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Temperature-Induced Degradation of Thin-Film III-V Solar Cells for Space Applications

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Abstract—**High efficiency, thin-film III-V solar cells offer excellent characteristics for implementation in flexible solar panels for space applications. In order to investigate the space compatibility of such cells. The temperature-induced degradation of both substrate-based cells with Au and Au/Cu contacts and thin-film cells on Au and Cu carriers was studied by accelerated ageing testing (AAT) at 200°C. With less than 3% decrease in efficiency after 37 days at 200°C (equivalent to 10 years at 100°C for E_a = 0.70 eV) the substrate-based cells show excellent results. With a 10% decrease in efficiency after 37 days of AAT the thin-film cells on an Au carrier exhibit promising results, given the early stage of development of the thin-film cells. On the other hand severe degradation is observed for thin-film cells on a Cu carrier (decrease in efficiency > 60% after 37 days of AAT). At least two factors contribute to this severe degradation: thermally-induced stress and Cu diffusion.**

I. INTRODUCTION

NOVEL III-V solar cell preparation approaches such as inverted metamorphic (IMM) growth [1]–[3], wafer-bonding [4] and mechanical stacking [5] require at least one growth substrate removal to create an operational device. Techniques such as epitaxial lift-off (ELO) [6]–[9] and controlled spalling [10], [11] allow for substrate removal without destruction of the expensive growth substrate, thus allowing for substrate re-use [12], [13]. The released active cell structures have the intrinsic potential to be turned into genuine thin-film devices if they are transferred to a thin and flexible carrier. Such thin-film III-V devices offer excellent characteristics for implementation in next generation space solar panels, as they allow for a significant weight reduction on panel level [14], while at the same time offering the highest possible solar cell efficiencies [3], [15], [16]. Space, however, also provides a harsh environment (vacuum, harsh UV, electron and proton radiation, temperature cycling) which adds additional design challenges. Unfortunately reports of space environmental testing of thin-film III-V solar cells are scarce [17]–[20] and a number of design challenges remain to be addressed. These include the need for thin-film interconnection techniques, suitable radiation and UV resistant flexible cover

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glasses [14], [21] and in particular a space compatible flexible carrier and support.

Both the ELO and controlled spalling techniques already implement metal foil carriers as flexible handling and thin-film support during substrate removal [22]–[26]. The ELO process currently used at Radboud University utilizes a Cu carrier for handling and support of the thin-film semiconductor structures, as Cu is relatively cheap, compatible with all (post-)ELO solar cell processing steps and can be easily applied by a number of chemical and physical deposition processes. However, Cu is also known to diffuse rapidly into many semiconductors [27] and to introduce mid band gap trap levels [28]. The harsh environment provided by space (particularly the elevated maximum temperature during temperature cycling) may induce Cu diffusion, having a potentially detrimental effect on the device performance. The main objective of this study is to investigate the temperature-induced degradation of thin-film solar cells on Cu carrier foils.

Unfortunately, standardized test procedures to address temperature-induced degradation of (bare) thin-film III-V solar cells have not yet been developed. Since Cu diffusion is exponentially temperature dependent ($D = D_0 exp(-E_a/kT)$) an accelerated life-time testing (ALT) procedure can be used to accelerate the degradation process. ALT assumes that exposure for a long period of time to a (relatively) low temperature equals a short exposure (few days) to a higher temperature. This can be described with the following equation [29]:

$$\frac{t_{op}}{t_{acc}} = exp\left[\frac{E_a}{k}\left(\frac{1}{T_{op}} - \frac{1}{T_{acc}}\right)\right] \qquad (1)$$

in which k is the Boltzmann constant, E_a the activation energy for the degradation process, T_{op} the regular operation temperature, T_{acc} the accelerated test temperature and t_{op} and t_{acc} the exposure times to the corresponding temperatures. Typically the actual operation conditions (illumination, bias voltage, vacuum) are simulated during such tests [30], [31]. In order to exclude additional (i.e. non temperature-induced) light- and/or electrically-induced degradation mechanisms these operating conditions were excluded in our test procedure. In order to distinguish this procedure from regular ALT we will refer to this adapted procedure as accelerated ageing testing (AAT).

