

Recent progress on CdTe/CdS thin film solar cells

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Received 12 December 2003; received in revised form 12 May 2004; accepted 15 July 2004

Available online 21 September 2004

Communicated by: Associate Editor T.M. Razykov

Abstract

The CdTe/CdS thin film solar cell is the most suitable to be fabricated on the form of thin films. The processes used to make all the films, which compose the cell, are quite simple and fast. An efficiency of 16.5% has been reached on laboratory scale and modules of $0.6 \times 1.2 \text{ m}^2$ with efficiency larger than 8% are now fabricated and commercialized. A strong contribution to the development of this type of solar cells has been given by the Parma University group with the discovery of a new ohmic back contact for CdTe which is very stable in respect to any other ohmic contact used for CdTe, and by the development of a new all dry process to make the cell. An efficiency of 15.8% has been recently obtained on a 10^{-4} m^2 soda-lime glass without using any copper or any other metal of the first group of the periodic table of the elements at the back contact.

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Keywords: Thin film; CdTe; CdS; Solar cells

1. Introduction

Bonnet and Rabenhorst (1972) published an interesting paper on CdTe/CdS thin film solar cells in 1972 reporting an efficiency of 6%. The 10% efficiency value was overcome by Tyan and Perez-Albuern (1982) and finally an efficiency of 15.8% has been reached by Ferekides et al. (1993). Very recently the group of NREL reported a record efficiency of 16.5% (Wu et al., 2001). However, all these groups used as a back contact for CdTe a material containing Cu, which is known to be a fast diffusor in CdTe. The diffusion of Cu could be

beneficial for the solar cell since it can dope CdTe p+ giving a momentary increase in the efficiency but, in any case, at long run the efficiency is doomed to decrease since Cu can segregate at grain boundaries (Dobson et al., 2000; Baetzner et al., 2001). In order to avoid Cu, a new contact, namely Sb_2Te_3 , was developed at the University of Parma (Romeo et al., 2000a).

Sb_2Te_3 is a p-type material and exhibits a resistivity of $2 \times 10^{-2} \Omega\text{m}$. We verified that Sb_2Te_3 makes an ohmic contact with CdTe. However, since this contact does not diffuse into CdTe (giving a high stability to the solar cell), it does not dope CdTe and so the efficiency that can be obtained is lower than the 16.5% NREL record.

In any case, with Sb_2Te_3 contact, made directly on a non-etched CdTe surface, we were able to obtain a stable efficiency of 15.8%.

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2. The structure of CdTe/CdS solar cell

The CdTe/CdS solar cell is composed of 4 layers that are:

1. a transparent and conducting oxide (TCO) which acts as a front contact;
2. a CdS film which is the so called window layer;
3. A CdTe film which is the absorber layer made on top of CdS;
4. the back contact on top of CdTe (Fig. 1).

3. The transparent conducting oxide (TCO)

The characteristics that are required in order for a TCO to be used as a front contact for CdTe/CdS thin film solar cells are:

1. a high transparency, better than 85% in the wavelength region of interest (400–860 nm);
2. a low resistivity on the order of $2 \times 10^{-2} \Omega\text{m}$ or a sheet resistance less than $10 \Omega_{\text{square}}$; ¹
3. a good stability at the maximum temperature (500 °C) at which one of the layer, namely CdTe, is prepared. This means that no diffusion from the TCO into the layers deposited subsequently must happen.

Several types of TCOs have been used and tested for the CdTe/CdS solar cell. Among these:

1. Fluorine doped SnO_2 , which is a quite stable material but it exhibits a sheet resistance of about $10 \Omega_{\text{square}}$ or more that could be not low enough for this kind of solar cells.
2. Tin doped In_2O_3 (ITO), which exhibits a low resistivity ($2.5 \times 10^{-2} \Omega\text{m}$) but we found out that some In can diffuse into CdS and/or CdTe when this material is used as a front contact. In this case a SnO_2 buffer layer can be used as a diffusion barrier (Visoly-Fisher et al., 2003).
3. Cadmium stannate (Cd_2SnO_4) which exhibits both the characteristics of high transparency and low resistivity but it is difficult to be handled since, as we found out, it is quite hygroscopic.
4. Finally we developed a new TCO that is fluorine doped In_2O_3 which exhibits all the characteristics which are needed. This material is prepared by sputtering using a target of pure In_2O_3 supplied by Cerac. The doping with fluorine is done by introducing in the sputtering chamber Ar containing $\approx 5\%$ of

