Overcoming Degradation Mechanisms in CdTe Solar Cells

Final Report

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NREL Technical Monitor: Bolko von Roedern

Prepared under Subcontract No. AAK-8-17619-15
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Abstract

The stability of CdTe solar cells, with special focus on possible effects of diffusion from the contact to the absorber towards other cell components, has been studied. Both whole cells and test systems containing only the ohmic contact and the absorber or only the window were used. Based on results obtained early on in this research, i.e., observing
- Fast Cu diffusion from Cu/HgTe/graphite back contacts appears enhanced by grain boundary diffusion
- Grain boundaries in CdTe/CdS cells are not fully passivated and are expected to be electrically active.
- CdTe films in the cells have significant porosity
- Decrease in cell performance correlates with increase in the heterogeneity of the spatial current collection efficiency of the cell
- Differences between module and cell stability can be due to the difference between encapsulated and air-exposed systems

we concentrated our efforts on

- **a**- finding an alternative ohmic back contact.
- **b**- isolate factors involved in cell degradation and find ways to
  - avoid cell degradation and
  - to get performance recovery of degraded cells.

For **a**- NiTe₂ was found to be a promising back-contact material.

For **b**- we find that Cu as such is not the dominant factor in the most common and quickest type of degradation of these cells. An additional factor appears to be formation of an oxide film on CdTe grains, which can be associated with the formation of the additional back contact barrier that has been deduced from electrical characteristics (evolving roll-over behavior of the current-voltage (I-V) curves) of the cells.

Further observations were:
- Cell degradation appears to be promoted by H₂O, O₂ and illumination, in that order.
- Less efficient cells are less stable than more efficient ones
- Some cells have been stabilized by heating in ultra-dry and O₂-free inert atmosphere (N₂ was used by us) before use, against subsequent degradation.
- Cells can recover by heating in dry N₂ or by sitting on the shelf in ambient atmosphere

Deliverables (cells that were prepared and tested here) are described and included with this report. In additions, suggestions are offered for further work by the team.
Introduction

The background and the statement of the problem to be solved for this project are described and discussed in great detail in our review paper in Solar Energy Materials and Solar Cells [1].

Specifically, the objectives of this project were:

To investigate the back “ohmic” contact to the p-CdTe film in the CdTe/CdS cell, with the aim of improving the stability of this contact while maintaining performance.

The following approaches were among those involved in this study:

(1) Separate back contact lifetime performance from overall cell lifetime performance.

(2) Investigate new back contact materials.

(3) Explore use of surface treatment of the CdTe to modify the contact.

In all our work we relied on cell structures from University of South California (USF) and from First Solar (FS) Inc., to which we added our own back contacts, either based on raw material from USF or home-made. The cells that resulted may therefore not be comparable to those prepared by USF and FS. Still, it should be clearly realized that, within the physical and chemical parameter spaces used, the materials chemistry and physics for the CdTe, CdS, conducting oxide parts and their interfaces, as found by us, will apply to these as well as to other structures composed of these materials unless clear materials scientific evidence to the contrary can be brought.

Our efforts concerning approach (1) concentrated on Cu-containing contacts and led to the conclusion that Cu is not automatically the major factor involved in cell degradation. This means that the back contact can lose Cu to the cell, while cell performance can be maintained, and that the cell can lose performance with no significant changes that can be observed in the composition of the back contact and its neighboring region. The experimental data on which these conclusions are based are mainly results from Secondary Ion Mass Spectroscopy (SIMS) analyses of degraded and of non-degraded cells and partial cell structures. These are detailed in our publications in Advanced Materials and in MRS Proceedings [2, 3].
Our efforts concerning approach (2) concentrated on NiTe$_2$–based contacts, prepared using a so-called Ni-P bath. The very promising results are described in our publications in *Thin Solid Films* [4] and in *Journal of the Electrochemical Society* [5].

Our efforts concerning approach (3) were unfortunately, due to a lack of time and funds, cut short. Apart from the use of atmospheres with and without O$_2$ and / or H$_2$O, the more detailed work had to be left at the stage that we reached during the beginning of the second year, and which we reported on in our second annual report, pages 13-20 [6].
Cell stability

Measurements of cell parameters after various stress conditions.

