

CdTe/CdS SOLAR CELLS WITH TRANSPARENT CONTACTS

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Summary

Evaporated CdTe/CdS solar cells with a transparent Cu-indium tin oxide contact have been made with an efficiency greater than 8.5%. The deposition of single-phase CdTe films from a compound source required a cadmium-to-tellurium flux ratio of 1.7 incident on the substrate. To obtain the needed p-type conductivity of the CdTe films required a high temperature heat treatment in air which reduced the transmission through the CdTe film owing to the formation of a CdTeO₃ surface layer. The heating and cooling rates used for the heat treatment affected the open-circuit voltage and contact resistance of the cells. The total subband gap absorption of the entire cell is 40% - 50%.

1. Introduction

CdTe-based solar cells have the potential of being an economically viable source of electrical energy. Single-crystal devices with efficiencies greater than 13% [1] and thin film devices with efficiencies greater than 10% have been demonstrated [2]. With a band gap of 1.5 eV, the CdTe cell is also a leading candidate as the top cell in a tandem structure [3]. The major problems with thin film CdTe devices are controlling the p-type conductivity of the CdTe and making ohmic contact. Although CdTe cells have been fabricated by a variety of processing techniques, all methods require at least one processing step with a temperature in excess of 400 °C. This is detrimental to the fabrication of monolithic tandem cells.

In Table 1 a summary of the best reported CdTe cell results is presented for different processing techniques and cell structures. The best thin film device results have been achieved by depositing CdTe onto a wide band gap semiconductor to form a heterojunction device. This structure leaves an exposed CdTe surface on which to form the ohmic contact. A variety of etching-contacting procedures have been developed to form the contact to CdTe [14 - 15].

Previous efforts to develop a process for depositing a CdTe/CdS solar cell onto a CuInSe₂/CdS cell to form a monolithic tandem cell have been

TABLE 1

Summary of best reported CdTe cells

<i>Structure^a</i>	<i>Area (cm²)</i>	<i>V_{oc} (V)</i>	<i>J_{sc} (mA cm⁻²)</i>	<i>FF (%)</i>	<i>Efficiency (%)</i>	<i>Refer ence</i>
<i>Single crystal</i>						
CdTe/In ₂ O ₃	0.02	0.892	20.1	75	13.4	1
CdTe/ITO	—	0.81	23.5	55	10.5	4
CdTe(n)/CdTe(p)	—	0.83	19.7	67	10.5	5
<i>Thin film</i>						
CdS/CdTe (screen print)	0.78 ^b	0.754	27.9	61	12.8	6
CdS/CdTe (CSV ^c T) ^c	0.1	0.75	22.7	62	10.5	7
CdS/(CdHg)Te (electroplating)	1.48	0.62	27.03	63	10.6	8
CdS/CdTe (CSV ^c T) ^c	1.2	0.748	22.2	63.4	10.6	9
CdS/CdTe/ZnTe (spray-electroplate-evaporation)	4.0	0.745	22.0	63.6	10.4	10
SnO ₂ /CdTe (CSV ^c T) ^c	4	0.663	28.1	56	10.5	11
CdS/CdTe) (evaporated)	0.09 ^b	0.612	20.5	60.5	8.7	This work
CdTe/CdS (CVD) ^d	1	~0.67	~16.4	59	6.5	12
CdTe/ITO (CVD) ^d	1	~0.72	~19	60	8.2	12
CdTe/CdS	0.14	0.52	17.3	77	6.9	13

^aMaterials listed in sequence of deposition^bActive area^cClose-spaced vapor transport^dChemical vapor deposition.

reported [16]. In those devices, Cu-indium tin oxide (ITO) was employed as the transparent interconnect between the devices. Two problems were encountered; (1) reproducibly obtaining an ohmic contact between Cu-ITO and CdTe, and (2) controlling the p-type conductivity of the CdTe. Research was subsequently focused on the development and analysis of the contact to CdTe in a CdTe/CdS/ITO/glass configuration [17] and on controlling the p-type conductivity of CdTe thin films deposited by a vacuum-evaporation process compatible with monolithic tandem cells. In this paper, we discuss the results of this research effort and report on vacuum-evaporated CdTe/CdS cells with efficiencies greater than 8.5% using transparent contacts.