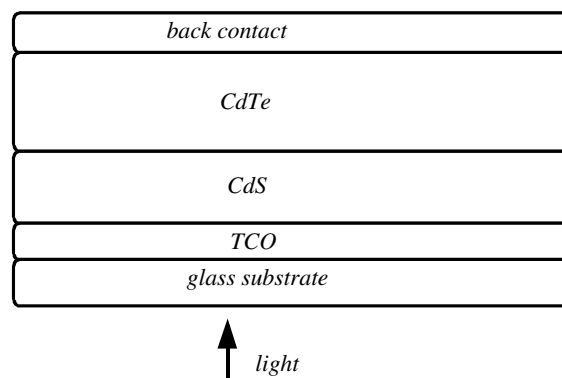


Fig. 1. Sequence of the layers constituting the solar cell. The structure of CdTe/CdS solar cells is of the superstrate arrangement, which means that the solar light enters the cell through the substrate glass.

CHF_3 (trifluoromethane). This gas is decomposed and ionized in the sputtering discharge freeing F which, being strongly electronegative, is directed to the substrate which is the positive electrode and dopes In_2O_3 during the growth. This material exhibits a resistivity of $2 \times 10^{-2} \Omega\text{m}$, a transparency better than 90% between 400 and 800 nm and is quite stable at a temperature of 500 °C as it has been verified by the good stability of CdTe/CdS solar cells grown on it (Romeo et al., 2003).

4. The CdS layer

CdS is prepared by several methods that are evaporation from a single crucible, sputtering, chemical bath deposition (CBD) and closed space sublimation (CSS).

All these methods are suitable to prepare a CdS layer that, when used in the CdTe/CdS solar cell, can give efficiency larger than 10% even though the highest efficiency (Ferekides et al., 1993; Wu et al., 2001) was obtained by using a CdS prepared by CBD. The choice of the CBD method was probably due to the fact that CBD makes a very compact film that covers perfectly the TCO layer. However it must be considered that CBD is not suitable for large-scale production since it is not fast and gives a waste that needs to be recycled. In our case we prepared CdS by sputtering or by closed space sublimation (CSS). CdS by sputtering was grown in presence of fluorine by introducing in the sputtering chamber Ar containing 3% of CHF_3 . Despite fluorine is a donor for CdS, we did not see any lowering of the CdS resistivity when it was grown in presence of fluorine.

CdS by CSS was prepared at a pressure of 10^3 Pa in an atmosphere composed of 50% of Ar and 50% of O_2 .

¹ Ω_{square} is the sheet resistance of a 1-cm² film.

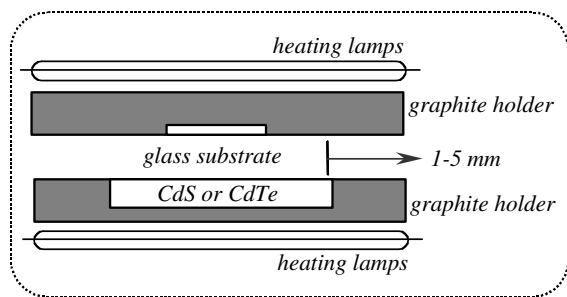


Fig. 2. Outline of the close-spaced sublimation system.

The CSS method (Fig. 2) consists in putting the sublimation source (in this case CdS) very close to the substrate ($2\text{--}4 \times 10^{-3}$ m) and doing the evaporation in an atmosphere that could be inert (Ar) or reactive (Ar + O₂). The CSS has the characteristic of allowing the deposition of materials at a substrate temperature much higher than that used in vacuum evaporation. As an example, when CdS is deposited by evaporation in high vacuum, the substrate temperature must be not much higher than 150 °C because, at temperatures higher than 150 °C, S has a very high evaporation rate and it is re-evaporated before combining with Cd.

