

Surface preparation effects on efficient indiumtin oxideCdTe and CdSCdTe heterojunction solar cells

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Surface preparation effects on efficient indium-tin-oxide–CdTe and CdS–CdTe heterojunction solar cells

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The effects of CdTe surface preparation and subsequent junction formation have been investigated through characterization of ITO/CdTe and CdS/CdTe heterojunction solar cells formed by electron beam evaporation of indium-tin-oxide (ITO) and CdS onto single crystal *p*-type CdTe. Surfaces investigated include air-cleaved (110) surfaces, bromine-in-methanol etched (110) and (111) surfaces, and the latter surfaces subjected to a hydrogen heat treatment. Both air-cleaved and hydrogen heat treated surfaces have a stoichiometric Cd to Te ratio. The ITO/CdTe junction formation process involves an air heat treatment, which has serious effects on the behavior of junctions formed on these surfaces. Etched surfaces, which have a large excess of Te, are less affected by the junction formation process and result in ITO/CdTe heterojunctions with solar efficiencies of 9% ($V_{oc} = 0.6$ V, $J_{sc} = 20$ mA/cm²). Use of low-doped CdTe results in junctions characterized by considerably larger open-circuit voltages ($V_{oc} = 0.81$ V) which are attributable to increasing diode factors caused by a shift from interfacial recombination to recombination in the depletion region. Resulting solar efficiencies reach 10.5% which is the highest value reported to date for a genuine CdTe heterojunction. CdS/CdTe heterojunctions show a strong dependence on CdTe surface condition, but less influence on the junction formation process. Solar efficiencies of 7.5% on an etched and heat treated surface are observed. All of these ITO/CdTe and CdS/CdTe heterojunctions have been stable for at least 10 months.

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INTRODUCTION

Because of its large absorption constant in the solar spectral range, CdTe is a prime candidate for all thin film solar cells. An all thin film cell of CdS/CdTe prepared by screen printing has been reported with an efficiency of 8.1%¹ and the same configuration deposited by close-spaced vapor transport (CSVST) has resulted in a 10.5% efficient solar cell.^{2,3} Cells using single crystal CdTe have been reported with solar efficiencies as high as 12%⁴ in a case in which the actual junction appears to be a buried homojunction. Actual heterojunctions have been reported with efficiencies of 8%–9%.^{5,6} Increasing experience with CdTe cells indicates that the junction preparation technique often controls the final cell properties: vacuum evaporation of CdS (Ref. 5) or spray-pyrolysis deposition of ZnO (Ref. 6) on CdTe result in heterojunctions, whereas sputtering of indium-tin-oxide (ITO)⁷ or chemical vapor deposition of doped CdS (Ref. 4) seem to produce buried homojunctions.

In general heterojunctions show much smaller open-circuit voltages than expected for an ideal junction because of leakage currents through interfacial defects. Such defects can arise during junction formation or be present on the as-prepared surface prior to junction formation. Recently we have investigated CdTe surfaces in detail using x-ray photoelectron spectroscopy,⁸ and also correlated the surface properties with the behavior of metal–CdTe junctions.^{9,10} It is

the purpose of the research reported here to investigate the properties of CdTe heterojunctions formed on various single crystal CdTe surfaces and to control the junction preparation so as to increase the open-circuit voltage. By using a vacuum evaporation technique to prepare these junctions it is found that the effects due to surface preparation and the effects due to junction formation can be separated.

SAMPLE PREPARATION

Substrates with dimensions 0.5×0.4 cm² were prepared from 0.1-cm-thick oriented wafers of single crystal *p*-type CdTe grown by a modified Bridgman method, doped with P and excess Te.¹¹ The typical bulk resistivity was about 2Ω cm; hole density and Hall mobility were 5×10^{16} cm⁻³ and 40 cm²/Vs, respectively. Orientations included (110), (111)_{Cd}, and (111)_{Te} (Cd-rich and Te-rich, respectively). A few substrates were obtained from highly doped CdTe (1×10^{17} cm⁻³), as well as from CdTe doped only with excess Te and having a hole density of 1×10^{15} cm⁻³. The standard preparation consisted of lapping the substrates and then polishing with 1- μ m alumina, followed by cleaning in organic solvents and etching in a 5% bromine-in-methanol solution (Br₂:MeOH) for a few minutes. A final etching step in Br₂:MeOH was always performed to achieve a shiny and residue-free surface prior to any further surface treatment or window material deposition. Polished and subsequently etched surfaces of various orientations provided the primary source for CdTe samples. A second method for preparing samples was cleaving in air (C). Cleaving in CdTe occurs

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along the (110) plane which is the only cleavage plane in the CdTe structure. In addition, some of the polished and etched (PE) samples were annealed in hydrogen (PEH).

