Damp-heat-induced degradation of layers in CIGS solar cells

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ABSTRACT

Investors require a guarantee of a minimum lifetime for PV installations. It is tempting to provide such a guarantee for a longer lifetime simply by specifying test conditions that are more and more severe. In this paper it is argued that, with a more detailed understanding of the basic mechanisms determining cell material behaviour under specific exposure conditions, not only can the inherent lifetime of solar cells and modules be improved, but also the predictive value and effectiveness of lifetime testing. An overview of the literature contributions regarding the influence of damp-heat exposure of the layers in Cu(In,Ga)Se₂ (CIGS) solar cells is presented. The material changes, as well as their potential influence on solar cell and module performance, are described. For the molybdenum back contact, it was observed that damp-heat exposure leads to a decrease in conductivity and reflectivity, most likely caused by molybdenum oxidation. The presence of a selenized-molybdenum (MoSe₂) top layer, in combination with the use of low sputtering pressure, resulted in more stable molybdenum films. For the transparent conductive front contact, a comparison of reports in the literature revealed that indium tin oxide (ITO) films are more stable in damp heat than ZnO:Al films. It was also observed that ZnO:Al films degraded as a result of the ingress of water and CO_2 via the grain boundaries, thereby lowering the conductivity of the material. The results of damp-heat studies of CIGS absorbers and buffers were difficult to quantify and varied between cases. In many instances, absorber exposure gave rise to the formation of spots on the surface, as well as to sodium migration, whereas buffer exposure often led to interdiffusion and reactions with the absorber and front contact. Literature reports also demonstrated that the Mo/ZnO stack in the P2 scribe is a vulnerable location in CIGS modules.

Reliability of thin-film PV

Thin-film PV modules based on Cu(In,Ga)Se₂ (CIGS) technology have shown themselves to be very promising. CIGS technology allows lightweight, low-cost PV products, and leads to the highest efficiencies among the various types of thin-film PV; the highest-performing CIGS solar cells today have yielded conversion efficiencies of up to 22.3% [1], thus even outperforming multicrystalline silicon cells.

For the large-scale market introduction of CIGS modules, solid performance stability, along with low initial costs and high efficiency, is an important prerequisite (Fig. 1). Besides the lower electricity costs afforded by reliable, longlifetime modules, the predictability of performance is also important: financiers, home-owners, utilities, planners and especially producers need to be able to predict when their modules will no longer function, so that the risks can be evaluated. Performance reliability is therefore very important in order to obtain the lowest and most predictable electricity costs.

"For the large-scale market introduction of CIGS modules, solid performance stability is an important prerequisite." In cases where the field performance of CIGS modules has been studied, large differences were observed: many modules were very stable (e.g. no degradation after seven years), but field failures were also seen [2,3]. Because of the relatively limited field experience of CIGS modules, combined with the multitude of techniques used for module production, the lifetime prediction for CIGS modules in general is a challenge. More information about the degradation behaviour of CIGS modules is therefore required in order to decrease degradation rates and make those rates more predictable, and thus to improve long-term performance. Studies have yielded the following observations regarding less stable modules in the field:

• Field exposure mainly affects the fill factor and the voltage, while the current is mostly relatively stable.



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• Degradation in the field is often related to the ingress of moisture into the CIGS modules, thereby affecting the CIGS cell material (Fig. 2) inside the module.

Since moisture ingress, often combined with elevated temperatures, has a negative impact on CIGS solar cells, modules are provided with water barriers. For rigid modules, glass is an excellent barrier choice; however, for flexible modules, expensive organic– inorganic multilayer coatings are often required. Therefore CIGS cells that are intrinsically more stable can help to minimize the barrier costs and facilitate the large-scale market introduction of flexible CIGS modules. Fig. 3 shows the various ways in which intrinsically more stable and predictable solar cells can help lower electricity costs with PV modules.

