

# An innovative process suitable to produce high-efficiency CdTe/CdS thin-film modules

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## ABSTRACT

A novel process, different in all steps from the common process usually used to fabricate CdTe/CdS thin-film solar cells, is described. This process on the one hand permits one to obtain high-quality layers and high-quality cells and on the other hand is particularly suitable to be scaled up at an industrial level. In fact, neither etching baths nor CdCl<sub>2</sub> deposition is used in this process. The back contact is made with a buffer layer capable of stabilizing Cu and, as a consequence, the devices are very stable.

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## 1. Introduction

CdTe with its energy gap of 1.45 eV is an ideal material to be used as an absorber in solar cells. Besides, its gap is “direct”, which means that only a few microns of the material are needed to absorb 90% of photons with energy greater than 1.45 eV. Its phase diagram is quite simple and only the stoichiometric compound can be formed at temperatures higher than 300 °C. At high temperature, namely 500–600 °C, that is used when it is deposited by closed-space sublimation (CSS), it grows p-type with a carrier concentration greater than 10<sup>14</sup> cm<sup>−3</sup> that are sufficient to form a good junction with an n-type partner. This is quite important since doping of polycrystalline thin films has to be avoided due to the fact that foreign elements, especially metals, tend generally to segregate into the grain boundaries. CdTe thin films can be prepared by several methods, such as simple evaporation [1], electrodeposition [2,3] and CSS [4]. Among these, CSS has been demonstrated to be most effective in giving high-efficiency solar cells since it allows the film to grow at high substrate temperatures (500–600 °C) at which CdTe grows polycrystalline with a large grain size (2–10 μm). High-efficiency thin-film solar cells are obtained using CdS as an n-partner (less than 100 nm thick) in the superstrate configuration. The highest efficiency so far reported for CdTe/CdS thin-film solar cells is 16.5% [5,6]. Here, the CdTe film is deposited by the CSS technique and CdS is deposited by chemical bath deposition (CBD), the front

contact is a layer of Cd<sub>2</sub>SnO<sub>4</sub> covered by a buffer of Zn<sub>2</sub>SnO<sub>4</sub> and the back contact is done with a layer containing a small amount of Cu. Due to the good photovoltaic properties of CdTe and the good scalability of the techniques used to produce CdTe/CdS thin-film solar cells, these devices are already produced in modules of 60 × 120 cm<sup>2</sup> by two companies, namely Antec Solar in Germany and First Solar in the United States. So far, CdTe/CdS modules exhibit efficiency between 7% and 10% [7]. An increase in the module efficiency can be obtained if the fabrication process is simplified in order to decrease the gap between the laboratory and the industrial scale. Here, we will describe a process that is notably simplified with respect to the ones that are currently used.

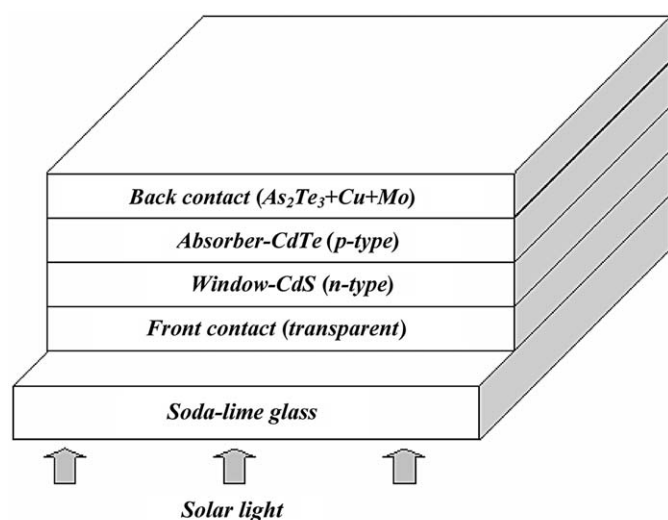
## 2. Structure of the cell

The CdTe/CdS thin-film solar cell is composed of four layers (Fig. 1):

- The TCO (transparent conducting oxide) layer that acts as the front contact.
- The CdS layer that is the n-type partner.
- The CdTe layer that is the p-type absorber.
- The back contact.

The configuration is of the “superstrate” type, which means that the light enters from the glass substrate. Both front and back contacts are composed of two sub-layers as it will be explained later on.

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**Fig. 1.** Sequence of the layers constituting the solar cell. The structure of CdTe/CdS solar cells has a superstrate arrangement.