2. Experimental details

CdTe films were deposited by thermal evaporation from the compound using an effusion source bottle. The bell-jar deposition system typically operated at 1×10^{-5} Torr during film growth. The system was equipped with two additional source bottles for elemental dopants. The deposition conditions used to grow CdTe films nominally 2 μm thick were the following: CdTe incident flux, $10^{16} - 10^{17}$ $\text{mol cm}^{-2} \text{s}^{-1}$; substrate temperature, 150 - 350 $^{\circ}\text{C}$; film growth rate, 0.01 - 0.2 $\mu\text{m min}^{-1}$. The dopant incident flux was $10^{11} - 10^{16}$ $\text{atoms cm}^{-2} \text{s}^{-1}$ of antimony and $10^{12} - 10^{17}$ $\text{atoms cm}^{-2} \text{s}^{-1}$ of cadmium. The substrate consisted of nine 2.5 cm \times 2.5 cm samples which were radiatively heated. The substrate temperature was controlled by a Eurotherm temperature controller using a thermocouple embedded in one of the samples. The substrates were Corning 7059 glass (7059) and Corning 7059 glass coated with ITO or ITO/CdS. The ITO was deposited by magnetron sputtering and the films were 400 nm thick with a sheet resistance of 10 Ω/\square . CdS films were deposited by thermal evaporation and were 1 μm thick with resistivities of 1 - 10 $\Omega \text{ cm}$.

To make devices, the 7059/ITO/CdS/CdTe structure was heat-treated in air for 4 - 15 min to lower the resistivity of the CdTe (discussed below). Then the structure was etched in 4 M KOH at 60 $^{\circ}\text{C}$ for 30 s to remove surface oxides and to prepare the surface for contacting. A copper layer nominally 2 nm thick was deposited to form the ohmic contact followed by a 200 nm thick ITO layer to carry the current. Cell areas were delineated by a photolithographic etching process or by depositing the contact through an aperture mask. This process yielded a sample with 12 3 mm \times 3 mm cells.

CdTe films were structurally characterized using Cu K α radiation on a Philips X-ray diffractometer with diffracted beam monochromation. Total transmission and reflection measurements were made using a Perkin-Elmer 330 UV-visible-IR spectrophotometer equipped with an integrating sphere. The cross-grain resistivity was measured using an ITO substrate with a 100 μm wide gap. The carrier type was determined with a hot probe.

3. CdTe film properties

CdTe films deposited on bare 7059, ITO-coated 7059 and CdS/ITO/7059 were analyzed with regard to their structure, morphology and electrical properties. The effects of heat treatment on these properties were analyzed.

3.1. Structure and morphology

As-deposited films appear specular, although scanning electron microscopy revealed slightly faceted surfaces at $\times 10\,000$ magnification with little discernible difference between films grown on different substrates. As-deposited CdTe films were all highly oriented with the film plane lying parallel to the (111) plane. The degree of the preferred orientation varied

TABLE 2

Relative X-ray diffraction intensities for as-deposited CdTe films on different substrates

<i>Substrate</i>	<i>CdTe reflections (%)</i>				<i>Intensity of (111) (counts)</i>
	<i>(111)</i>	<i>(220)</i>	<i>(311)</i>	<i>(422)</i>	
Random	100	60	30	10	
7059	100	ND ^a	0.04	ND ^a	16500
ITO/7059	100	ND ^a	0.4	ND ^a	8100
CdS/ITO/7059	100	ND ^a	ND ^a	0.1	5000

^aND, not detected

consistently depending on the type of substrate used. As shown in Table 2, CdTe films deposited on bare 7059 had the highest degree of preferred orientation.