A number of experiments were carried out to investigate the effects of different types of stress on the cell output parameters. These are:

- Heating in air at 200°C  
  (Figs. 1, 2)
- Heating in humid N₂ at 200°C  
  (Fig. 3)
- Heating in dry N₂ at 200°C  
  (Figs. 4-6)
  
  All these measurements were carried out under exclusion of light (for earlier experiments, under stray light).
- Heating in dry N₂ at 200°C under simulated 1-sun illumination (open circuit)  
  (Figs. 7-10)
- Heating in dry N₂ at 200°C in the dark followed by heating in dry N₂ at 200°C under simulated 1-sun illumination  
  (Figs. 11, 12)
- Storage (including recovery; see below)  
  (Figs. 13-17)

The effects of stress are shown as bar graphs, with results given for cells with the highest initial efficiency and the average of cells within certain initial efficiency ranges. Additionally, representative current-voltage characteristics are given and, for two cases, the time evolution of the change in cell output parameters.

As noted in [2], we use conditions to induce maximum degradation in minimum time, something that can, admittedly, activate additional modes of degradation. At the same time, though, there can be little doubt that mechanisms operative at room temperature, up to the maximum operating temperature of cells in modules in the field, will be accelerated under such conditions. The actual extent (factor) of acceleration is not known. We did some experiments that suggest that the extent of acceleration, when working under dry conditions is less than the rule of the thumb often-used (2 x acceleration for every 10 degrees C temperature increase). However, we note that humidity will most likely increase stress acceleration significantly.
**Heating in air at 200°C**

All cells degrade severely (Fig. 1) when heated in air. Higher efficiency cells degrade more than lower efficiency cells, i.e., after degradation there is a smaller variation in cell parameters of different cells than before. The degradation is caused mainly by comparable relative drops in Fill Factor (FF) and short-circuit current (I_{SC}), and a smaller drop in open-circuit voltage (V_{OC}). Degradation occurs initially relatively fast (Fig. 18) and some improvement subsequently occurs after further stressing, followed by further, slow degradation upon continued stress. I_{SC} remains low after the initial drop, while FF and V_{OC} increase after the initial drop, which reduces the overall efficiency loss.

Fig. 2 shows a typical current-voltage characteristic, in the dark and under illumination, of a cell before and after air stressing. The unstressed cell shows a classic diode relationship between dark and illuminated characteristics, with no crossover between the dark and illuminated plots at high forward bias. The dark diode characteristics do show rather large currents in both bias directions, which could be explained by shunting; however, this does not affect the photovoltaic characteristics much. (This behavior is not typical; most cells show low reverse current). After stressing, the most obvious change is in the much higher resistance, both in the dark and under illumination, as seen at high forward bias, and the roll-over in the illuminated characteristics near V_{OC}, as well as cross-over between the light and dark forward characteristics. The latter is usually ascribed to photoconduction, and it is likely that this occurs in the CdS, since most of the migrating Cu resides in the CdS (see SIMS data below and in [2, 3]) and Cu is known to cause photoconductivity in CdS. The high resistance of the stressed junction may be due both to increased resistance of the CdS and, most likely the main factor, partial loss of ohmic contact to the CdTe due to loss of Cu from this contact. The roll-over as well as the drop in both I_{SC} and V_{OC} suggest increase in recombination in the cell.

**Heating in humid atmosphere or in dry air at 200°C.**

A few experiments were carried out under such conditions. In the case of Fig. 3 water vapor was deliberately added to N₂. While significant degradation was found, it was smaller than in air. Degradation upon stressing in dry air was also smaller than in humid air. This means that air and humidity together induce a more severe degradation than each of them does separately.
**Heating in dry N₂ at 200°C.**

There was an element of irreproducibility in these results, beyond obvious statistically significant differences between the FS plates from which the test cells were prepared (99012-936L4 and to some extent also 990 20147K3 gave better results than others). Several of the cells were essentially completely stable (frequently a small increase in efficiency was measured, mainly due to increase in $I_{SC}$ and $V_{OC}$; FF often dropped slightly, particularly for poorer cells) after many tens of hours at 200°C in N₂ (Figs. 4, 5). The cells degraded much less than upon stressing in air. Also, there was a wide variation in the degradation from cell to cell. As for air-stressed cells, the main drop upon stress was in FF and $I_{SC}$, with a smaller, but appreciable drop in $V_{OC}$ (Fig. 6). The degradation appears to be due mainly to increase in cell resistivity.