### 3. The TCO layer

The most important characteristics that a TCO front contact must exhibit are a low sheet resistance and a high transparency in the visible region. Indium tin oxide with a 10 wt% of Sn fulfils these requirements. However, due to the high temperature at which CdTe is deposited and/or treated in the presence of chlorine, some In can diffuse from the ITO into the subsequent deposited layers. The use of a 100–200-nm-thick buffer layer between ITO and CdS, such as SnO<sub>2</sub> or ZnO, can hinder the In diffusion. Recently we have demonstrated, by SIMS depth profile measurement [8], that In has a concentration of  $5 \times 10^{15} \text{ cm}^{-3}$  in CdTe when SnO<sub>2</sub> is used as a buffer layer while its concentration rises to values of  $8 \times 10^{16}$ – $10^{17} \text{ cm}^{-3}$  when the buffer layer is not used. Besides, due to its relatively high resistivity, the buffer layer reduces the effect of a shunt resistance that could come from pinholes in the very thin CdS layer.

In prevision of an industrial production, the ITO is deposited by DC magnetron sputtering that is less expensive than RF magnetron sputtering while SnO<sub>2</sub> and ZnO are prepared by reactive DC magnetron sputtering in the presence of O<sub>2</sub> using pure Sn or Zn targets. In order to avoid the formation of nodules on the ITO target surface that could introduce a discharge instability, a rotating ITO target is being used.

### 4. The CdS layer

High-performance CdTe/CdS thin-film solar cells can be obtained with CdS prepared by three different techniques, namely chemical bath deposition, close-spaced sublimation and RF sputtering. We chose this last technique since it is the most suitable for industrial production. However, in order for a sputtered CdS film to give high-efficiency cells, it has to be made with some fluorine (F) in the sputtering chamber. We prepared F-doped CdS films by introducing in the sputtering chamber Ar containing  $\approx 3\%$  of CHF<sub>3</sub>. This gas is decomposed in the sputtering chamber and fluorine, being negative, is directed to the substrate. We expected that F will decrease the resistivity of the CdS film but this did not happen. On the other hand, F-doped CdS exhibits a forbidden gap a little larger than the un-doped CdS [9], a stronger cathodoluminescence emission [10], a stronger photoconductivity and, what is most important, gives higher

efficiency solar cells [11]. Possibly, fluorine, bombarding the CdS film during the growth, eliminates any excess of Cd and S and gives a more stoichiometric film. On the other hand, it could form some CdF<sub>2</sub>, which could passivate the grain boundaries. F-doped CdS has revealed to be more stable with respect to the CdS–CdTe mixing when the CdTe/CdS structure is treated at 400 °C in the presence of chlorine.

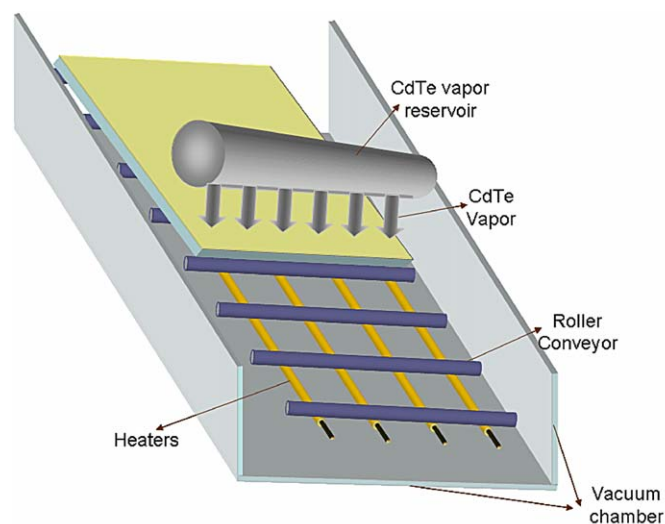
### 5. The CdTe layer

The most suitable method to deposit CdTe is close-spaced sublimation (Fig. 2), even though electrodeposition has also been demonstrated to be a very suitable technique [6]. However, CSS allows high deposition rates of the order of several microns per minute and it has been used for the preparation of the highest efficiency CdTe/CdS [5,11,12] solar cells. A slit 60 cm long and 2.5 cm wide was used and the vapour was supplied from top such that the glass substrate is well sustained on the carrying rollers. This is done in order to avoid the bending of the glass at 500 °C. The substrate temperature is typically 500 °C and the substrate–crucible distance is  $\approx 5$  mm. The deposition is done in an Ar atmosphere with a pressure of  $10^{-1}$ –1 mbar. With a crucible temperature of 700 °C, deposition rates between 1 and 4  $\mu\text{m}/\text{min}$  can be obtained.