In addition, a tellurium second phase was identified in the X-ray diffraction scans of the CdTe films. The strongest reflections observed were the (100) and (003) peaks of hexagonal tellurium. The intensity of the tellurium peaks could not be correlated with the substrate temperature, the CdTe incident flux at the substrate or the type of substrate, as shown in Table 3

TABLE 3

Tellurium (100) peak intensity for different substrates and substrate temperatures for growth rates in the range 1 - 3 nm

<i>Substrate temperature (°C)</i>	<i>Tellurium (100) intensity</i>		
	<i>7059</i>	<i>ITO/7059</i>	<i>CdS/ITO/7059</i>
160	31	—	—
250	24	24	26
280	31	18	13
300	24	—	0

Using energy-dispersive spectroscopy and Auger techniques the level of excess tellurium in the films could not be quantified. On the basis of these results and the X-ray analysis, the tellurium second phase is estimated to be less than 0.1% of the film.

The tellurium second phase was eliminated by co-evaporating cadmium during film growth. A cadmium- to -tellurium incident flux ratio greater than 1.7 was needed to deposit single-phase CdTe films. The tellurium second phase can also be eliminated by heat treatment above 500 °C. Figure 1 shows representative diffraction scans in the region of the CdTe (111) for a single-phase CdTe film and for a film having a tellurium phase.

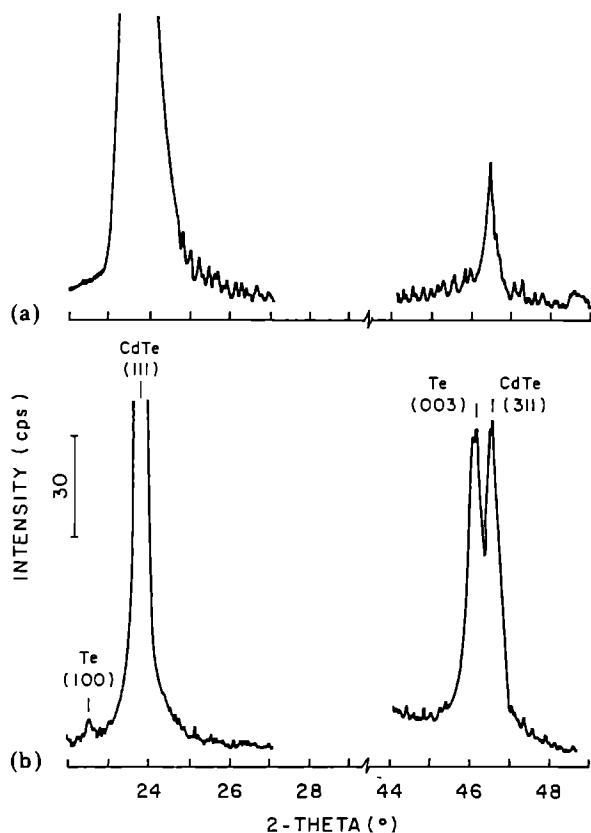


Fig 1. 2θ diffraction scans for CdTe films (a) single phase, (b) with tellurium second phase

3.2. Electrical properties

As-deposited CdTe films are highly resistive with a resistivity greater than $10^7 \Omega \text{ cm}$. Films deposited with an excess cadmium flux had resistivities an order of magnitude lower. In general, heat treatment of CdTe films lowers the CdTe resistivity. The effects of temperature, time and atmosphere are discussed below.

The effects of temperature and time on the resistivity using heat treatment in air are tabulated in Tables 4 and 5. The heat treatment at 550°C in air reduced resistivities by three orders of magnitude with the full effect occurring after 4 min. An additional heat treatment time does not lower the resistivity further but does adversely affect optical transmission through the CdTe film. At temperatures of 600°C and above, heat treatment in air results in reaction-evaporation of the CdTe film making electrical measurements impossible. Below 400°C , no change occurs in resistivity in the 15 min time interval. The effects of different atmospheres on resistivity are shown in Table 6. Although heat treatment in all the atmospheres reduced

TABLE 4

CdTe cross-grain resistivity as a function of temperature for heat treatment in air for 15 min

Sample condition	Temperature (°C)	Cross-grain resistivity (Ω cm)	
		Dark	Light
As-deposited	—	3×10^8	1×10^6
Heat treated	300	3×10^8	1×10^6
Heat treated	400	1×10^7	2×10^4
Heat treated	500	7×10^7	1×10^4
Heat treated	550	3×10^6	4×10^3