**Heating in dry N₂ at 200°C under illumination (open circuit).**

Heating in N₂ under illumination (“light stress”) resulted in some degradation (Fig. 7, 8), less for good cells and considerably more for poorer cells. Over-all, there was no obvious difference in the change in parameters between stress in the dark and under illumination for good cells, except for the important fact that, whereas dark stressed cells were sometimes stable, light stressed ones invariably degraded to some extent (but see below). There is also a tendency for the main degradation parameter to be the FF, rather than an approximately equal mixture of FF and $I_{SC}$, particularly for earlier cells which were the ones (made from plate 936L4) that were usually stable to stress in the dark in N₂.

The drop in efficiency and FF (the latter being the main cause for drop in efficiency in this case) with stressing occurs gradually during ca. 50 h (in contrast to air-stressed cells, which degrade relatively rapidly initially) (Fig. 18). $V_{OC}$ is stable initially (the slight increase is probably not significant) while $I_{SC}$ drops initially (as for air stress, only much less) and is then more or less stable.

Current-voltage characteristics before and after stress are shown for two samples, a relatively stable one and a relatively unstable one. This was judged to be important because, considering the wide variation in degradation, no single representative example can be given (Figs. 9, 10). Again, the qualitative changes after stressing are similar to previous examples.

Some cells that were stressed in N₂ under illumination were found to recover their original performance after some time, either under ambient conditions or heating in N₂ in the dark, some-
times even after severe degradation. This recovery was seen for cells stressed under illumination for relatively short times (ca. 20 h). Cells stressed for much longer times (> 60 h) did not recover. This implies that a reversible change occurs at shorter times but becomes irreversible (to storage at ambient) upon long degradation times (Fig. 17).

**Heating in dry N₂ at 200°C in the dark followed by heating in dry N₂ at 200°C under illumination.**

Here we divide the results into two groups: those that were stable after heating in the dark and those that were not.

**Stable cells after dark N₂ stress.**

These cells (made from 936L4) did not degrade after stress under illumination, in contrast to cells that were stressed under illumination from the beginning. Such cells are therefore stabilized by initial heating in N₂ in the dark.

**Cells which degraded after dark N₂ stress.**

When stressed under illumination, these cells recovered some (about 50%) of what they had lost after dark N₂ stress (Fig. 11). The recovery was mostly in I_SC, a small amount in V_OC, and essentially not at all in FF, although there was considerable variation in such division of parameters. The changes in the current-voltage characteristics after the light stress are shown by the representative example in Fig. 12. The resistivity increases, as can be seen from the increase in slope of the dark forward plot. Also the pronounced cross-over (photoconductivity, presumably) exhibited after dark stress is strongly reduced after subsequent light stressing.

**Storage at room temperature.**

These experiments were carried out either under normal ambient or in a dessicator.

**Normal ambient (humid conditions).**

Small drops in all parameters were often observed after several days, but mostly in FF (Fig.13, 14). Current-voltage characterization showed that this drop was due to slightly increased resistivity (Fig. 15). Note that no roll-over occurs under these conditions; the increase in resistivity appears to be the only important factor in the small degradation which occurs during this type of storage.

**Dessicator (dry conditions).**

There is no decrease, but rather an increase, mainly due to a change in short circuit current.(cf. Fig. 13, 16).
Cells from earlier experiments that tended to be stable under dry N₂ stress, were also found to be reasonably stable upon storage under normal atmospheric conditions (Fig. 14).