### 6. Treatment of the CdTe/CdS structure in the presence of Cl<sub>2</sub>

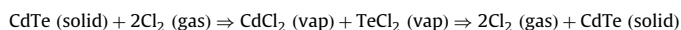
Treatment of CdTe in the presence of Cl<sub>2</sub> is necessary in order for the solar cells to exhibit high efficiency. This treatment is generally done by depositing CdCl<sub>2</sub> on top of CdTe by evaporation or by dipping CdTe on a solution of CdCl<sub>2</sub>–methanol and with a subsequent annealing at 400 °C in air or in an inert gas such as Ar [11]. In view of the industrial production we found out another method to carry out the Cl<sub>2</sub> treatment that does not use any CdCl<sub>2</sub>.

The CdTe/CdS structure is annealed in a mixture of 100–500 mbar of Ar and 20–50 mbar of a non-toxic gas containing Cl<sub>2</sub> such as difluorochloromethane (HCF<sub>2</sub>Cl). The annealing duration was  $\approx 5$ –10 min at 400 °C. After the treatment the CdTe morphology is completely changed with an increase in size (Fig. 3). Since HCF<sub>2</sub>Cl is decomposed at 400 °C and CdTe starts to decompose at around 400 °C, we suppose that the following reaction happens



**Fig. 2.** Sketch of CSS as it is foreseen for a production line.

especially for the small grains that are the first ones to decompose:



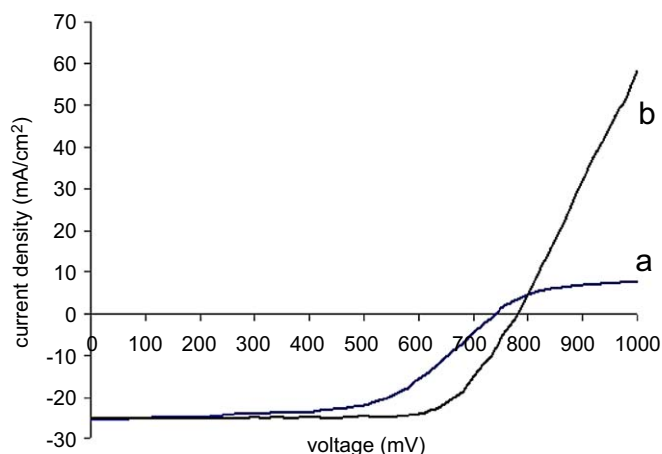
After the  $\text{Cl}_2$  treatment the sample was maintained in vacuum for few minutes at  $400^\circ\text{C}$  in order to permit any  $\text{CdCl}_2$  formed on the CdTe surface to re-evaporate and to have a clean CdTe surface ready for the back contact.

The  $\text{Cl}_2$  treatment can be carried out with any gas of the Freon family. The only requirement is that the gas contains Cl. This method is very effective in producing high-efficiency cells. It avoids the use of  $\text{CdCl}_2$  that could be dangerous and instead it uses a gas that is stable, inert and non-toxic at room temperature. Besides, it eliminates the step of  $\text{CdCl}_2$  evaporation and as a consequence it is much more suitable for an industrial production. This process has been patented [13].