When CdS is made by CSS, a substrate temperature as high as 500 °C can be used since the sticking of CdS is assured by the gas pressure that is maintained in the chamber and by the fact that the substrate is very close to the source. Furthermore, the deposition of materials at a high substrate temperature guarantees high crystalline quality, due to the high adatoms superficial diffusion coefficient.

We found out that both CdS(F) by sputtering or CdS(O) by CSS are suitable to make high efficiency cells if they are treated for 20 min. in an atmosphere of Ar containing 20% of H₂ at 400 °C substrate temperature (Romeo et al., 2003, in press). We explained this fact by considering that CdS(F) contains CdF₂ segregated in the grain boundaries and in the surface. While the CdF₂ segregated in the grain boundaries is useful to passivate the grain boundaries, the CdF₂ segregated in the surface has to be removed in order to allow CdS to interact with CdTe thus forming a good CdTe/CdS junction.

The same happens when CdS is made in presence of O₂ but in this case CdO and/or CdSO₃ is formed in the grain boundaries and in the CdS surface. CdO and/or CdSO₃ formed in the surface is removed by the H₂ treatment.

The thickness of the CdS layer is, in both cases, 80 nm.

5. The CdTe layer

CdTe is a material that exhibits a forbidden gap of 1.45 eV very close to the maximum of solar energy con-

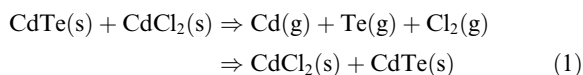
version. Besides, its gap is direct and as a consequence its absorption coefficient is higher than 10^7 m^{-1} for energy larger than the forbidden gap. This means that only a few microns of material are enough to absorb all the light. A practical efficiency of 18.5% could be expected for this material with an open-circuit voltage of 0.880 V and a short-circuit current density of 270 A/m² (with a negligible thin CdS) (Sites and Liu, 1985).

Several methods have been used to deposit CdTe, namely vacuum evaporation, (Romeo et al., 2000b) electrodeposition (Johnson, 2000) and closed space sublimation (CSS). All these techniques are able to produce cells with efficiency larger than 10%. However, the highest efficiency solar cells have been prepared by closed space sublimation (CSS) (Ferekides et al., 1993; Wu et al., 2001; Romeo et al., 2000). This last technique has been also used in our laboratory but with a substantial modification, which consists in the use of a compact block of CdTe, previously melted and resolidified, instead of CdTe granules. The use of a compact block of CdTe is effective in getting a uniform heating of the material thus avoiding any spitting of microparticles from the source to the substrate.

The higher efficiency obtained by using CSS seems to be due to the fact that a high substrate temperature, namely 500–600 °C, is used. At these temperatures part of the CdS mixes with CdTe forming a mixed CdS_xTe_{1-x} (with $x \approx 0.07$), since only 7% of CdS can enter in the matrix of CdTe (Lane et al., 2000). If CdTe is deposited on top of CdS at low substrate temperature, as in the case of vacuum evaporation or electrodeposition, than the mixing process only happens when CdTe is treated with CdCl₂ at 400 °C substrate temperature. The CdS–CdTe mixing seems to be very important in order to get solar cells with efficiencies larger than 10%.

6. The CdCl₂ treatment

This treatment is necessary independent of the technique used to deposit CdTe. If the CdCl₂ treatment is not performed, the short circuit current of the solar cell is very low and also the efficiency is very low. The treatment consists in depositing 300–400 nm of CdCl₂ on top of CdTe with a subsequent annealing at 400 °C for 15–20 min in air or in an inert gas like Ar. During this process the small CdTe grains are put in vapour phase and recrystallize giving a better-organized CdTe matrix following possibly the reaction shown in Eq. (1):



The presence of Cl_2 could favour the crystalline growth of CdTe by means of a local vapour phase transport. In this way the small grains disappear and the CdS/CdTe interface is reorganized. If this process is carried out in air, some oxides are formed on the CdTe surface. These oxides need to be removed before making the back contact and an acid etching or a cleaning in a solution of Br-methanol is necessary (Romeo et al., 2000b). In an in-line production any use of acids or liquid solutions should be avoided in order to not interrupt the process and also for safety considerations.

For this reason we developed a new method for CdCl_2 treatment that consists simply in exposing the CdTe surface to a CdCl_2 vapour in an inert atmosphere such as Ar.