Knowledge of the stability of CIGS solar cells and modules is often obtained by the simulation of degradation phenomena in accelerated lifetime tests (ALT). In order to study the effect of moisture ingress at elevated temperatures, the IEC procedure 61646 for thin-film PV includes 1,000 hours' exposure to 'damp heat' (85°C/85% relative humidity (RH)).

In learning about the stability of CIGS solar cells and modules, many aspects should be considered, including:

- The damp-heat stability of the individual layers in the CIGS solar cells and modules.
- The interaction between these layers and between the stack and the module package material.
- The influence of biases, such as illumination [3,4].

Although none of these aspects in isolation can be used to predict the failure mechanisms occurring in CIGS modules in the field, they can give an indication of the vulnerable materials and interfaces, and allow improvements to the various parts of the module. In this paper the results of damp-heat exposure of individual layers, and of the interconnection areas within CIGS solar cells and modules, are summarized.

A short overview of the data previously published in Theelen [3] (and in the paper by Theelen & Daume [4], submitted for publication) will be given. In these cited works, more extensive literature references can also be found (in particular for the data used for Figs. 5 and 7).



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Figure 2. Cross section of the make-up of a typical CIGS solar cell.

Back contact: Molybdenum (0.3-1 µm)

Figure 3. Routes by means of which more intrinsically stable and predictable solar cells can reduce the cost of electricity generated by PV modules. 'Slower degradation rate' and 'longer module lifetime' are naturally related: the former refers to the real non-linear loss of output power in the field ('stability'), while 'lifetime' is the economic lifetime (e.g. maximum 20% efficiency loss) as defined before installation and on which the electricity cost calculations, installation planning and risks assessments are based ('predictability').

Molybdenum back contact

Sputtered molybdenum (Mo) is used as the back-contact material for CIGS solar cells and modules by nearly every research group and manufacturer. The main requirements for a functional molybdenum back contact are good conductivity and reflectivity, the latter especially when thin absorber layers are used. Molybdenum films within CIGS cells and modules are normally covered by a thin layer of MoSe₂, while the molybdenum is uncovered between the molybdenum and CIGS deposition steps.

Stability of the Mo back contact

The degradation of molybdenum can occur at two points in time [5]:

- 1. Degradation of bare metallic molybdenum in storage. Molybdenum which is not immediately used for further processing can be stored, but exposure of bare molybdenum to the atmosphere can affect the electrical properties of the later-processed CIGS solar cells and modules.
- 2. Long-term stability of the molybdenum back contact in CIGS







cells and modules. These layers are covered, in most places, by a thin $MoSe_2$ layer (Mo/MoSe₂ stack), as well as by the rest of the cell stack. These molybdenum layers are therefore mostly vulnerable in the P2 and P3 scribes (see scribe section later). An assessment of the stability of this material should be based on the actual composition of the molybdenum in these particular areas.

In many cases, exposure to elevated temperatures and humidity and oxygen levels leads to oxidation of the films: this is often visible as black and blue stains on both molybdenum and selenized-molybdenum (Mo/MoSe₂) films, as shown in Fig. 4. These stains contain molybdenum oxide (MoO_2/MoO_3), which is generally poor in terms of conductance and reflectance. The molybdenum oxide layers sometimes even show cracks and needles on the surface.

The formation of a thick layer of non-conductive molybdenum oxide can lead to a very rapid decrease in conductivity when measured from the top. Prior to this plummet, most films exhibit a slow decline in conductivity: an overview of the decrease in conductivity for various types of molybdenum films is shown in Fig. 5.

Differences in degradation rate are caused by the presence of the MoSe₂ top layer: selenized molybdenum degrades more slowly than bare molybdenum films, most likely because it is easier to oxidize metallic molybdenum than MoSe₂. Another factor that influences the degradation rate is the sputter pressure. Degradation effects are the most severe for molybdenum deposited at high pressures during sputter deposition, thereby forming more porous molybdenum layers, which are more susceptible to the ingress of, among other things, water and oxygen [5].