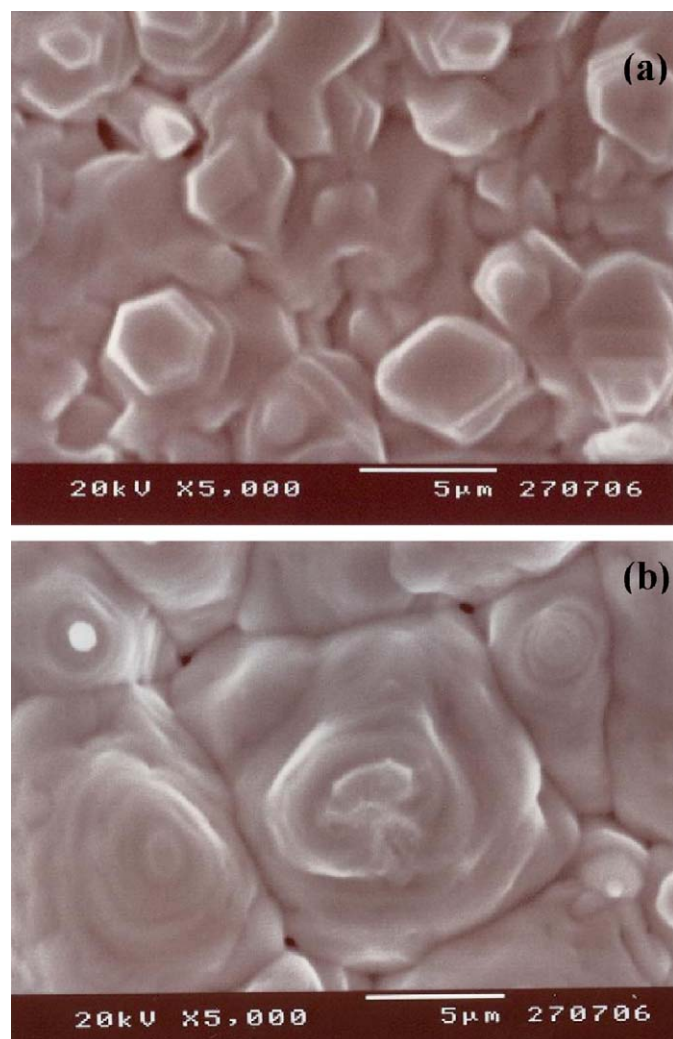
We are aware that a gas of the Freon family containing Cl is dangerous for the ozone belt. However, in an industrial production the Freon gas can be completely recovered and recovering machines are commercially available (RefTech International Inc., Bradenton, FL).

## 7. The back contact

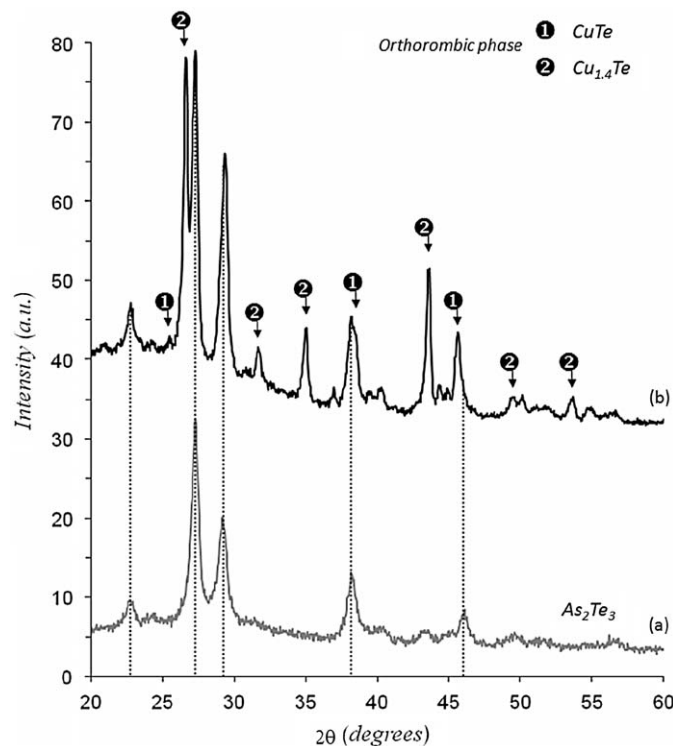
The back contact in the CdTe/CdS thin-film solar cell is a very important part of device fabrication. A non-ohmic contact causes a “roll-over” in the  $I$ - $V$  characteristic that lowers the fill factor and, as a consequence, the cell efficiency [14]. Since CdTe exhibits a high electronic affinity ( $\chi$ ), most metals form a Schottky barrier that limits the hole transport in the p-type CdTe. The most commonly used metal to make a non-rectifying contact with CdTe is copper. However, before Cu deposition, a chemical etch of CdTe is needed in order to create a Te-rich surface that, reacting with



**Fig. 4.**  $J$ - $V$  characteristic of a CdTe/CdS solar cell in which the back contact is deposited with different substrate temperatures: (a)  $\text{As}_2\text{Te}_3$ +Cu both deposited at room temperature; fill factor = 0.57 and (b)  $\text{As}_2\text{Te}_3$ +Cu both deposited at  $200^\circ\text{C}$ ; fill factor = 0.7.