The process works like a closed space sublimation (CSS) in which the source is CdCl_2 and the substrate is CdTe. In order to remove any residual of CdCl_2 which could be present on the CdTe surface, after the treatment, the CdCl_2 source temperature is lowered and a high vacuum is maintained in the chamber keeping the substrate at 400 °C. At this temperature in vacuum, the residual CdCl_2 re-evaporates from the CdTe surface leaving a surface that is ready for the subsequent back-contact deposition. In this way no etching or cleaning process of the CdTe surface is necessary.

7. The back contact

Most researchers make a back contact on CdTe with a material containing Cu such as Cu–Au alloy, Cu_2Te , ZnTe:Cu (Gessert et al., in press) and Cu_2S (Kim et al., in press). It is believed that Cu is necessary to make an ohmic contact on CdTe since CdS/CdTe solar cells made with contacts not containing Cu have a high series resistance.

In our opinion the series resistance does not come from the contact but from CdTe which possibly is more conducting at the interface than in the bulk. The higher conductivity, close to the interface could come from the fact that CdTe mixes with CdS and its gap lowers down. In any case, in useful cells Cu must not be used since in the long run it segregates at grain boundaries and the solar cell degrades. It must be considered that the highest efficiency solar cell so far reported has been made with some copper at the back contact. Copper, by diffusing into CdTe lowers its resistivity and for a while it gives a higher efficiency.

We developed a new contact, namely Sb_2Te_3 , which has a low gap (≈ 0.3 eV), is p-type and exhibits a very low resistivity ($2 \times 10^{-2} \Omega\text{m}$) when it is deposited at a substrate temperature of 300 °C. Sb_2Te_3

can be deposited easily by sputtering, but it can also be deposited by vacuum evaporation (Romeo et al., 2000a).

We verified that Sb_2Te_3 makes an ohmic contact with CdTe thin films. We also found out that Sb_2Te_3 makes an ohmic contact on a p-type CdTe single crystal whose resistivity is on the order of $10^7 \Omega\text{m}$. With Sb_2Te_3 as a back contact we obtained CdS/CdTe thin film solar cells with a maximum efficiency of 15.8%. These cells are heated up to 200 °C under 20 suns without degrading. Instead, they generally exhibit a higher efficiency after the 200 °C treatment.

8. Recent results

Recently, a further improvement in the efficiency of CdTe/CdS solar cell has been obtained in our laboratory. We found out that, by depositing CdTe on top of CdS by CSS in presence of O_2 , the fill factor of the CdTe/CdS solar cell increases from around 0.65–0.66 to 0.71–0.74. In this way we were able to obtain on a 10^{-4} m^2 solar cell, prepared on soda lime glass, an efficiency of 15.8% with an open circuit voltage of 0.862 V, a short circuit current density of 255 A/m^2 and a fill factor of 0.72. On average, the efficiency of our cells made in similar conditions is in the range of 15.2–15.8%. This efficiency has been measured with an Oriel™ solar simulator under 10^3 W/m^2 AM 1.5 solar light. The measurement system has been calibrated with a 14% efficient CdTe/CdS thin film solar cell previously certified at the Renewable Energies Unit of the Joint Research Center, Ispra, Italy. An important remark is that no copper or any other element of group I of the periodic table of the elements has been used to make the back contact. Instead, we used 300 nm of Sb_2Te_3 followed by 300 nm of Mo deposited by sputtering. It is generally believed that copper is necessary to make a good contact on CdTe since it is generally observed that both fill factor and efficiency increase very much with a contact containing copper. We believe that Cu is not necessary to make a good contact and that the momentary improvement of the efficiency is due only to the doping of CdTe by copper since this last element is a strong diffusor. However, the cells made with some copper at the back contact are doomed to degrade since copper, at long run, segregates at the grain boundaries.