Various references have reported on the presence of sodium, which probably plays a role in molybdenum degradation. Sodium can occur, for example, in the form of needles on the molybdenum surface, but it can also intercalate via a reduction reaction into MoO₃, thereby forming Na_xMoO₃ [6].

Influence on module stability

The presence of molybdenum oxide in the scribes is discussed in the scribe section, and so only the effect of oxidation of the Mo/CIGS interface will be described here. Oxidation can, on the one hand, simply increase the series resistance and decrease the fill factor of a solar cell or module; on the

81

other hand, it can change the ohmic behaviour at the Mo/CIGS region, thereby influencing various electrical parameters. It should be noted that oxidation is not always detrimental to cell or module performance; mild oxidation could actually even result in slightly improved efficiency [7].

The CIGS absorber and the buffer layer

The CIGS absorber and the buffer layer form the p and n parts of CIGS solar cells and modules. A study of the damp-heat stability of these individual layers is complicated, since their interaction with each other and with the front and back contacts greatly influences the stability of the layers. Consequently, these layers are mainly tested as part of the complete solar cell stack, while measurements for the individual layers are scarce, and the interpretation of the effect of the observations on the solar cell or module output is especially complicated.

Stability of the CIGS absorber

The CIGS absorber has a variety of point defects with complex interactions that determine their benign electronic behaviour. The CIGS absorber is an impurity-tolerant material with radiation hardness, and it can be stated that CIGS grains themselves are thus intrinsically stable, irrespective of their complicated structure [8]. Changes in the CIGS absorber due to extrinsic influences, such as damp heat, are therefore often caused by changes around the grain boundaries within the polycrystalline absorber film and the interfaces with the back contact and buffer.

It has been observed that the exposure of bare CIGS absorbers to even ambient conditions has led to a rapid reduction in carrier lifetime [9]. This degradation process can be prevented by buffer deposition on top of the CIGS absorber. Another source [10] has reported that when bare CIGS absorbers were exposed to damp heat, spots were formed on the surface. These spots exhibited low concentrations of gallium, copper and selenium, but high sodium content, caused by, for example, hydrolysis or oxidation. Furthermore, the formation of sulphate has been reported as a result of damp-heat-induced oxidation of sulphur in Cu(In,Ga)(Se,S)₂ solar cells [11].

Sodium has been found to have a very large impact on CIGS absorber stability: oxidation of CIGS absorbers occurs in the presence of sodium and water. Experiments have shown that this oxidation, catalysed by water, promoted an enhanced removal of selenium from the absorber layer via the formation of Na_2Se_x compounds, leading to severe efficiency loss, mainly because of reduced shunt resistance [12]. Additionally, the presence of a large sodium content due to a porous molybdenum back contact even leads to physical distortions in the CIGS absorber layer [13].

Damp-heat stability is also influenced by the gallium content. Experiments with complete solar cells with CIGS absorbers containing different levels of gallium have shown that for a medium concentration of gallium, the degradation rate was the lowest, whereas cell degradation was more intense in the case of pure CIS or CGS absorbers [14].

Stability of the buffer

As a buffer in CIGS solar cells, CdS has often been the material of choice. However, because of the toxicity of cadmium and the use of a wet-chemical deposition technique, other materials are being used more and more [15]. The buffer has always been studied in terms of its contact with the other layers.

The stability of the buffers CdS, $Zn(O,S,OH)_x$ and In_2S_3 under dampheat conditions has been widely

reported in the literature. It has been found that the CdS buffer can diffuse into the CIGS and ZnO layers under damp-heat conditions. Possible reaction products are ZnSO₄ or some other sulphate [11], and Cd_{1-x}Zn_xS and $ZnO_{1-x}S_x$ [16]. The interaction between CdS and the doped ZnO front contact can possibly lead to an increase in the sheet resistance of this contact. An interaction between the buffer and the front contact has also been found when $Zn(O,S,OH)_x$ is used as a buffer, which quite likely resulted in the hydration and dehydration of ZnO into $Zn(OH)_2$, and vice versa [17]. When In_2S_3 is used as an alternative buffer, no major differences in stability behaviour from that of CdS-based solar cells have been found [18].