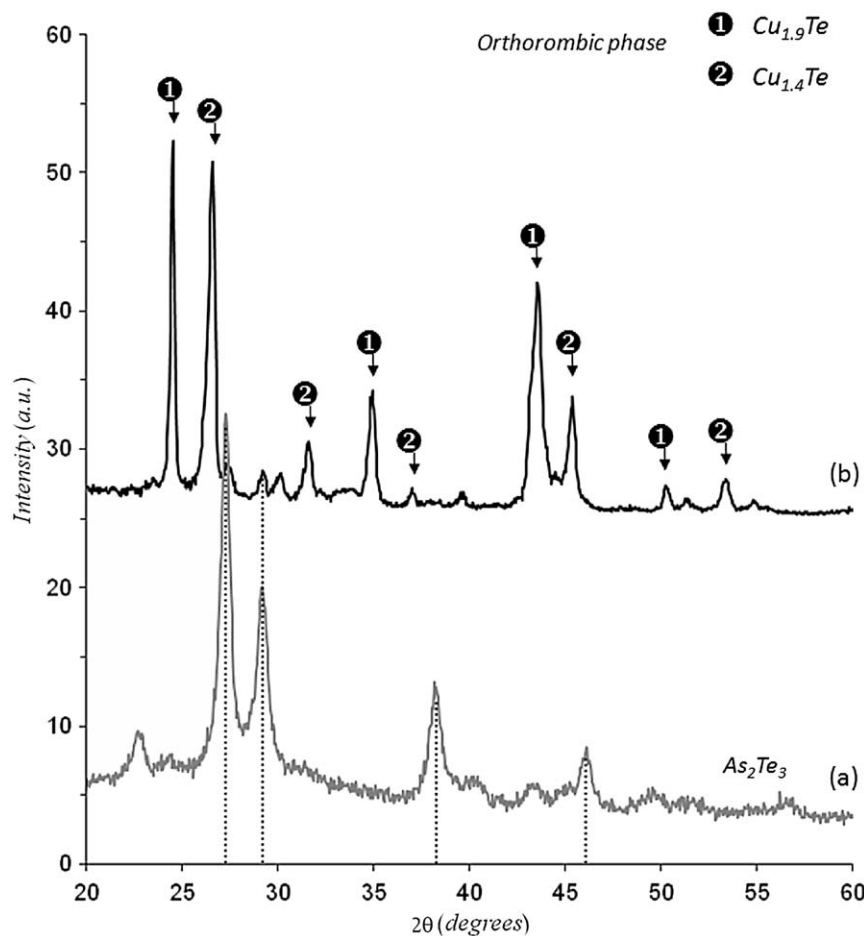
**Fig. 3.** SEM photos of the surface of CdTe films: (a) morphology of an untreated CdTe film deposited by the CSS method and (b) morphology of the same film after thermal treatment in  $\text{Ar}+\text{HCF}_2\text{Cl}$  atmosphere at a temperature of  $400^\circ\text{C}$  for 5 min.



**Fig. 5.** XRD analysis of (a) 200-nm-thick  $\text{As}_2\text{Te}_3$  deposited on glass at  $200^\circ\text{C}$  substrate temperature and (b) 200-nm-thick  $\text{As}_2\text{Te}_3$  film deposited on glass at  $200^\circ\text{C}$  substrate temperature on which 20-nm-thick Cu is deposited at the same temperature.

Cu, forms the compound  $\text{Cu}_x\text{Te}$  ( $1 \leq x \leq 2$ ) [15–18]. The ohmic contact is not done directly with Cu, which easily diffuses into the CdTe matrix, but with the  $\text{Cu}_x\text{Te}$  compound that is formed on the CdTe surface [19].  $\text{Cu}_x\text{Te}$  with  $x > 1.4$  is not a stable material [20] and, as a consequence, it frees copper that, being a fast diffuser, penetrates into the CdTe matrix causing two drawbacks: (1) it segregates into the grain boundaries forming shunting paths and (2) it arrives into the CdTe/CdS interface and forms a deep level in CdS that captures electrons, increasing the CdS resistivity. Since Cu is a positive ion, its diffusion inside the CdTe/CdS structure depends on the junction internal field that, in turn, depends on the fact that the cell is driven by an external bias or is illuminated by one or more suns. The device degradation is accelerated both if it is heated up at temperatures higher than 60 °C or if it is illuminated by several suns. When copper is used to make an ohmic contact, one has to be very careful in order to avoid any copper diffusion. For this reason, first solar uses only 2 nm of Cu onto a CdTe etched surface to make the back contact on its CdTe/CdS thin-film solar cells [21]. In the past, in order to avoid any degradation, we did not use any copper, but we used a low-gap p-type material, namely  $\text{Sb}_2\text{Te}_3$  [11]. This compound exhibits a very low resistivity, on the order of  $10^{-4} \Omega \text{cm}$ . If this material is deposited by sputtering at a substrate temperature around 300 °C, it can make a good contact with CdTe and can give solar cells with an efficiency close to 16%. This type of contact turned out to be very stable even when the device is illuminated by 10–20 suns and at a temperature higher than 100 °C. However, in some cases, depending on the starting  $\text{Sb}_2\text{Te}_3$  target, the presence of a roll-over in the  $I$ – $V$  characteristics has been found,