TABLE 5

CdTe cross-grain resistivity as a function of heat treatment time at 550 °C

Time (min)	Cross-grain resistivity (Ω cm)	
	Dark	Light
—	3×10^8	2×10^6
2	3×10^8	1×10^4
4	2×10^7	7×10^3
8	1×10^6	5×10^3
15	6×10^6	8×10^3

TABLE 6

CdTe cross-grain resistivity as a function of the atmosphere for heat treatment at 550 °C

Sample condition	Atmosphere	Cross-grain resistivity (Ω cm)	
		Dark	Light
As-deposited	—	1×10^8	2×10^6
Heat treated	Air	5×10^6	5×10^3
Heat treated	Argon	2×10^7	9×10^3
Heat treated	10vol %H ₂ –90vol %Ar	1×10^7	2×10^3
Heat treated	Hydrogen	4×10^5	2×10^4

the dark and light resistivity, only the treatment in air resulted in an improved cell performance.

Hot-probe (Seebeck) measurements of CdTe films on glass suggest that as-deposited films are intrinsic, but after heat treatment at 550 °C the films become p type. Heat treatment at 550 °C in air for 4 - 15 min was consequently used in the fabrication of the CdTe/CdS cells.

4. CdTe/CdS cells

The best results for an all-evaporated CdTe/CdS cell with a transparent Cu-ITO contact are given in Table 7. The short-circuit currents and fill factors are as high as those reported for CdTe/CdS cells with efficiencies greater than 10% [7, 8] but the voltages are 0.05 - 0.1 V lower. The low V_{oc} is attributed to interface recombination and high CdTe resistivity. These efficiencies are the highest reported for an evaporated CdTe/CdS polycrystalline thin film cell.

TABLE 7
ITO/Cu/CdTe/CdS cells

V_{oc} (V)	J_{sc}^a (mA cm ⁻²)	FF (%)	Efficiency (%)
0.612	20.5	60.5	8.7
0.663	19.8	57.3	8.6
0.523	19.4	68.6	8.0
0.552	13.6	71.6	6.1

^aELH simulation at 87.5 mW cm⁻²; cell temperature, 32 °C

4.1. Effects of heat treatment

Heat treatment in air at 550 °C for 4 min was required to produce cells with efficiencies greater than 6%. Further, it was found that the heating and cooling rate for the heat treatment was important for obtaining a high V_{oc} . Increasing the heating and cooling rates from 10² °C min⁻¹ (slow) to 10³ °C min⁻¹ (rapid) resulted in an increase in V_{oc} of 50 - 100 mV. Table 8 summarizes the J - V results for CdTe/CdS cells made from two CdTe depositions using the different heating and cooling rates. An increase in V_{oc} and small increases in FF and J_{sc} were obtained for all cells using the rapid heat treatment. The heating and cooling rates affect both the contact and bulk properties of the CdTe as discussed below. Similar behavior was observed for CSVT-deposited CdTe cells [18].

TABLE 8
Effect of cooling rate on V_{oc}

Heat treatment	V_{oc} (V)	J_{sc}^a (mA cm ⁻²)	FF (%)	η (%)
Slow cooling	0.491	15.8	47.9	4.3
Rapid cooling	0.605	18.0	57.0	7.1
Slow cooling	0.513	16.1	50.0	4.7
Rapid cooling	0.583	16.8	51.9	5.8

^aELH simulation at 87.5 mW cm⁻², cell temperature, 32 °C.

TABLE 9

Effect of heat treatment time at 550 °C in air on CdTe cell results

Heat treatment time at 550 °C in air (min)	V_{oc} (V)	J_{sc}^a (mA cm ⁻²)	FF (%)	η (%)
1	0.53	13.4	45	3.6
2	0.57	14.4	64	6.0
4	0.63	17.2	58	7.1
4	0.62	19.5	59	8.2
8	0.60	17.6	65	7.9

^aELH simulation at 87.5 mW cm⁻², cell temperature, 32 °C.