TCO front contact

The transparent conductive oxide (TCO) functions as a front contact of CIGS solar cells or modules. It should be transparent in order to allow the influx of photons into the CIGS layer, while conductivity is required of it for the transport of the produced electrons. The main requirements for a TCO in a solar cell or module are therefore conductivity and transparency. Several types of TCO are possible candidates for the front contacts of CIGS solar cells



Figure 6. The presence of grain boundaries in doped zinc oxide can lead to grain boundary scattering, thereby reducing the electron carrier transport (on the basis of Theelen et al. [19,21]). The presence of unwanted species due to damp-heat exposure can increase this effect, leading to a decrease in conductivity of the film.

82

and modules; of these, the costeffective sputtered ZnO:Al is the most frequently used, but sputtered indium tin oxide (ITO) is also implemented.

"An increased resistivity of ZnO:Al films is often found to be the main cause of efficiency losses in solar cells and modules."

Stability of the TCO front contact

An increased resistivity of ZnO:Al films is often found to be the main cause of efficiency losses in solar cells and modules. This increased resistivity is primarily driven by a decrease in mobility and is typically caused by the diffusion of species from the atmosphere into the grain boundaries. The migration of, among others, water and CO₂ can lead to the formation of molecules such as Zn(OH)₂ and $Zn_5(CO_3)_2(OH)_6$ [19,20], while the adsorption of atmospheric species in the grain boundaries might also occur. The presence of these species can lead to the formation of a potential barrier at the grain boundaries, resulting in grain boundary scattering (Fig. 6) and thus increased resistivity. Additionally, spot formation has been observed [19], but large changes in the transparency of the ZnO:Al due to damp-heat exposure have not been reported.

More stable ZnO:Al films have been obtained by thicker layers [22], higher deposition temperatures [19] and higher doping concentrations [22]. Furthermore, it has been found that ZnO:Al on rough substrates shows a faster increase in resistivity than on smooth substrates, as a result of damp-heat exposure. Rough underlying absorber and buffer lavers can therefore also negatively impact the damp-heat stability of the ZnO:Al film [23]. Increases in resistivity can be largely reversed by annealing in a vacuum or in a reducing atmosphere at elevated temperatures [22,24], but the feasibility of high-temperature annealing of ZnO:Al on the top of complete CIGS solar cells or modules needs to be further investigated.

The more expensive ITO is generally more stable than ZnO:Al in the presence of humidity and elevated temperatures (Fig. 7). The conditions that favour the deposition of dampheat-resistant ITO films are higher deposition temperatures [25] and lower partial pressures of oxygen [26]. The damp-heat exposure of ITO also often results in the formation of spots [25].



Figure 7. Box plots showing increase in resistivity per hour (assumed to be linear) for ZnO:Al, ITO and i-ZnO/ZnO:Al films exposed to damp-heat conditions. The resistivity changes for ITO in particular, and in one case for ZnO:Al, can be zero or negative (not shown in the logarithmic graph).

Influence on module stability

There are various ways to determine the impacts of the TCO on the stability of a complete CIGS solar cell or module. The first of these impacts is the one that is mainly addressed in this paper:

- 1. The impacts of increasing resistivity and decreasing transmission. The effect of an increase in resistivity, as often observed, can mainly be seen in a series resistance increase and thus in a fill factor decrease. A decrease in transmission, on the other hand, reduces the current of the solar cell or module. On the basis of damp-heat experiments, the effect of a transmission change is not expected to be large.
- 2. Changes in TCO properties due to chemical reactions between TCO and, for example, the encapsulation material, such as ethylene vinyl acetate (EVA). Similarly to point 1, these effects can be found by changing resistivity and transmission values.
- 3. Water barrier properties of the TCO itself (e.g. i-ZnO and ITO are reported to function as water barriers [27]).
- 4. The changes in carrier concentration in both the TCO and the CIGS layer can lead to changes in the Fermi level, which will induce a change in voltage.
- 5. The possibility of atmospheric species diffusing through the TCO,

thereby allowing the underlying CIGS/buffer layers to react with these atmospheric species, leading to a change in the absorber or buffer properties, which can impact, for example, the voltage.