indicating that some rectification, even if not very much pronounced, is present in the back contact. Recently, we developed a novel contact that, despite containing Cu, maintains the stability of the device and does not present any roll-over in the  $I$ – $V$  characteristics. The novel contact is constituted by a layer of  $\text{As}_2\text{Te}_3$  deposited directly onto the CdTe surface without any etching of CdTe, followed by a thin layer of Cu deposited onto  $\text{As}_2\text{Te}_3$  at a substrate temperature of about 200 °C. Cu is then covered by 150 nm of Mo, which behaves as the external back contact.  $\text{As}_2\text{Te}_3$  is a p-type semiconductor that has a forbidden gap of about 0.6 eV and exhibits a room-temperature resistivity of  $10^{-3} \Omega \text{cm}$ . It melts at 360 °C and can evaporate at temperatures higher than 250 °C in vacuum. In order to get a good contact, one has to follow some simple rules: the  $\text{As}_2\text{Te}_3$  thickness can be varied between 100 and 300 nm while the Cu thickness can be varied between 2 and 20 nm.  $\text{As}_2\text{Te}_3$  can be deposited at a substrate temperature between RT and 200 °C, while Cu has to be deposited between 100 and 200 °C substrate temperature. In our experiments, both  $\text{As}_2\text{Te}_3$  and Cu are deposited by sputtering,  $\text{As}_2\text{Te}_3$  with a deposition rate between 10 and 20 Å/s and Cu with a deposition rate of 5 Å/s. If both  $\text{As}_2\text{Te}_3$  and Cu are deposited at room temperature, the contact is rectifying as one can see in Fig. 4, curve (a). Here the “roll-over” is clearly visible in the first quadrant of the  $I$ – $V$  characteristic. If Cu is deposited at a substrate temperature around 200 °C, the “roll-over” disappears and the fill factor is much higher (Fig. 4, curve (b)). In order to understand the behaviour of the double-layer  $\text{As}_2\text{Te}_3$ +Cu, some samples were prepared by depositing  $\text{As}_2\text{Te}_3$ +Cu directly on glass with Cu deposited at 200 °C substrate temperature. Besides, some samples



**Fig. 6.** XRD analysis of (a) 200-nm-thick  $\text{As}_2\text{Te}_3$  deposited on glass at 200 °C substrate temperature and (b) 200-nm-thick  $\text{As}_2\text{Te}_3$  deposited on glass at 200 °C substrate temperature on which 50-nm-thick Cu is deposited at the same substrate temperature.