The single crystal CdTe samples were subsequently mounted in a maskholder defining a 0.12 cm² area. Both ITO and CdS were deposited by means of electron beam (*e*-beam) evaporation. The system consists of a diffusion pump assisted evaporation chamber fitted with a Varian 6-kV, 4 crucible *e*-gun.¹² The ITO source material consisted of pressed powder pellets of 80 mole % In₂O₃ and 20 mole % SnO₂.¹³ ITO depositions were typically carried out in the presence of 5×10^{-5} Torr of dry oxygen. Samples and glass substrates were kept at room temperature, and the deposition rate was kept at about 5 Å/s. The CdS source material consisted of undoped, sintered grains,¹⁴ and the CdS evaporations were carried out at a chamber pressure of 2×10^{-6} Torr with the samples heated to 130–140 °C by quartz infrared lamps. The evaporation rate was about 10 Å/s to achieve a final thickness of about 1 μm. Evaporated In or Al made low-resistance contacts to the CdS films, whereas removable In discs were sufficient to make contact to the ITO films. To form low-resistance ohmic contacts to CdTe, necessary for electrical characterization as well as final solar cell characterization, the back side surface of the sample was etched with K₂Cr₂O₇·H₂SO₄·H₂O, which has been successfully used for low-resistance contacts to *p*-type CdTe and has been found to leave a Te-rich layer on the CdTe surface.^{8,15}

RESULTS

ITO films

As-deposited ITO films of 2000 Å on glass were almost opaque to visible light. TEM observations¹⁶ showed that the films deposited on CdTe were amorphous. The amorphous nature of the film was confirmed by a diffraction pattern. The mean diameter of the amorphous ring corresponds to the *d* spacing for the strongest reflection of the In₂O₃ structure. X-ray diffraction analysis confirmed the TEM measurements in that no crystalline diffraction peaks were obtained.

ITO films heat treated in air at 225 °C for 2 min showed crystalline structures and the *d* spacings obtained from the diffraction pattern can be interpreted as belonging to the In₂O₃ diffraction pattern. A separate x-ray diffraction analysis confirmed the TEM measurements. All observable diffraction peaks corresponded to the In₂O₃ diffraction pattern, implying that the Sn present does not assume a crystalline structure. The presence of Sn in the ITO films was confirmed by a microprobe analysis performed in conjunction with the TEM studies.

The values of film resistivity, electron mobility, and electron density are presented in Table I for both as-deposited and air heat treated ITO films of various thicknesses. The air heat treatment of the initially low conductivity, low mobility amorphous ITO film increases both the electron density and the mobility, leading to highly conductive films. Extensive heat treatments in air (> 24 h) were found to further increase the mobility but simultaneously to decrease the electron density, resulting in decreased conductivity.

TABLE I. Properties of selected ITO films.

Thickness (Å)	Resistivity (Ω cm)	Mobility (cm ² /Vs)	Carrier concentration (cm ⁻³)
2400 (as-deposited)	0.3	0.8	2.6×10^{19}
2400 (air heat treated)	1.9×10^{-3}	21.0	1.6×10^{20}
1600 (air heat treated)	2.5×10^{-3}	11.0	2.3×10^{20}

Transparency depended on both temperature and time; the choice of 225 °C for 2 min was made to minimize both temperature and time, still achieving transparency but reducing the air heat treatment for ITO-CdTe junctions.

The plots of the transmission characteristics obtained for three films of different thicknesses are given in Fig. 1. All films were deposited under the same conditions and subjected to a 2 min air heat treatment at 225 °C. The transmission rises to 90% at 4500 Å after which a plateau is reached. Interference peaks correspond to the different thicknesses.

CdS films

CdS films of 1–1.5 μm thickness deposited on glass were analyzed by x-ray diffraction. Most peaks could be assigned to the hexagonal structure with a strong preferred orientation close to the *c* axis. CdS films deposited on CdTe were found to depend on the CdTe surface orientation and preparation. Optical microscopy analysis indicated that on the etched (110) surfaces, CdS grew without visible features, whereas growths on etched as well as heat treated (111)_{Te} surfaces resulted in visible large area grains. Growth on the etched as well as heat treated (111)_{Cd} surfaces showed no visible features whereas CdS grown on a freshly cleaved surface showed features related to the cleavage steps induced during the cleaving process.

Recent observations of the CdS/CdTe interface using high resolution transmission electron microscopy (HRTEM) verify the dependence of CdS growths on surface orientation.¹⁷

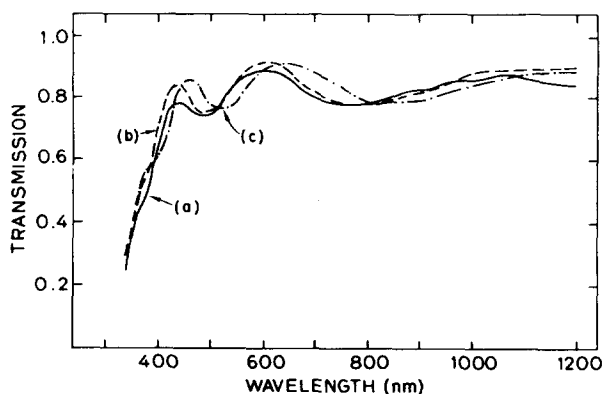


FIG. 1. Transmission plot of ITO films on glass: (a) 1600 Å, (b) 2400 Å, and (c) 2800 Å.