**LBIC**

Light beam-induced current (LBIC) measurements showed that degradation is accompanied by spatial heterogeneity of the cells (cf. also [3]). Fig. 19 shows LBIC images of (a) a non-stressed cell; (b) the cell stressed at open circuit under illumination and (c) a dark stressed cell. The light stressed cell clearly shows more dark areas (lower collection efficiency) than the others. Since \( I_{SC} \) is, in general, less affected by degradation than the fill factor, or at least appears less representative of changes in efficiency that the fill factor, such measurements repeated close to maximum power should show this effect more clearly.

**SIMS**

In view of our conclusion that Cu *per se* does not appear to be a direct cause for the earliest signs of instability of the cells, it became of interest to look at the fate of Ag, used often to complete the back contact to the CdTe, on top of the Cu-containing C paste. For this purpose cells were prepared with Ag, with Au and with Al as the completion stage for the back contact and within 1-2 days subjected to SIMS analyses. Because of technical difficulties, Al deposition on the cells was not comparable to the application of Au by evaporation and of Ag via a paste. Time restrictions prevented further work with Al and we will thus concentrate on the Ag and Au metallized cells. Their initial performances were roughly comparable. The cells were stressed in dry N₂ in the dark or the light and in air. SIMS measurements were made before and after stress.

*Au* The SIMS analyses show some accumulation of Au at the interfaces in comparable concentrations in ALL cells, whether or not Au was used as contacting material. Therefore this can be traced to Au present in the material rather than to Au penetrating from the back contact (cf. Durose et al. in [7], who reported on large amounts of Cu in so-called Cu-free CdTe/CdS cells). The traces of Au accumulated at the CdS/SnO₂ and SnO₂/glass interfaces. In stressed cells Au was also found in the back contact but not in the CdTe bulk. The presence of Au in ALL cell types, especially its presence near the glass, suggests that its origin is there.

*Ag* penetrates in a manner similar to that observed earlier for Cu, with accumulation in the CdS and at the CdS interfaces (Au and Al do not show this behavior). Similar concentrations of
Ag are found in stressed than in unstressed cells in the CdS. In air stressed or light-stressed cells Ag concentration in the CdTe is higher than in the unstressed samples, suggesting oxide assisted in-diffusion, or, maybe even more importantly, grain surface adherence (Fig. 20).

_Cu_ concentration profiles are as previously reported [2, 3]. In addition Cu content is higher (in all parts of the cell, but most significantly in CdS) in light-stressed in N2 or dark stressed in air cells than in dark N2 stressed ones. All Cu profiles closely follow the S ones.

_Sn_ is seen to penetrate rather deep into the CdTe, something that appears to be more than just an expression of surface roughness or other experimental artifacts. This penetration is not significantly affected by stress.

In summary the SIMS data

- confirm earlier results concerning Cu (+Cl) accumulation irrespective of stress, adding that Ag behaves similar to Cu, while Au does not.
- do not give evidence for a clear correlation between Cu (or Ag) distribution and cell performance.
**Major conclusions concerning stability of cells**

Conclusions concerning stability of cells with Cu-containing back contacts, after stress at 200 ºC under various conditions are (all refer to the C/Cu/HgTe type of back contact):

1. Stabilization of cell performance has been possible (at times, see -5- below) by heating in dark, in dry N₂. In particular, immunization against time- (storage) and light stress- induced degradation were shown.

2. Cells are least stable after stress in air + H₂O, therefore air and H₂O should be avoided in order to improve CdTe cell stability.

3. We confirm that cells that have degraded may recover by heating in dry N₂ or even by sitting on the shelf

4. Ag and Cu appear to behave similarly in the cell, in terms of their distribution after being applied to the back contact. Neither Ag, nor Cu appears to be by itself the determining factor for degradation as cells containing either or both have been made that are stable.

5. There remains an important aspect of IRREPRODUCIBILITY of stress effects in cell stability and efficiency with some connection between them. This implies that in the time that was available to us we could not identify all factors that are involved in stabilizing the Cu-containing back contact containing cells.

