHT) was performed in a SunEvent SUN/340/S climate chamber from Weiss Umwelttechnik. Modules were first heated to 85°C under dry conditions for 30 minutes to avoid condensation. Then the relative humidity (R.H.) was also increased to 85% R.H. with the samples remaining in the climate chamber. After a certain time, the modules were removed from the climate chamber to perform I-V measurements. Then the same procedures were repeated until the total test time of 900 h was reached.

The current–voltage (I–V) characteristics of CIGS/polyimide mini-modules were measured at 25 °C under 1000 W/m² simulated AM1.5G illumination (WACOM WXS-90S-5, Voss Electronic GmbH, Kirchheim-Heimstetten, Germany). Before each measurement the samples were light soaked for 10 min at 25 °C under open circuit condition.

3 RESULTS AND DISCUSSIONS

A total number of 8 mini-modules (3×10 cm²) of CIGS on polyimide were fabricated and all modules were encapsulated with back sheet supplied from Lenzing Plastics. On the front side, 3 different encapsulation schemes as detailed in Table I were employed. The growth of 25 nm ALD-Al₂O₃ barrier layer was carried out at 160 °C for about 1 h. To make the performance evolution of the mini-modules comparable, the 2 modules without front encapsulation were also annealed in the ALD reactor at 160 °C for 1 h. Hence, all the modules underwent the same

thermal treatment before the damp heat test (DHT). The initial efficiency before DHT of these modules is about 8-11%. The relative change of efficiency η as a function of damp heat time is summarized in Fig. 1. For the modules without front encapsulation (*i.e.* without ALD-Al₂O₃, without front sheet), the efficiency dropped to ~20% of the initial value at as early as 150 h DHT. After 900 h DHT, the modules were fully degraded with complete loss of efficiency. For those encapsulated with 25 nm ALD-Al₂O₃ and front sheet from Feron, ~80% of initial efficiency was preserved at 150 h DHT; however, with further exposure to 900 h, the modules also completely lost efficiency. By comparison, the modules with front sheet from 3M remain ~87% of their initial efficiency at 150 h DHT, and survived after 900 h DHT although the final efficiency is still unsatisfactorily low: ~33% of initial value.

Table I: Three different front encapsulation schemes for 3×10 cm² CIGS/polyimide mini-modules

Front encapsulation scheme	# modules
Without ALD-Al ₂ O ₃ , without front sheet	2
25 nm ALD-Al ₂ O ₃ , front sheet from Feron	3
25 nm ALD-Al ₂ O ₃ , front sheet from 3M	3

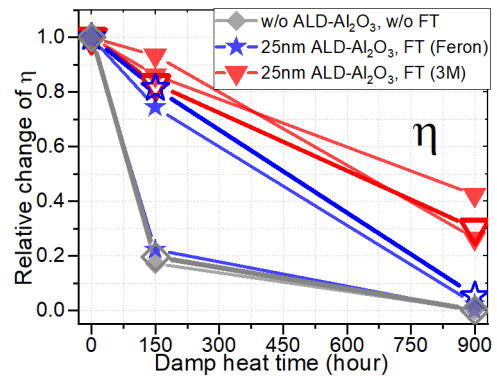


Figure 1: Relative change of efficiency η as a function of damp heat time for 3×10 cm² CIGS/polyimide mini-modules with different front encapsulations as indicated. FT stands for front sheet. All modules were encapsulated with back sheets supplied from Lenzing Plastics. The I-V curves of the modules plotted with open symbols are shown in Fig. 2.

Indeed, for the modules without front encapsulation, aggressive moisture penetration from the front side can be expected. As a consequence, the modules underwent severe degradation already at early stage, evidenced by the poor I-V response at 150 h DHT (Fig. 2a); extended exposure to 900 h entirely destroyed the diode behavior of the modules. On the contrary, for the modules with front encapsulation (Fig. 2b-c), due to the presence of moisture-blocking ALD-Al₂O₃ barrier layer and front sheet (Feron or 3M), the moisture penetration is slowed down at 150 h DHT, thus the resulting I-V curves are fairly similar to the ones before DHT. However, with further exposure, moisture still progressively penetrates into modules thus very poor I-V curves at 900 h DHT are seen (Fig 2b-c).