The effects on the cell performance of a heat treatment time from 1 to 8 min at 550 °C in air with rapid cooling are given in Table 9. As the heat treatment time was increased from 1 to 4 min, V_{oc} , J_{sc} and FF all increased, the increase being consistent with a decrease in the CdTe bulk resistivity (see Table 5). Again, no significant change was seen after a time longer than 4 min.

4.2. Cu-ITO contact

The Cu-ITO contact was analyzed and compared with gold contacts using a method which separates the light-dependent resistance R_L and the contact (light independent) resistance R_c components of the series resistance of a device [17]. A J - V analysis at different light intensities was done for three CdTe/CdS cells. Two of these had Cu-ITO contacts; one cell was heat treated and rapidly cooled and the other was heat treated and slowly cooled. The third cell was rapidly cooled and had a gold contact. The results are given in Table 10.

TABLE 10

Comparison of Cu-ITO with gold contacts for CdTe/CdS cells after different heat treatment cooling rates

	Au	Cu-ITO	Cu-ITO
Cooling rate	Rapid	Slow	Rapid
V_{oc} (V)	0.574	0.488	0.591
J_{sc}^a (mA cm ⁻²)	17.9	15.9	18.5
FF (%)	53.4	43.7	59.6
η (%)	6.3	3.9	8.4
R_c (Ω cm ²)	6	7	2
R_L (Ω cm ²)	1	1	0.5
J_0 (mA cm ⁻²)	6×10^{-5}	5×10^{-3}	3×10^{-5}

^aELH simulation at 87.5 mW cm⁻², cell temperature, 32 °C.

The contact resistance of the Cu-ITO is significantly lower than that of the gold when the sample is rapidly cooled. This is reflected in a higher FF. The light-dependent resistance, however, is essentially the same for all the samples suggesting that the bulk CdTe properties are not affected by the cooling rate or contact material. R_L does not represent the bulk photoconductivity in the CdTe film owing to non-uniform photogeneration in the CdTe film.

The heat treatment cooling rate affects both the contact and the junction. For the slowly cooled sample the reverse saturation current J_0 is 100 times larger than that for the two rapidly cooled samples. This is reflected in a low V_{oc} . Also, the contact resistance is larger for the slowly cooled sample than for the comparable rapidly cooled sample.

4.3. Cell transparency

The transmission T and reflection R of CdTe/CdS cells have been measured for the subband gap energy region, *i.e.* 850 - 1400 nm. The sum of R and T is shown in Fig. 2 after the five steps required to fabricate a CdTe/CdS cell. For the 7059/ITO (curve 1), the fall-off in R plus T at long wavelengths is due to free-carrier absorption in the ITO layer. After the CdS/CdTe deposition (curve 2), absorption in the entire structure increased by 8% - 15%. The decrease in R plus T at long wavelengths is attributed to changes in the ITO during the CdS and CdTe deposition. Curves 3 and 4 were measured after the 550 °C heat treatment and KOH etching respectively. The heat treatment increases the absorption by 15% - 20%. Curve 5 shows R plus T for a completed CdTe cell with a Cu-ITO contact. The total absorption in the structure varies from about 40% at 900 nm to about 50% at 1300 nm.

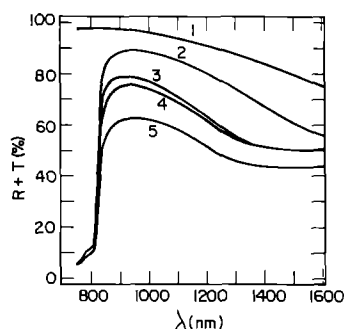


Fig. 2. The sum of the reflection and transmission of a CdTe/CdS cell after five cell fabrication steps.