The main difficulty with both ALT and AAT is that it requires a known activation energy. Unfortunately experimental determination is difficult and hence there are very few activation energies reported for solar cell degradation [29], [32], [33] and none of these concern (Cu) diffusion. The European Cooperation for Space Standardization (ECSS) standard for

solar cell structure
> 300µm p-GaAs substrate

a) substrate-based structure

solar cell structure
100nm AlGaInP etch stop
600nm n-GaAs spacer
10nm n-AlAs release layer
300nm GaAs buffer layer
> 300µm n-GaAs substrate

b) ELO structure

solar cell structure
100nm AlGaInP etch stop
300nm GaAs buffer layer
> 300µm n-GaAs substrate

c) substrate etch structure

300nm n-GaAs contact layer
20nm n-AlInP/n-InGaP window
75nm n-GaAs emitter
2000nm p-GaAs base
75nm p-InGaP BSF
50-300nm p-GaAs contact layer

d) n on p cell structure

200nm p-GaAs contact layer
30nm p-InGaP window
400nm p-GaAs emitter
1600nm n-GaAs base
75nm n-InGaP BSF
300nm n-GaAs contact layer

e) p on n cell structure

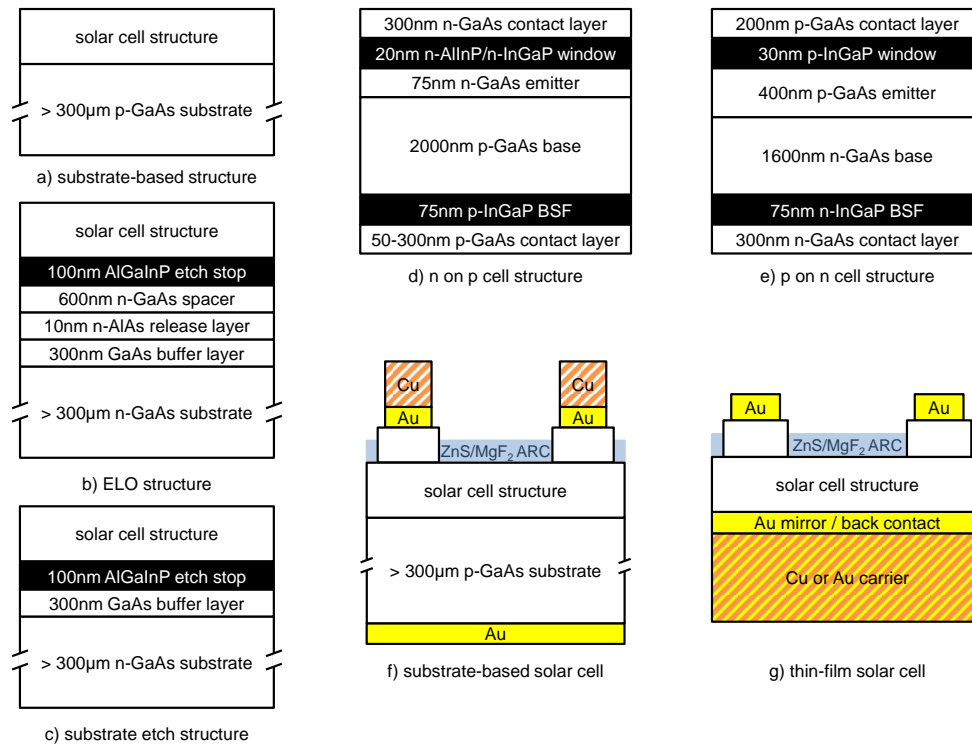f) substrate-based solar cell

g) thin-film solar cell

Figure 1. Schematic representations of a) a substrate-based structure b) an ELO structure c) a substrate etch structure d) a n on p solar cell structure e) a p on n solar cell structure f) a substrate-based solar cell and g) a thin-film solar cell. The solar cell structures (d and e) were grown upright on top of the substrate-based (a), ELO (b) or substrate etch (c) structures unless specified otherwise. The substrate-based structures (a) were processed into substrate-based solar cells (f) and the ELO (b) and substrate etch (c) structures were processed into thin-film solar cells (g).