6. Degradation is characterized, among other things, by increasing spatial heterogeneity in photovoltaic activity.

7. In most cases the FF and the I_{sc} are the parameters with closest correspondence to efficiency decrease. This corresponds to an increase in cell resistance, something that can be due to contact degradation.
8. **Roll-over** in the dark, as well as high dark, series resistance are, *per se*, not detrimental to cell performance. They can result from increase in CdS (and to lesser extent, probably CdTe) resistivity, which is compensated for by appreciable photoconductivity. As long as roll over in this case occurs at forward bias higher than the $V_{OC}$, it will not affect cell performance.

9. In air strong roll-over is seen, stronger than in the case of light-stressed cells. This suggests that we create a barrier for hole transport (near the back contact?).

10. Preliminary XPS data on both CdTe and CdS (not shown) indicate oxide formation on the CdTe AND on the CdS, not only upon air stress but also upon light stress.

11. Preliminary photoresponse data (not shown) suggest that most of the changes in short-circuit current upon degradation (and stabilization) occur in the red region between 750 and 800 nm.
Electroless-deposited NiTe₂ and Ni-P back contacts to p-CdTe

This study arose based on some earlier work on electroless Ni contacts to p-CdTe [8]. The annealing conditions used for this contact (250°C for 90 min in air) suggest it to possess high stability. Details of this study are given in [4, 5] and a brief synopsis of the main results is given here.

Normally, electroless deposition of Ni, using hypophosphite, gives a Ni-P alloy with varying amounts of P. Deposition of such films onto the CdTe gave, in our hands, poor contacts (low fill factor and V_{OC}). However, by carrying out the deposition under conditions where the Ni formation was slow (low hypophosphite concentration) and at high temperatures, the slowly formed Ni reacted with elemental Te present at the CdTe surface. The CdTe surface was found to be enriched in Te by an etching action of the deposition solution, and this enrichment allows the known solid state reaction between slowly-formed Ni (presumably in a finely-divided state) and Te to occur to give NiTe₂, a metallic conductor. Another possible mechanism is the exchange reaction between Ni^{2+} and CdTe, although this reaction, by itself, is very slow.

The optimum annealing treatment for the NiTe₂-CdTe junction was 200°C in air for 90 min. This treatment attests to the stability of the junction. Water vapour was found to reversibly degrade the junction (i.e., the performance recovered on storage in dry air).
**Deliverables**

Cells that degraded after dark N₂ stabilization 10.9.01S6; 3.9.01S4 (936L4 plate)
23.8.01S9; 23.8.01S6 (31904I5 plate)

Cells that did NOT degrade > dark N₂ stabilization, 936L4-5; 936L4-6; 936L4-25;
936L4-28 (dark + subsequent light N₂ stress)

Cells that recovered 936L4-11
936L4-12
936L4-10
936L4-9

Those cells may be worthwhile objects for further analyses by the new team, such as XPS, SIMS,
Advanced I-V analyses, photoresponse.

Tables of all relevant CdTe/CdS cells stability results are available upon request.

**Suggestions for future work**

- Work in clean room and in controlled humidity environment;
  it will be worthwhile also to control CO₂ concentration

- Apply back contacts in more controlled manner than use of paste.

- Use sub-monolayers of molecules with voltage-controllable dipole moments.

- Perform LBIV and equivalent measurements under ca. maximum power;
  should be more sensitive to degradation than I_{SC}.

- Do stress measurements on NiTe₂ contacts with encapsulant against water.

- Do SIMS Cu measurements **immediately** after cell preparation
Publications and Presentations at major meetings


9. E-MRS 2000 spring meeting, Strasbourg, France (2 presentations).

References


Fig. 1

**air stress (Dark)**

**highest $\eta^{\text{initial}}$ cell**

**$\eta^{\text{initial}} >9\%$ cells’ average**

$\eta^{\text{initial}} = 6.7\div8.8\%$ cells’ average

**Figure 1**

Bar graphs, showing the change in cell output parameters (efficiency, fill factor, short circuit current and open circuit voltage, all under simulated 1 sun illumination), after air stress at 200 °C in the dark. Data are shown for the cell with the best initial efficiency, and for the average of cells within the indicated range of initial cell efficiencies.
Figure 2