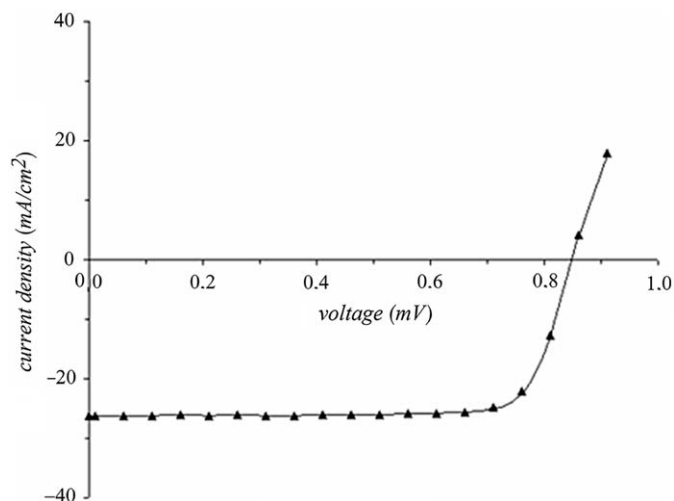
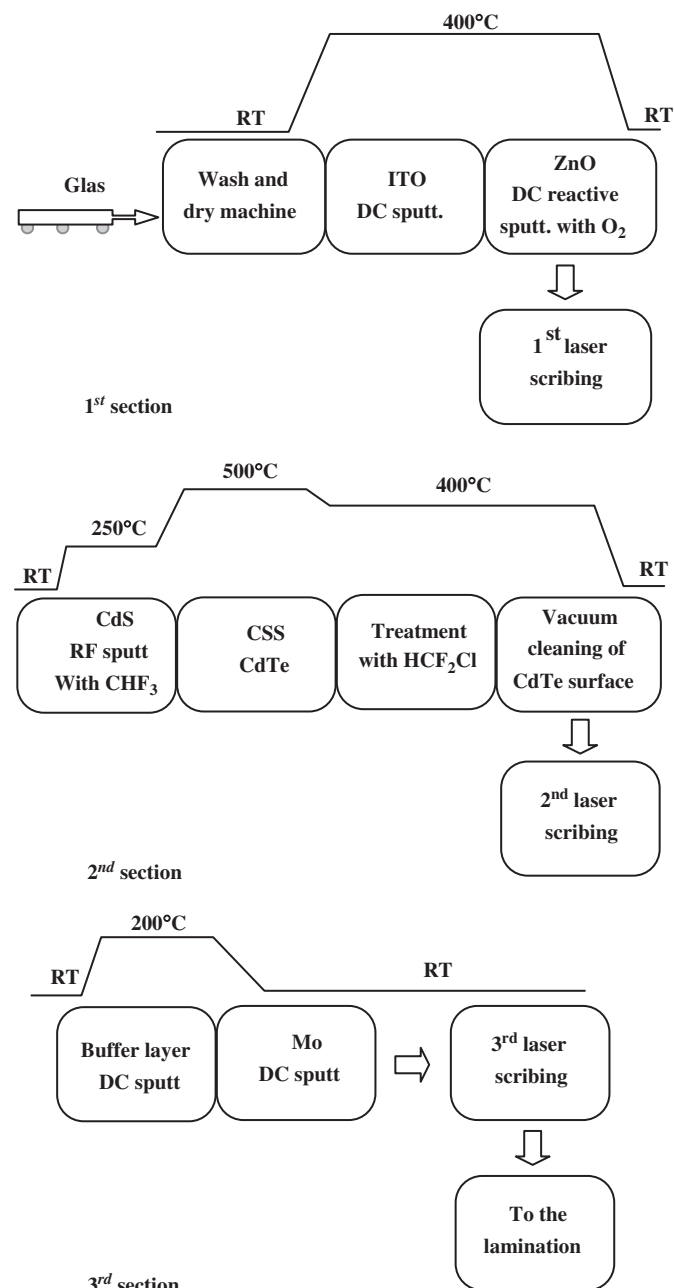