photovoltaic assemblies and components (ECSS-EST-20-08C [31]) advises to use an E_a of 0.70 eV in calculations for solar cell assemblies (solar cells with cover glass, interconnect and (if used) bypass diode), this appears to be a suitable initial estimate for the activation energy. For an accelerated test temperature of 200°C this E_a of 0.70 eV results in ALT/AAT times of 8.5, 37 and 55 days for simulation of a Geosynchronous orbit mission (GEO, 15 years, max 70°C), a Low-Earth orbit mission (LEO, 10 years, max 100°C) and an extreme scenario (15 years, max 100°C) respectively.

In previous studies [34], [35] a substrate-based model system utilizing a 45% coverage front contact grid was used to investigate the effects of Cu diffusion on GaAs solar cells. In these studies it was shown that at temperatures < 250°C Cu diffusion has no significant effect on the J-V characteristics of (substrate-based) solar cells. In this paper we report the first results of accelerated ageing testing (AAT) at 200°C of thin-film III-V solar cells on Cu and Au carriers and compare them with the AAT results of substrate-based solar cells with and without Cu. Three different types of cells on a Cu carrier were compared: regular ELO n-type emitter on p-type base cells, ELO p-type emitter on n-type base cells and n on p cells obtained by substrate etching. These three different cell types were chosen as Cu diffusion may affect n-type and p-type material differently and Cu is at the n-type side in the substrate-based cells and on the p-type side in conventional n

on p ELO cells; and in order to exclude the potential effects of the ELO process (particularly the sample bending during lift-off) on the diffusion process, substrate etched cells were prepared in such a way that they remained planar during the entire production process.

II. MATERIALS AND METHODS

A. Solar cell processing

The substrate-based, ELO and substrate etch solar cell structures as schematically depicted in figures 1a, 1b and 1c were grown on 2" substrates in an Aixtron 200 MOCVD reactor at Radboud University or obtained from a third party supplier on 4" substrates. The epilayers for the cell structures were grown in upright order and Zn was used for p-type doping and Si for n-type doping, the n-GaAs contact layers were Te-doped. Except for the ELO cells on an Au carrier which were grown inverted and with a Si-doped n-type contact layer. InGaP windows were used for the p on n cells and the cells on an Au carrier, while AlInP windows were used in the n on p cells on a Cu carrier and for the substrate-based cells. The thickness of the p-type contact layer of the n on p cells was varied: 300 nm for the substrate-based cells and ELO cells on an Au carrier, 50 nm for the first series of ELO cells on a Cu carrier (labelled (1)) and 100 nm for the substrate etched cells and the second series of ELO cells on a Cu carrier (labelled (2)).

First a flexible handle was applied to the thin-film solar cell structures after which they were removed from their growth substrates either by epitaxial lift-off [12] or by substrate etching with a 5:1 citric acid (1 kg in 1 kg H_2O) : H_2O_2 (32%) solution. After substrate removal the cell structures were transferred to a ~ 20 μm Au mirror/back contact/carrier or to a ~ 25 μm Cu carrier with a 100 nm Au mirror/back contact (the Au acts as a photon confining mirror [36]–[39]) and then mounted on a temporary glass carrier for further processing. Then a 45% coverage 200 nm thick Au front contact grid was applied by e-beam evaporation. For the substrate-based cells first 45% coverage metal front contacts (100 nm Au or 100 nm Au / 3 μm Cu) and 100 nm Au back contacts were applied by e-beam evaporation. After application of the contacts 6 mm x 6 mm solar cells were created by a MESA etch. A 1:2:10 NH_4OH:H_2O_2:H_2O solution was used for the GaAs layers and a 1:100:200 Br_2:HBr:H_2O solution (thin-film cells) or 37% HCl solution (substrate-based cells) was used for the AlInP and InGaP layers. For the thin-film cells all semiconductor layers were removed, in such a way that the Au mirror / back contact became exposed after the MESA etch, for the substrate-based cells only the front contact, window, emitter and base layers were removed. Then the front contact layer between the metal grid fingers was removed with 2:1:10 NH_4OH:H_2O_2:H_2O solution. Finally a ZnS (42.5 nm)/MgF_2 (88.0 nm) anti-reflection coating was applied by e-beam evaporation. The thin-film samples were then removed from their temporary glass carrier and cut into smaller pieces typically including 3-6 cells with a scalpel.