Fig. 3 summarizes the detailed changes of electrical parameters V_{oc} (open circuit voltage), I_{sc} (short circuit current) and FF (fill factor) of the CIGS/polyimide mini-modules as a function of damp heat time. A close look reveals that the efficiency loss observed after these mini-

modules were exposed to damp heat was predominantly due to loss in I_{sc} and FF. This is most likely due to increase in series resistance as a consequence of electrical degradation of the front Al:ZnO (AZO) window layer [17] and/or the back Mo layer [18] upon moisture ingress. Moreover, in modules, the conductivity loss in AZO layer would contribute significantly to the contact resistance in P2 interconnection [19], which further increases the series resistance. As such, at 150 h DHT, I_{sc} in modules without front encapsulation drops most significantly while I_{sc} in modules with front encapsulation (with front sheets from both Feron and 3M) remain more or less reasonable (Fig. 3b). With further exposure up to 900 h, moisture nevertheless penetrates through front encapsulation and so I_{sc} in modules with front encapsulation drops significantly.

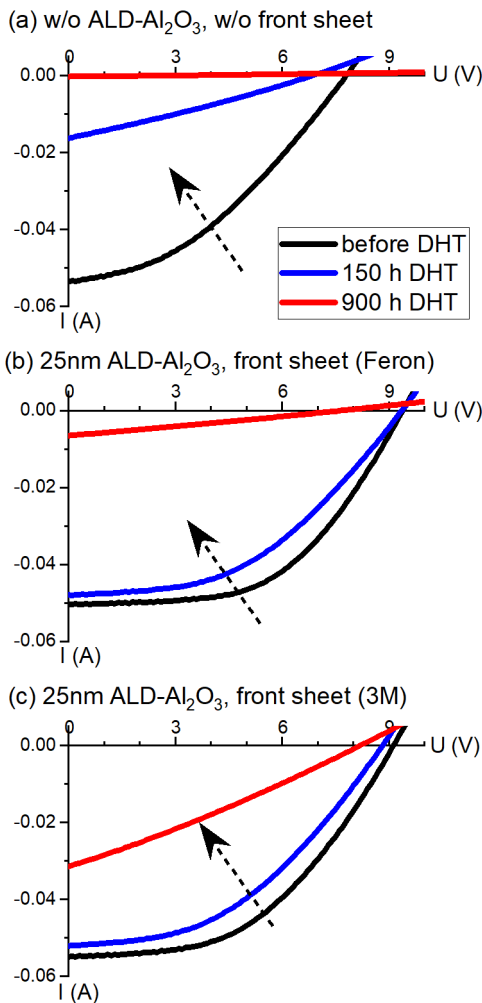


Figure 2: I-V characteristics before DHT (black curve) and after 150 h (blue curve), 900 h (red curve) damp heat test (DHT) time for $3 \times 10 \text{ cm}^2$ CIGS/polyimide (a) without ALD- Al_2O_3 and without front sheet, (b) with 25 nm ALD- Al_2O_3 and commercial front sheet from Feron, (c) 25 nm ALD- Al_2O_3 and commercial front sheet from 3M. All modules were encapsulated with back sheets supplied from Lenzing Plastics. The dashed arrow indicates the direction of increasing DHT hour.

For the modules without front encapsulation, further moisture ingress reduces not only I_{sc} but also V_{oc} , which could be that moisture penetrates deep into the cell degrading the CIGS and/or CIGS/CdS junction [20]. The V_{oc} loss is much less significant in modules with front

encapsulation, suggesting that due to the presence of front encapsulation and thus slower humidity penetration, the inner CIGS and CdS layer are less impacted. One of the modules encapsulated with Feron front sheet degrades even worse than modules without front encapsulation. This could be that the lamination was problematic (for example with presence of holes or defects), and thus this module is excluded for consideration.