To make these cells useful for tandem devices, absorption losses must be minimized. Lower heat treatment temperatures reduce absorption in the structure; however, the 550 °C heat treatment was needed to make efficient CdTe cells. Reducing the heat treatment time also reduces the absorption as shown in Fig. 3. However, conductivity measurements on CdTe films (Table 5) and cell results (Table 9) as a function of heat treatment time

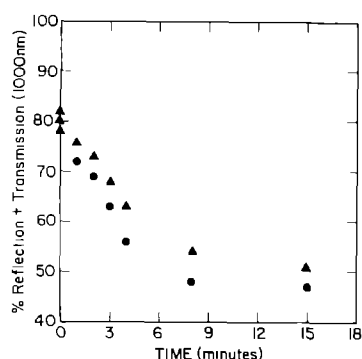


Fig 3 The sum of the reflection and transmission of a CdTe/CdS sample as a function of heat treatment time in air at 550 °C with rapid cooling ▲, heat-treated in air at 550 °C and quenched, ●, after etching with KOH.

indicate that heat treatment for at least 4 min is necessary. A comparison of the absorption after heat treatments with slow and rapid cooling showed little difference.

The 550 °C heat treatment was found to have two effects; the formation of a complex surface layer and the restructuring of the CdTe. The surface layer was mechanically removed from the CdTe surface and the absorption remeasured. No change in absorption was observed after removal of 100 - 200 nm of the film. After subsequent polishing, the absorption decreased until, after 600 - 1000 nm was removed, it was comparable with its value before the heat treatment. Further, optical microscopy of a tapered cross-section also revealed a two-layer surface structure with thicknesses consistent with those determined by polishing. The top surface layer is removed after etching in KOH.

These surface layers have been further studied using X-ray diffractometry and Auger depth profilometry. After the 550 °C heat treatment, X-ray diffraction scans revealed a significant number of reflections indexed as CdTeO₃, which are consistent with other reported results on heat-treated CdTe [19]. The intensity of the CdTeO₃ reflections increased with heat treatment time. While the KOH etching eliminated many of the CdTeO₃ reflections, some remained, indicating that the exposed surface of heat-treated and etched CdTe still contained CdTeO₃. Auger electron spectroscopy results confirm the presence after heat treatment of a layer containing nearly 50% oxygen. After etching a much lower level of oxygen is seen at the surface.

Finally, cells were made from heat-treated and mechanically polished 7059/ITO/CdS/CdTe. In these cells, four different etchants were used prior to the deposition of the Cu-ITO contact; KOH, 0.1% bromine in methanol and those used by Tyan [14] and Basol [15]. All the cells had a curvature in the forward-bias region of the *I-V* curve and therefore low fill factors, indicating non-ohmic contacts. Thus the surface containing CdTeO₃ formed by the heat treatment and KOH etching plays an important role in forming an

ohmic contact between the bulk CdTe and the Cu-ITO. The cells made from the polished samples gave values of J_{sc} and V_{oc} as high as 19.8 mA cm^{-2} and 0.603 V respectively. Therefore the bulk properties necessary to obtain high efficiency CdTe/CdS cells remain unaffected.

5. Discussion

Evaporated CdTe/CdS solar cells with a transparent Cu-ITO contact have been made with efficiencies greater than 8.5%. The subband gap transmission through the device is about 50% which is limited by a surface layer formed during a 550°C heat treatment. The effect of the heat treatment is to increase the p-type conductivity of the CdTe. Further, it was found that the heating and cooling rate of the heat treatment affected the open-circuit voltage and contact resistance of the cell. For these transparent CdTe/CdS cells to be useful as the top cell in a tandem structure, the transmission through the cell must be improved. This could be accomplished by reducing or eliminating the heat treatment required to control the p-type conductivity. If the processing temperature is low enough (less than 300°C), CdTe/CdS could be used in monolithic cell structures.

One approach to lowering the processing temperature is to use an extrinsic dopant to control the conductivity. Encouraging results have recently been reported on doping CdTe p type by a photo-assisted molecular beam epitaxy technique [20]. Epitaxial films with carrier concentrations of $5 \times 10^{16} \text{ cm}^{-3}$ and mobilities of $40 - 45 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ were achieved using antimony at a substrate temperature of 230°C . If similar techniques can be developed for thin polycrystalline CdTe films both monolithic and mechanically stacked tandem cells will be possible.

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