B. Characterization and accelerated life-time testing

After cell preparation the solar cells were characterized by J-V measurements with an ABET 2000 solar simulator and ReRa Tracer 3.0 measurement software and by EQE measurements with a ReRa SpeQuest system with ReRa Photor 3.1 measurement software. Sets of 3-6 cells were then exposed to stepwise accelerated ageing testing (AAT) in a vacuum oven at 200°C up to a total of 37 days (equivalent to 10 years at 100°C for an E_a of 0.70 eV), for some of the cells the test was extended to 55 days (equivalent to 15 years at 100°C for E_a = 0.70 eV). After each AAT step the J-V and EQE of the cells were measured and for the short-circuit current density (J_{sc}), the open-circuit voltage (V_{oc}), the fill factor (FF) and the efficiency (η) remaining factors (R) were calculated according to:

$$\text{R-parameter} = \frac{\text{parameter value after AAT}}{\text{parameter value as processed}} . \quad (2)$$

The R values were then averaged over the set of cells.

Based on the results of the AAT, a number of cells were selected for transmission electron microscopy (TEM) analysis. These solar cells were covered with a thin Pt protection layer. A cross-section was made and then thinned using focused ion beam milling to allow for cross-sectional TEM analysis of the front contacts. TEM images were obtained with a FEI Titan G2 microscope.

Table I
AVERAGE BEGINNING OF LIFE (BOL) VALUES OF J_{SC}, V_{OC}, FF AND η FOR THE DIFFERENT CELL TYPES.

	J_{sc} (mA/cm^2)	V_{oc} (mV)	FF (%)	η (%)
Au Sub n on p	15.6	1022.4	82.4	13.1
Cu Sub n on p	15.2	970.4	83.0	12.3
Au ELO n on p	15.6	1029.3	83.5	13.4
Cu ELO n on p (1)	16.4	1008.8	83.9	13.9
Cu ELO n on p (2)	16.2	1003.3	82.4	13.3
Cu ELO p on n	14.7	986.7	78.5	11.4
Cu SubEtch n on p	11.4	989.8	82.6	9.4

III. RESULTS AND DISCUSSION

The average beginning of life (BOL) values for J_{sc}, V_{oc}, FF and η of the different cell types are given in table I. The V_{oc} values of ~ 1.00 V and FFs of 82-83% are typical values for reasonable quality GaAs solar cells. The J_{sc} values may at first glance appear to be rather low, but can be accounted for by the 45% coverage front grid which significantly limits the amount of light entering the cells. The somewhat poorer efficiencies of the Cu ELO p on n and Cu substrate etch n on p cells can be accounted for by the fact that device design and cell processing were not yet optimized for these types of samples.

In figure 2 the average J_{sc}, V_{oc}, FF and η remaining factors of substrate-based and thin-film cells with and without Cu are plotted as a function of AAT time. Both the Au and Cu substrate-based cells show little degradation, indicated by the R-η values which remain above 0.97 after 37 days at 200°C. The other remaining factors indicate that this minor decrease in performance is mainly due to decreases in V_{oc} and FF. V_{oc} losses are typically caused by enhanced non-radiative recombination, while decreases in FF may be related to changes in J_{sc}, V_{oc} and resistance. As these decreases occur for both Au and Cu contacts, they must be caused by general (i.e. non Cu specific) temperature-induced degradation mechanisms. These results clearly indicate that, under these conditions, the application of a thick (3 μm) Cu layer in a III-V solar cell does not affect device performance significantly and hence that Cu diffusion from the contact into the active solar cell should not be an issue for application of Cu in space solar panels.