In order to obtain high efficiency solar cells with a high fill factor, as said before, one has to deposit CdTe by CSS in presence of O_2 . In our case, CdTe is deposited with a gas pressure of 10^4 Pa of Ar containing 2–10% of O_2 with a distance between crucible and substrate of about $4 \times 10^{-3} \text{ m}$. The substrate temperature is varied between 480 and 520 °C. CdTe is

covered by 100–500 nm of CdCl_2 and annealed in air or Ar at a temperature varying between 380 and 420 °C for 20 min. The efficiency of the solar cell depends on the amount of O_2 in the CSS chamber, on the substrate temperature during the deposition and the CdCl_2 annealing temperature. A high amount of O_2 or a high CdCl_2 annealing temperature tend to give a higher fill factor and a lower open circuit voltage.

The short circuit current is generally not affected but in the case in which a very large amount of O_2 and/or a high CdCl_2 annealing temperature is used. In extreme cases the short circuit current density could be much lower than 255 A/m^2 . A high substrate temperature, namely 520 °C, during the CdTe deposition tends to decrease the fill factor since, at this temperature, less O_2 is incorporated in the film. In order to interpret these results, we made the hypothesis that O_2 , mixing with CdS and CdTe, forms at the interface a mixed compound containing Cd, S, Te and O with a forbidden gap close to that of CdTe which is n-type. In this way a pseudo-homojunction is formed between this compound and p-type CdTe and this can explain the strong increase in the fill factor.

This hypothesis is corroborated by several facts:

1. The I – V characteristic in the dark exhibits an A factor close to 1 where A is given in the diode formula $J = J_0 \exp[(eV/AkT) - 1]$. An A factor close to 1 is typical of a homojunction.
2. When a high amount of O_2 , namely more than 10%, and/or a high CdCl_2 annealed temperature, namely more than 420 °C, are used, the short circuit current density is much lower than 250 A/m^2 despite a high open circuit voltage and principally a high fill factor (0.74–0.75) are obtained. In our vision, this means that a thick n-type layer larger than the absorption length is formed and a good part of the carriers created by the light do not reach the junction region. As a consequence, we deduce that the amount of O_2 used during the CdTe deposition is very important in order to control the formation and the thickness of this presumed n-type layer. The formation of an n-type thick layer has been confirmed by measurements of the photocurrent as a function of wavelength: when the solar cell exhibits a J_{sc} much lower than 250 A/m^2 the photocurrent response drops very quickly for wavelengths shorter than that corresponding to the gap of CdTe. The difference between the characteristics of two cells, one prepared using O_2 and the other prepared without O_2 is put in evidence in Fig. 3.
3. It has been reported (Hernández-Fenollosa, 2003) that O_2 in presence of Chlorine enhances the concen-

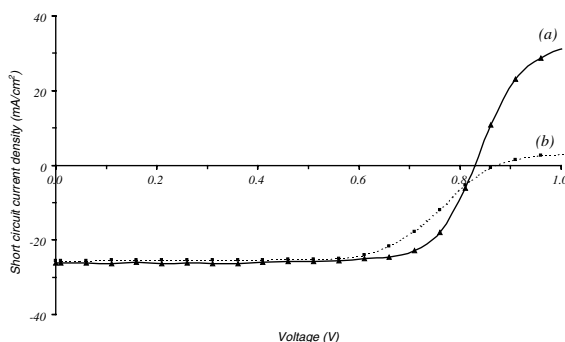


Fig. 3. Comparison between the J – V characteristics of two CdTe/CdS solar cells taken under 10^3 W/m^2 AM 1.5 simulated solar light: (a) J – V characteristic of a CdTe/CdS solar cell in which CdTe has been prepared by CSS in an atmosphere of Ar containing 2% of O_2 . Fill factor and efficiency are respectively: 0.73% and 15.45%. (b) J – V characteristic of a CdTe/CdS solar cell in which CdTe has been prepared by CSS in an atmosphere of pure Ar. Fill factor and efficiency are respectively: 0.63% and 13.67%. All the other layers that compose the cells, namely TCO, CdS and back contact have been prepared in the same way.

tration of $\text{Cl}_{\text{Te}}^{2-}$ which is a donor for CdTe and can form an n-type CdTe(S,O) layer at the interface.

9. A possible in-line dry process

The techniques, which we use to make CdTe/CdS solar cells, are CSS and sputtering both fast and easily scalable. A possible in-line process is shown in Fig. 4. Modules of $0.6 \times 1.2 \text{ m}^2$ can be covered with a process time of a few minutes.