I-V characteristics in the dark and under simulated 1 sun illumination before and after 43 h air stress at 200 °C.
Figure 3

Bar graph as in Fig. 1, showing the change in cell output parameters (efficiency, fill factor, short circuit current and open circuit voltage, all under simulated 1 sun illumination), after stress in wet N₂ at 200 °C. Data are shown for one of the cells tested.
Figure 4

Bar graphs, as in Fig 1, for stress in the dark in an atmosphere of N₂.
Figure 5

Bar graphs, as in Fig 4, but early set of experiments using only samples from plate 936L4.
Figure 6
I-V characteristics in the dark and under simulated 1 sun illumination before and after 43 h stress in the dark in N2, at 200 ºC.
Figure 7
Bar graphs, as in Fig 1, for stress under illumination in an atmosphere of N₂.
**Fig. 8** Light stress (early experiments)

$\eta_{\text{initial}}^{\text{highest cell}}$ and $\eta_{\text{initial}>9\% \text{ cells' average}}$

$\eta_{\text{initial}} = 6.5\% - 8.8\% \text{ cells' average}$

**Figure 8**
Bar graphs, as in Fig 7, but early set of experiments using only samples from plate 936L4.
After light stress, N2 flow, 200ºC, 22h

I-V characteristics in the dark and under simulated 1 sun illumination before and after 22 h stress under illumination in N2, at 200 ºC, for a sample that shows strong degradation.
Figure 10
As Fig. 9, but for relatively stable sample.
Figure 11

Bar graphs, as in Fig. 1, for stress in the dark, followed by stress under illumination, both in an atmosphere of N₂.
Figure 12

I-V characteristics in the dark and under simulated 1 sun illumination, after 43 h stress in the dark and after subsequent stress of 44 h under illumination, both in N$_2$ at 200 °C.
Figure 13
Bar graphs, as in Fig. 1, for samples that were stored at room temperature under atmospheric ambient conditions in the dark.
Figure 14

Bar graphs, as in Fig.13, but for earlier samples taken only from plate 936L4, and stored in a chemical hood under a flow of air.
Figure 15

I-V characteristics in the dark and under simulated 1 sun illumination, after 7 days storage under ambient atmospheric conditions at room temperature.
Figure 16

As Fig. 15, but for a sample stored in a desiccator for nearly 6 days.
Figure 17
Bar graphs, as in Fig. 1, for earlier samples taken only from plate 936L4, showing their performance after stress in N₂ under illumination (top left and right and bottom left) or after stress in N₂ in the dark (stray light; bottom right), and subsequently let to recover by storing in the hood (top left and bottom left and right) or by stressing in N₂ in the dark at 200 °C (top right).
Figure 18

Time evolution of cell degradation, in terms of the output parameters of two cells, one stressed in air and one stressed in N₂ under illumination.
Fig. 19

Figure 19
Front-wall LBIC images of three completed cells, supplied from NREL, recorded following; (a) no stress, (b) stressed under illumination at open circuit and (c) stressed in dark at \( V > 0 \). Cell (b) had exhibited increased degradation with respect to cell (c).
Ag SIMS profiles in Ag-metallized cells

Figure 20
SIMS depth profiles for the Ag-metallized cells, air-stressed and N₂-stressed. Experimental details are as given in [2].
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This report describes the stability of CdTe solar cells, with special focus on possible effects of diffusion from the contact to the absorber towards other cell components. Both whole cells and test systems containing only the ohmic contact and the absorber or only the window were used. We found that NiTe₂ is a promising back-contact material. We also found that Cu as such is not the dominant factor in the most common and quickest type of degradation of these cells. An additional factor appears to be the formation of an oxide film on CdTe grains, which can be associated with the formation of the additional back-contact barrier that has been deduced from electrical characterization. Further observations were: Cell degradation appears to be promoted by H₂O, O₂, and illumination, in that order; less efficient cells are less stable than more efficient ones; some cells have been stabilized by heating in ultra-dry and O₂-free inert atmosphere (N₂ was used by us) before use, against subsequent degradation; and cells can recover by heating in dry N₂ or by sitting on the shelf in ambient atmosphere.