For the thin-film cells on an Au carrier the decrease in efficiency upon AAT is almost 10% after 37 days at 200°C, mainly caused by decreases in V_{oc} and FF. This slightly larger degradation compared to substrate-based cells is likely to be (at least partially) caused by non-optimized thin-film cell processing which is in an early stage of development compared to substrate-based processing. In contrast with the cells on an Au carrier, all cells on a Cu carrier show severe and rapid degradation as is indicated by the R-η values (see figure 2d) which are already below 0.85 after 4.25 days. Although there are differences between the different sample types, the general degradation trends are similar for all cells on a Cu carrier. Initially rapid decreases in all parameters can be observed. Whereas the initial rapid degradation of the n on p cells starts
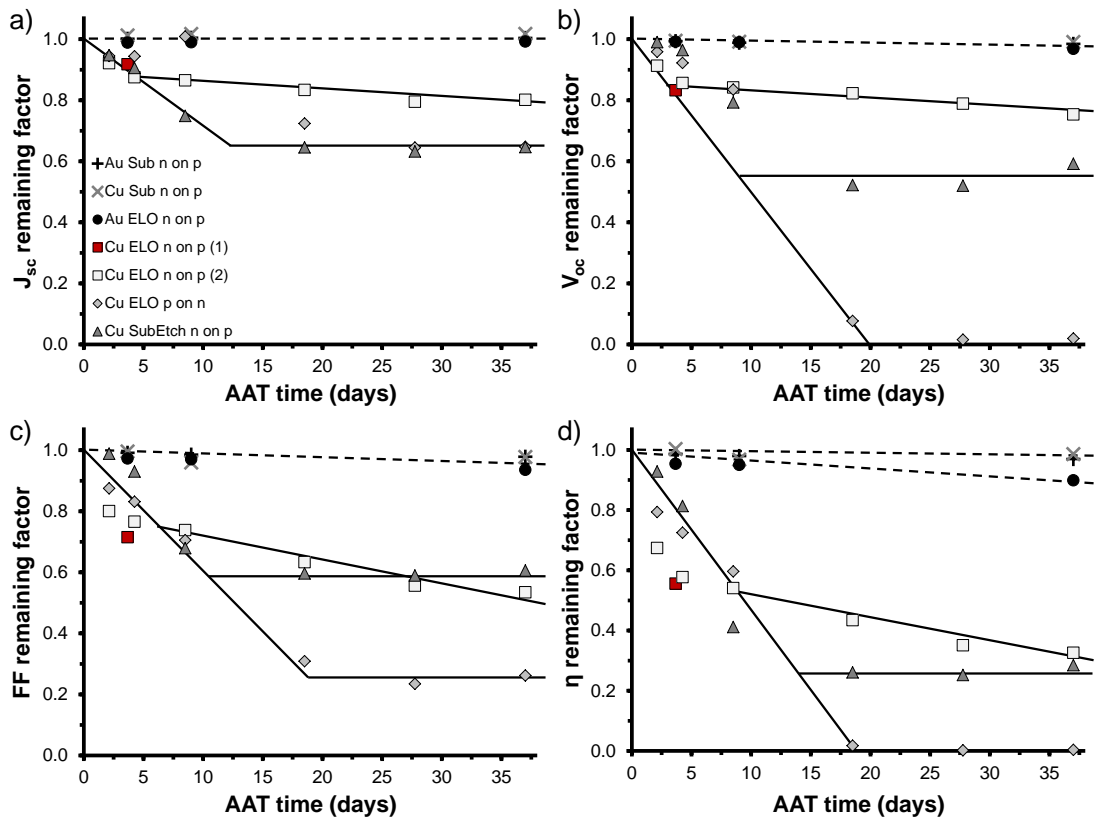
Figure 2. Average a) J_{sc} b) V_{oc} c) fill factor (FF) and d) efficiency (η) remaining factors plotted versus AAT time for n on p substrate-based cells with 100 nm Au front contacts (black + signs), n on p substrate-based cells with 100 nm Au / 3 μm Cu front contacts (grey x signs), n on p ELO cells on an Au carrier (black circles), n on p ELO cells on a Cu carrier (red and light grey squares), p on n ELO cells on a Cu carrier (grey diamonds) and n on p substrate etched cells on a Cu carrier (grey triangles). The dashed lines indicate the general degradation trends for substrate-based cells and thin-film cells on an Au carrier, the solid lines indicate the trends for thin-film cells on a Cu carrier.