were prepared by depositing onto  $\text{As}_2\text{Te}_3$  a Cu layer up to 20 nm thick, while other samples were prepared by depositing a 50-nm-thick layer. In all cases the thickness of  $\text{As}_2\text{Te}_3$  was 200 nm. These samples were analyzed by X-rays and were compared with samples containing only  $\text{As}_2\text{Te}_3$ . As one can see in Figs. 5 and 6 curves (a) and (b), the samples containing Cu up to 20 nm exhibit several  $\text{Cu}_x\text{Te}$  phases with  $1 \leq x \leq 1.4$ , while the samples containing 50 nm of Cu exhibit also the  $\text{Cu}_{1.9}\text{Te}$  phase. Back contacts on CdTe/CdS thin-film solar cells made using 50 nm of Cu resulted to be not much stable, while cells made with 20 nm or less of Cu resulted to be very stable and more efficient. This is in accord with what is reported by Wu et al. [20], that is, the  $\text{Cu}_x\text{Te}$  phases with  $x > 1.4$  are not stable. A CdTe/CdS thin-film solar cell characteristic, in which the back contact has been done by depositing, in sequence, at a substrate temperature of 200 °C, 200 nm of  $\text{As}_2\text{Te}_3$  and 20 nm of Cu, is shown in Fig. 4, curve (b). No etching of CdTe has been done. Fill factor of this cell is  $\sim 0.7$ . The cell is stable even if a light soaking of 10 suns in the open-circuit mode is done at a temperature of 100 °C for 8 h. In order to further understand the  $\text{As}_2\text{Te}_3$ +Cu double-layer behaviour, some experiments were done by substituting  $\text{As}_2\text{Te}_3$  with  $\text{Sb}_2\text{Te}_3$ . In this case, the substrate temperature was varied between 200 and 350 °C for both  $\text{Sb}_2\text{Te}_3$  and Cu. In all the completed cells the back contact was not stable even though the Cu thickness was only 2 nm. The cell efficiency is quite high at the beginning, but decreases very fast. The results above reported can be explained in the following way. When Cu is deposited onto  $\text{As}_2\text{Te}_3$  at a sufficiently high temperature, namely 200 °C, Cu makes a solid-state reaction with  $\text{As}_2\text{Te}_3$  in which Cu substitutes As forming  $\text{Cu}_x\text{Te}$ . Depending on both  $\text{As}_2\text{Te}_3$  and Cu thickness,  $\text{Cu}_x\text{Te}$  phases with  $x \leq 1.4$  or  $x > 1.4$  can be formed. The stable phase with  $x \leq 1.4$  can be formed using an  $\text{As}_2\text{Te}_3$  thickness of 200 nm and a Cu thickness of 20 nm or less at a substrate temperature of around 200 °C. This type of reaction cannot happen with  $\text{Sb}_2\text{Te}_3$ , since  $\text{Sb}_2\text{Te}_3$  is a much more stable material than  $\text{As}_2\text{Te}_3$ . With  $\text{Sb}_2\text{Te}_3$ , Cu remains free and diffuses into the CdTe/CdS structure, damaging the cell. This novel method of making the back contact on CdTe is similar, in some aspects, to the one commonly used in which first a Te-rich surface is created by means of a chemical etching of CdTe and then a very thin layer of Cu is deposited in order to form the  $\text{Cu}_x\text{Te}$  with  $x \leq 1.4$ . However, a substantial difference lies in the fact that, in our case, no etching of the CdTe is done since we do not need a Te-rich surface (this is substituted by  $\text{As}_2\text{Te}_3$ ) and that a Cu thickness ten times higher can be used. This renders less critical the formation of the

non-rectifying contact giving a good stability to the CdTe/CdS thin-film solar cell. This novel contact has been patented [22].

## 8. The CdTe/CdS solar cell

One square centimeter CdTe/CdS solar cells have been prepared using the layers described before onto a 1 in<sup>2</sup> soda lime glass. These cells have been tested using an Oriel solar simulator (with a reference calibrated Si cell) under an AM1.5, 100 mW/cm<sup>2</sup> simulated solar light. Open-circuit voltages range between 840 and 870 mV, short circuit currents between 24 and 27 mA/cm<sup>2</sup> and fill factors between 0.6 and 0.72. The highest efficiency has been 15.8%. The *I*–*V* characteristic of this cell is shown in Fig. 7. Stability tests have been done leaving the cells for several hours under ten or more suns at a temperature higher than 100 °C and



**Fig. 7.** *J*–*V* characteristic of our best completely dry-processed cell under a 100 mW/cm<sup>2</sup>, AM1.5 simulated solar light.  $V_{OC} = 0.862$  V,  $J_{SC} = 25.5$  mA/cm<sup>2</sup>,  $ff = 0.72$ , Efficiency of 15.8%.

**Fig. 8.** The overall in-line process together with the temperature profile.

no degradation has been observed. In some cases, an increase in the open-circuit voltage has been obtained.

### 9. The in-line process

The in-line process together with the temperature profile is shown in Fig. 8. Glass with a size of  $0.6 \times 1.2 \text{ m}^2$  will be used. The process is divided in three sections. After each section a laser scribing is done in order to obtain 1-cm wide and 60-cm long solar cell stripes all put in series. The process is completely automated and a production of one module every 2 min is foreseen. In this way, with three shifts the line has a production capacity of 15–18 MW per year.

### 10. Conclusions

The process that we use to produce CdTe/CdS thin-film solar cells is quite simplified with respect to the one normally used in other laboratories; there are at least four innovations that simplify the process:

- (1) CdS is done by RF sputtering in an Ar+CHF<sub>3</sub> atmosphere. A better reproducibility and a better thickness control are obtained.
- (2) Treatment of CdTe is done using a gas that is inert and not toxic at room temperature. The step of CdCl<sub>2</sub> deposition has been removed.
- (3) CdTe is not etched before back contact deposition. The step of immersing CdTe in a chemical bath has been removed.
- (4) A novel back contact, namely As<sub>2</sub>Te<sub>3</sub>+Cu that is stable and not rectifying, has been developed.

Due to the simplification of the process, we believe that not much problems can rise in scaling it up to a production scale.

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