The use of the more expensive ITO instead of the cost-effective ZnO:Al can be considered. Because of the higher stability of the ITO films, the barrier costs in flexible modules can thus be reduced, which can lead to a module with a lower total cost price.

Scribe degradation

The monolithic interconnection scheme of a CIGS module plays an important role in module degradation. Fig. 8 shows a scribe area, where the degradation risks are indicated [28]:

- An increase in absorber conductivity around the P1 scribe, which can decrease the shunt resistance. A similar effect has been reported by Allsop et al. [18], but it was not observed very often.
- A series resistance increase in P2, where a Mo/ZnO:Al contact is responsible for the current transport between the solar cells. Possible reasons are the introduction of an oxide layer at the Mo/ZnO:Al interface, deteriorating the contact, or the increased resistivity of ZnO:Al films. This effect has been observed in model systems, such as in Klaer et al. [29].
- Degradation of P3, due to oxidation



of molybdenum, for example in locations that have been damaged by the scribing process. This could lead to an increase in series resistance. However, the metal molybdenum has a high conductivity, which means its degradation will only impact module efficiency when the molybdenum layer is almost completely oxidized [6]; this will most likely happen only after harsh exposure.

Furthermore, the increased resistivity of the TCO also has an additional impact on modules in the case where the molybdenum resistivity remains constant: this leads to the creation of a difference in voltage drops at different locations, which can result in local heating in solar cells [30].

Conclusions and outlook

On the basis of these observations, some practical changes that might lead to more stable solar cells and modules could be implemented. It should be noted that these recommendations are based on the experience of the author with specific CIGS cell fabrication approaches, in combination with literature data; small variations in the composition might lead to other dependencies. The suggested changes are:

- Use molybdenum with a low deposition pressure, especially near the Mo/CIGS interface.
- If possible, keep the selenizedmolybdenum film intact in P2 and P3.
- Use ITO films instead of ZnO:Al films.

- Use thicker layers, higher deposition temperatures or doping concentrations, or post-deposition treatments for the ZnO:Al deposition.
- Implement minor changes in, for example, the absorber, which can have a major influence on cell stability.
- Recognize that sodium is often involved in degradation processes, while the role of potassium is still unknown.

"Future lifetime studies need to focus more on understanding the degradation mechanisms, instead of simple go/no-go judgments being made on the basis of IEC tests."

Future studies

Since moisture ingress is often the reason for degradation, many CIGSspecific degradation problems can be prevented by an adequate water barrier; however, since intrinsically stable CIGS solar cells are preferred, more knowledge is undoubtedly required in this area. In order to expand the current level of knowledge, future lifetime studies need to focus more on understanding the degradation mechanisms, instead of simple go/no-go judgments being made on the basis of IEC tests.

A better understanding of the degradation can even help to lower the required test loads. Currently, investors might ask for damp-heat tests lasting more than 1,000 hours (IEC test) in order to be certain about the stability of CIGS modules. However, the calculations by Coyle [31] indicate that the current 1,000 hours' dampheat exposure can simulate between 1.1 and 110 actual years in Miami, depending on the composition of the CIGS solar cells, but also on the nature of the package material. Simply increasing the damp-heat exposure time is therefore only relevant in cases where degradation phenomena with a relatively low acceleration factor are the dominant mechanisms.

To conclude, it is very important to compare accelerated lifetime effects with field test results; this comparison allows the appropriate accelerated tests to be selected. It is therefore of interest (particularly to this author) to always examine modules that have failed in the field and make comparisons with laboratory test results.

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