to level of after 5-10 days, resulting in R-η values of about 0.3 after 37 days of AAT, the rapid degradation of the p on n cells continues (in particular for V_{oc} and FF), resulting in non-operational cells after 18 days. This hints that n-type GaAs is more prone to Cu diffusion, if Cu diffusion is a process contributing to device degradation. However the fact that the substrate-based cells also have the Cu at the n-type side and hardly show any degradation indicates that there is at least one other degradation mechanism that has a major impact on the degradation process. The fact that both the substrate-etched and ELO n on p thin-film cells show a very similar degradation trend indicates that the manipulation of the thin-film during the ELO process does not inflict damage that is detrimental to the solar cell device operation.

The significant decrease in J_{sc} observed for the thin-film cells on Cu carriers is in sharp contrast with the small changes (typically 1-2%) observed for cells on an Au carrier and substrate-based cells with and without Cu (both in this study and in previous work [34], [35], [40]). This indicates that the degradation process causing the decrease in J_{sc} is specific for thin-film cells on a Cu carrier and therefore of interest for further investigation. The decrease in J_{sc} is (at least partially) caused by a reflectivity loss of the Au mirror that is applied to reflect transmitted and recycled photons back into the solar cell [36], [37], [39]. Figures 3a and 3b show an as processed
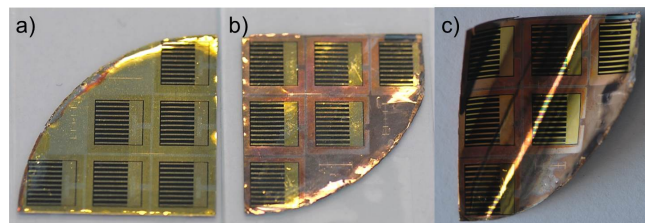

Figure 3. ELO n on p thin-film solar cells on a Cu carrier with 100nm Au mirror a) as processed and b) and c) after 37 days AAT at 200°C. For images a) and b) the samples were placed behind a glass plate to keep them flat. Without this glass plate the samples show significant curvature, in particular after AAT as is shown in image c).

ELO thin-film sample and an ELO thin-film sample after exposure to AAT. Around the cells the as processed sample shows the yellowish colour of the gold mirror/back contact, while the sample exposed to AAT has a reddish Cu colour, indicating that Cu from the carrier has diffused through the Au mirror/back contact during AAT. The same colour change is observed for all samples on a Cu carrier.