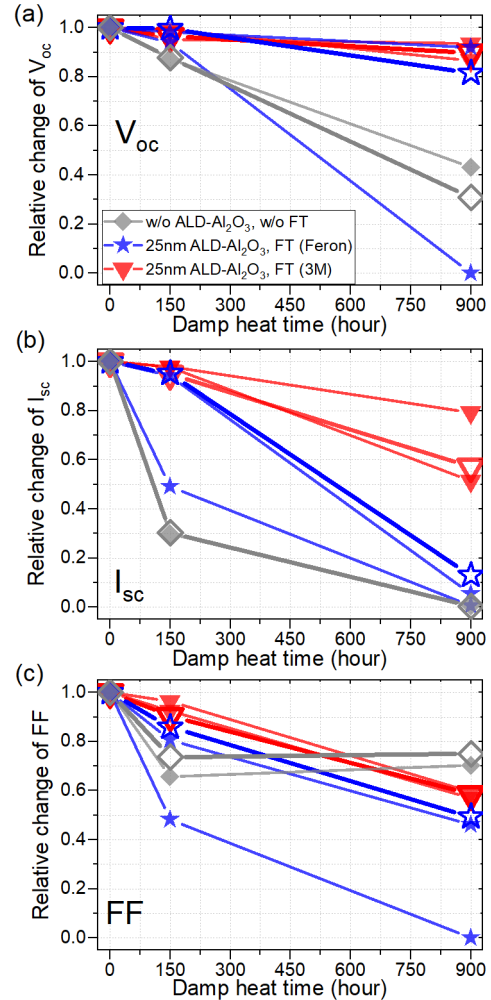


Figure 3: Relative changes of (a) open circuit voltage V_{oc} , (b) short circuit current I_{sc} and (c) fill factor (FF) as a function of damp heat time for $3 \times 10 \text{ cm}^2$ CIGS/polyimide mini-modules with different front encapsulations as indicated. FT stands for front sheet. All modules were encapsulated with back sheets supplied from Lenzing Plastics. The I-V curves of modules plotted with open symbols are shown in Fig. 2.

To sum up, when exposing the $3 \times 10 \text{ cm}^2$ CIGS/polyimide mini-modules with inorganic-organic hybrid encapsulation to damp heat test:

- 1) In the first 150 h, severe moisture penetrates into modules without front encapsulation, resulting in a significant drop in I_{sc} ; for the modules with front encapsulation (both front sheets from Feron and 3M), moisture ingress is slowed down thanks to the presence of front encapsulation thus negligible loss in I_{sc} is seen.
- 2) After 150 h, moisture penetrates progressively even in modules with front encapsulation, resulting in almost complete loss of I_{sc} in

modules with Feron front sheet while less than 50% I_{sc} loss occurred in modules with 3M front sheet. It thus appears that the front sheet from 3M behaves slightly superior than that from Feron in blocking moisture ingress although the efficiency of modules with 3M front sheet is still not satisfactory. On the other hand, the presence of front encapsulation still helps to prevent the humidity to penetrate deep into the CIGS cell. Thus, the V_{oc} did not show significant drop in modules with front encapsulation while this is not the case in modules without front encapsulation.

An effective front encapsulation to block moisture penetration is prerequisite for the damp heat stability of flexible CIGS/polyimide mini-modules. In the inorganic-organic hybrid encapsulation scheme introduced in this work, the two front sheets from Feron and 3M do not appear effective in encapsulating CIGS/polyimide mini-modules with respect to the criteria set by the international standard 61215-2 (*i.e.* 5% efficiency drop after 1000 h DHT). However, it could be that the ALD- Al_2O_3 employed is still too thin since CIGS cells made on polyimide substrates are more fragile than those on reference glass substrate. Thus in the future it could be interesting to test thicker ALD- Al_2O_3 barrier layers and as well other front sheets which are better humidity-resistant.

4 CONCLUSION

We demonstrate in this work the concept of an inorganic-organic hybrid encapsulation scheme and have encapsulated 3×10 cm² CIGS/polyimide mini-modules with a 25 nm ALD- Al_2O_3 barrier layer in combination with back sheet (supplied from Lenzing Plastics) and with front sheets (commercial ones purchased from Feron and 3M). When exposed to damp heat test, moisture enters the modules and degrades mainly the I_{sc} by most likely increasing the series resistance. Due to the presence of front encapsulation (ALD- Al_2O_3 and front sheet), the moisture ingress is slowed down in the first 150 h, but further damp heat exposure still leads to progressive moisture ingress resulting in significant I_{sc} loss up to 900h.

It is clear that the moisture ingress imposes stringent requirement in front encapsulation of flexible CIGS/polyimide mini-modules, and more challenges need to be tackled in encapsulating flexible CIGS modules compared to rigid counterparts. In the future, thicker ALD- Al_2O_3 and/or better H_2O -resistance front sheets will be tested and studied.

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