The glass, moved on a rail, first enters in a washing machine and then is heated up to 400 °C and goes into a sputtering chamber where 500 nm of TCO are deposited. A laser scribing eliminates the TCO along parallel lines 10^{-2} m distant one from each other. The temperature is raised up to 500 °C and 80 nm of CdS and 4000–5000 nm of CdTe are deposited in sequence both by CSS.

A CdCl_2 treatment is done in another chamber at 400 °C substrate temperature, in an Ar atmosphere as described before. Then, a second laser scribing is done in order to remove CdS and CdTe in parallel lines close to the ones made before. Afterwards, the glass enters in a sputtering machine where 300 nm of Sb_2Te_3 are deposited at a substrate temperature of 300 °C. In sequence 300 nm of Mo are sputtered on top of Sb_2Te_3 in order to increase the conductivity of the back contact. Finally a third laser scribing is done in parallel lines close to those made by the second laser scribing in order to have a module made up of solar cells 10^{-2}

² Cl which substitutes Te.

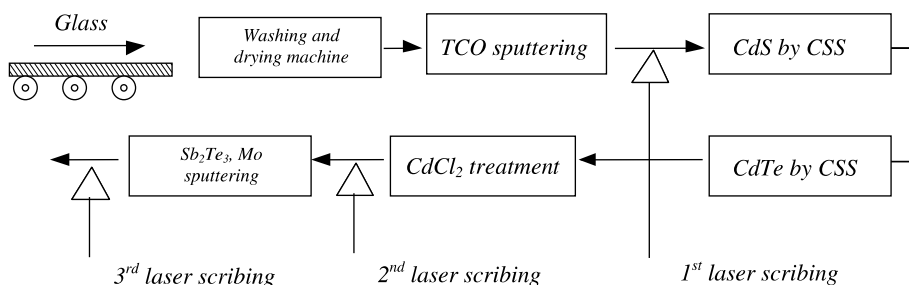


Fig. 4. Schematic of a possible in-line dry process for the production of CdTe/CdS solar cells modules.

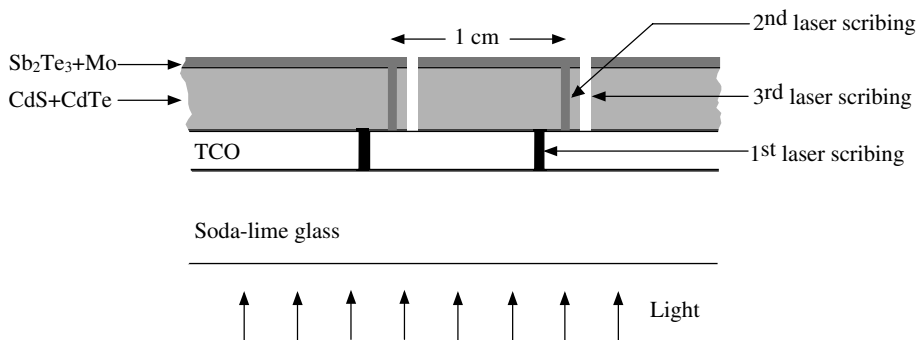


Fig. 5. Typical interconnect scheme for a CdTe/CdS solar cell module.

m large and as long as the length of the glass all put in series (Fig. 5).

10. Conclusion

The technology to fabricate CdTe/CdS thin film solar cells can be considered mature for the large-scale production of CdTe/CdS modules. The stable efficiency of single laboratory 10^{-4} m^2 cells is around 15.8% and it is expected that a $0.6 \times 1 \text{ m}^2$ module can exhibit an efficiency of 12% or more. Due to the simple and fast techniques that are used for the preparation of the cells, a production cost less than 1 Euro/Watt is foreseen.

The contribution of Parma University group to the development of such type of solar cells has been substantial. Three parts of the preparation process have been implemented:

1. A new stable TCO has been realized, namely fluorine doped In_2O_3 .
2. A dry process, which excludes any use of acid or liquid solutions, was shown to be effective in treating CdTe with CdCl_2 .
3. A new stable and ohmic back contact for CdTe, namely Sb_2Te_3 , has been invented.

A stable efficiency of 15.8% has been obtained by depositing CdTe by CSS in presence of O_2 .

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