TEM images show that the rear interface of thin-film cells on Au and Cu carrier are remarkably different upon exposure to AAT. After 55 days at 200°C the cell on an Au carrier shows a smooth Au/GaAs interface at the back (see figure
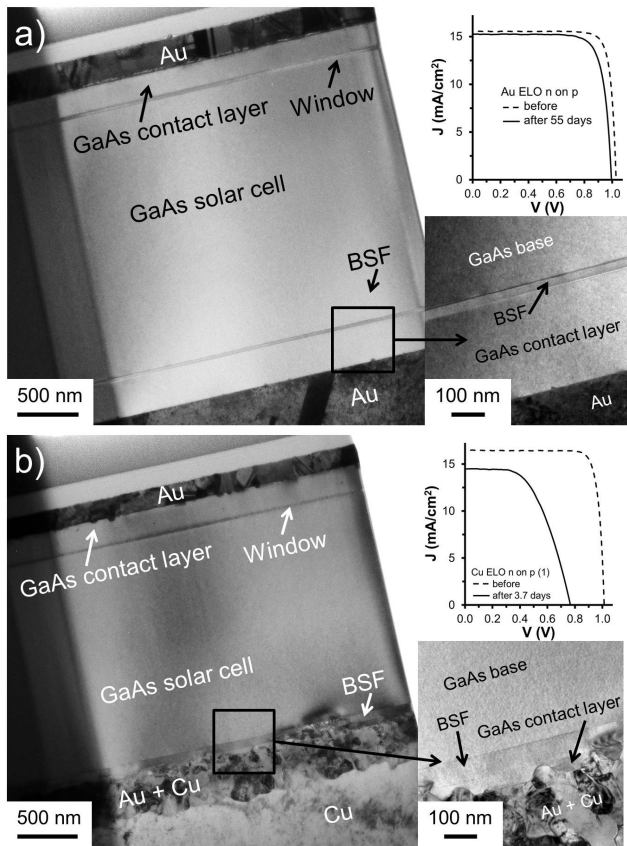
Figure 4. TEM images of a) a thin-film ELO cell on an Au carrier after 55 days at 200°C and b) a thin-film ELO cell on a Cu carrier after 3.7 days at 200°C. The inserts on the right show the J-V curves before (dashed curve) and after (solid curve) AAT and an enlargement of the back of the solar cell.
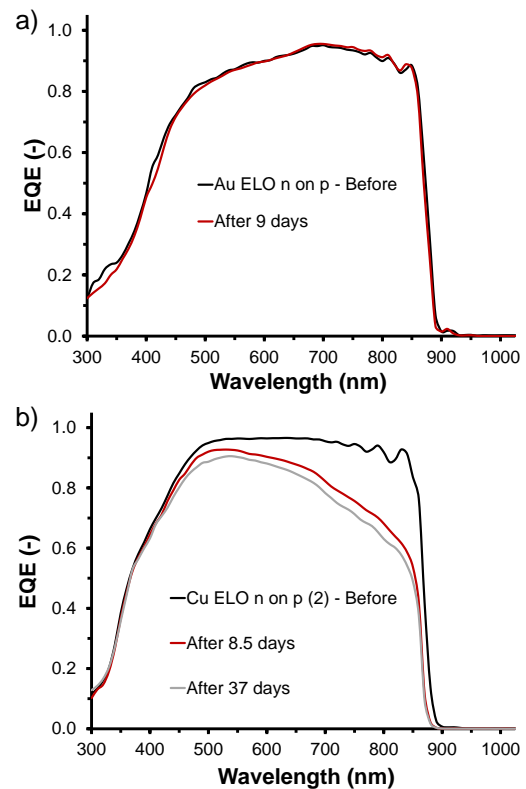

Figure 5. External quantum efficiency (EQE) of a) a thin-film ELO n on p solar cell on a Au carrier before AAT (black curve) and after 9 (red curve) days of AAT at 200°C and b) a thin-film ELO n on p solar cell on a Cu carrier with 100nm Au mirror before AAT (black curve) and after 8.5 (red curve) and 37 (grey curve) days of AAT at 200°C.

4a), whereas the cell on a Cu carrier shows an intermixed Au/Cu layer and a GaAs contact layer that appears to have been almost fully consumed by the Au/Cu matrix already after 3.7 days at 200°C (see figure 4b). This undulation of the metal/GaAs interface and intermixing of the Au and Cu is similar to the undulation and intermixing previously observed for substrate-based cells after 1320 h (55 days) at 200°C (see figure 6b in reference [35]). The intermixing of the Au and Cu is in agreement with the observed colour change of the carrier foil (see figure 3). The intermixing and particularly the undulation of the Au/GaAs interface indicates severe degradation of the Au mirror.

The reflectivity loss of the mirror can also be deduced from the EQE data of the cells. In figure 5a the EQE of an n on p thin-film ELO cell on an Au carrier is plotted before AAT and after 9 days at 200°C and in figure 5b the EQE of an n on p ELO thin-film cell on a Cu carrier is plotted before AAT, after 8.5 days and after 37 days at 200°C. After AAT the interference fringes in the 700-900 nm range caused by the reflection of the mirror [41] have disappeared for the cell on a Cu carrier, but remain visible for the cell on an Au carrier. This clearly shows that in the cells on a Cu carrier the mirror properties of the Au are severely degraded upon AAT. Additionally the EQE data show that the loss in J_{sc} for the cells on a Cu carrier is mainly caused by a reduced collection

efficiency of carriers created by long wavelength (600-900 nm) photons, which are mostly absorbed at the back of the cell. The fact that no reduction in collection efficiency is observed for the cell on an Au carrier suggests that the decrease for the cells on a Cu carrier is Cu related. Two mechanisms contribute to the loss in collection efficiency: reduced reflection of photons by the mirror and diffusion of Cu into the active cell, creating trap levels that reduce the carrier collection efficiency.

Next we discuss V_{oc}. The decrease in V_{oc} of the thin-film cells on Cu carrier is far more severe than the decreases observed for substrate-based cells and thin-film cells on an Au carrier. As decreases in V_{oc} are typically related to increases in the (non-radiative) recombination current and Cu is known to introduce trap levels in the band gap [28] which act as recombination centres, the decreases in V_{oc} are most likely related to the introduction of Cu trap levels as a result of Cu diffusion. The colour change of the metal foil as depicted in figure 3, the intermixing of Au and Cu and the undulation of the Au/GaAs interface observed in the TEM images (figure 4b) and the reduced collection efficiency in the long wavelength range of the EQE (figure 5) all indicate that Cu diffusion indeed takes place. Direct quantitative measurement of Cu diffusion by for example SIMS is difficult as the concentration of Cu causing an electrical effect is most likely significantly below the detection limit of such methods. A problem that was already discussed by Istratov et al. for Cu diffusion in

silicon [42]. However, as the substrate-based Cu cells do not show signs of Cu diffusion there must be a thin-film related process that induces/enhances the diffusion process. The image in figure 3c provides a possible explanation for the enhanced Cu diffusion in thin-film cells, as it shows that the carrier tends to curl upon exposure to AAT. This curling of the sample can be explained by the difference in thermal expansion coefficient between the Cu ($16.5*10^{-6}$ K^{-1}) and Au ($14.2*10^{-6}$ K^{-1}) on the one hand and GaAs ($5.4*10^{-6}$ K^{-1}) on the other. Upon exposure to AAT the metal expands more rapidly, thereby creating stress in the solar cell material. Such stress is likely to induce (micro-)cracks and other defects which are known to enhance diffusion [43], [44].

IV. Conclusions & Outlook

In order to investigate the space compatibility of thin-film III-V solar cells, the temperature-induced degradation of thin-film cells on Cu and Au carriers and substrate-based cells with Au and Au/Cu front contacts was studied by accelerated ageing testing. The substrate-based cells show little degradation with a decrease in efficiency of 3% or less for both Au and Au/Cu contacts (after 37 days, equivalent to 10 years at 100°C for an E$_a$ of 0.70 eV), which indicates that application of Cu in itself is not a threat to the long term device performance. The slight efficiency loss of ~10% of the cells on an Au carrier after 37 days at 200°C, indicates that there is a definite potential for the thin-film III-V cells in space modules. The severe degradation observed for the cells on Cu carriers (efficiency losses typically > 60% after 37 days at 200°C) shows that the carrier material significantly influences the device stability. The fact that both ELO and substrate-etched thin-film cells on a Cu carrier show severe degradation indicates that the ELO process itself does not affect the device performance. At least two factors contribute to the decrease in solar cell performance: thermally induced stress and Cu diffusion. These two factors may influence each other as diffusion processes are likely to be enhanced by damage (such as cracks and defects) induced by thermal stress.

In order to improve long term stability of thin-film solar cells a better understanding of the effect(s) of thermally induced stress on the solar cell / carrier combination is required. As the actual maximum temperature reached during solar cell operation in space is significantly lower (typically max ±100°C, possibly somewhat higher in extreme circumstances) than the AAT temperature applied in this study (200°C) the critical levels of stress induced by the AAT may not be reached during an actual space mission. Lowering the AAT temperature to a value well below 200°C may significantly reduce the thermally induced stress. Additionally, mounting the thin-film cells on a space compatible flexible mechanical support might help reducing the stress effects in the thin-film cells (i.e. preventing the thin-film cells from bending upon exposure to thermal stress). Furthermore, a solution for the intermixing of the Au mirror and Cu carrier and diffusion of Cu into the device has to be found. This could be achieved either by implementation of a barrier that prevents intermixing of the Au mirror/back contact and the Cu carrier or by replacement of the Cu carrier with a different (metal) carrier.

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