1- μm -thick CdS films deposited on glass at different temperatures showed different resistivities. For substrate temperatures above 180 °C, resistivities were of the order of $10^3 \Omega \text{ cm}$, at 150 °C of the order of $50 \Omega \text{ cm}$, and at 125 °C a resistivity of $10 \Omega \text{ cm}$ could be achieved. Lower temperatures resulted in resistivities around $1 \Omega \text{ cm}$ but at the low temperatures the films were increasingly more opaque. The typical deposition temperature was 135 °C at which temperature the resulting films on glass had an average transmission of 85% over the visible range.

The electrical properties of CdS films deposited on CdTe were found to depend strongly on the CdTe surface condition and orientation. On the etched (PE) (110) and (111)_{Cd} surfaces, the resistivity for a 1- μm film was $0.01 \Omega \text{ cm}$, whereas on the (PE) (111)_{Te} surface the resistivity was $0.5 \Omega \text{ cm}$. On the cleaved (C) surface, the resistivity was larger than $1 \Omega \text{ cm}$. Electron mobilities in CdS films on glass and CdTe surfaces were on the order of 6 and 30–60 cm^2/Vs , respectively.

ITO–CdTe junction characteristics

1. Effects of surface preparation

As-deposited ITO films on CdTe resulted in junctions whose usefulness as solar cells was prevented by the nontransparent ITO films. Measurements of the junction characteristics were performed to study the effect of air heat treatment necessary for transparency. The transition from amorphous to crystalline ITO was accompanied by drastic changes of the J - V characteristics for junctions formed on cleaved (C) surfaces with only minor changes being observed for etched (PE) surfaces; junctions on heat treated (PEH) surfaces show

similar results to those of junctions formed on cleaved surfaces. The results presented here are limited to the junctions formed on (C) and (PE) surfaces.

The dark J - V characteristics at 300 K for ITO/CdTe junctions are shown in Fig. 2. They can be fit over several decades by a standard diode equation

$$J = J_0 \exp(qV/AkT), \quad (1)$$

where J_0 is the dark saturation current and A is the diode ideality factor. The characteristics of the junction prepared on a (C) surface are drastically altered by the air heat treatment to achieve transparency. Initial J_0 values of about $1 \times 10^{-8} \text{ A/cm}^2$ increase to $1 \times 10^{-6} \text{ A/cm}^2$ and A factors change from about 2 to 1.5. Opaque ITO films on (C) surfaces result in junctions with the parameters $V_{oc} = 0.45 \text{ V}$ and $J_{sc} = 0.2 \text{ mA/cm}^2$. Transparent ITO films on (C) surfaces produced by heating in air result in junctions with V_{oc} of the order of 0.3 V and $J_{sc} = 18\text{--}20 \text{ mA/cm}^2$. The large dark saturation current caused by the air heat treatment is reflected in decreased V_{oc} values. Diffusion voltages as found from the voltage intercept from capacitance measurements were of the order of 1.0 V for junctions with opaque ITO and 0.7 V for junctions with transparent ITO on cleaved surfaces. The reduction in J_0 is thus attributable to lowering of the diffusion potential.

Etched (PE) surfaces resulted in ITO/CdTe junctions with the dark J - V characteristics also shown in Fig. 2. The characteristics of junctions with opaque and air-heated transparent ITO on (PE) surfaces are quite similar. The light J - V behavior of the junctions with transparent ITO is typically characterized by J_{sc} of 20 mA/cm^2 , V_{oc} of 0.6 V, and fill factors of 0.65 as shown in Fig. 3 for a junction prepared on an etched (110) surface. Resulting solar efficiencies of about 9% at 85 mW/cm^2 are typically observed. The effect of CdTe surface orientation was investigated with the results

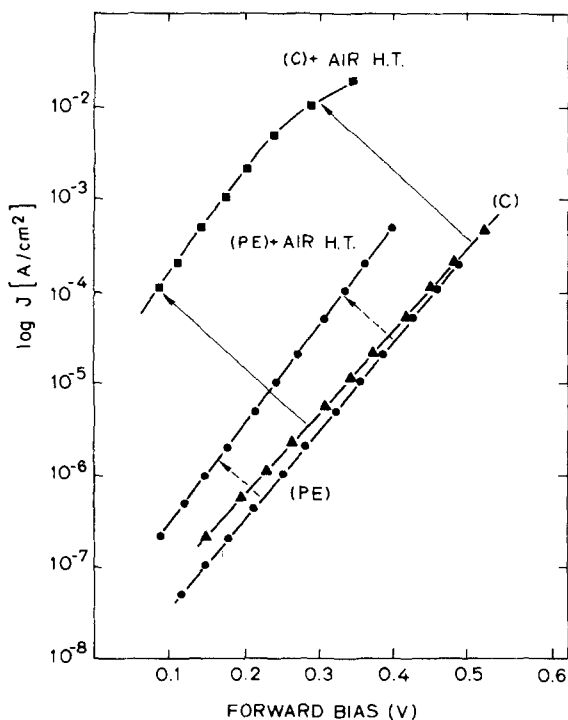


FIG. 2. Dark J - V characteristics of ITO/CdTe junctions on cleaved (C) and etched (PE) surfaces prior to and after air heat treatment.

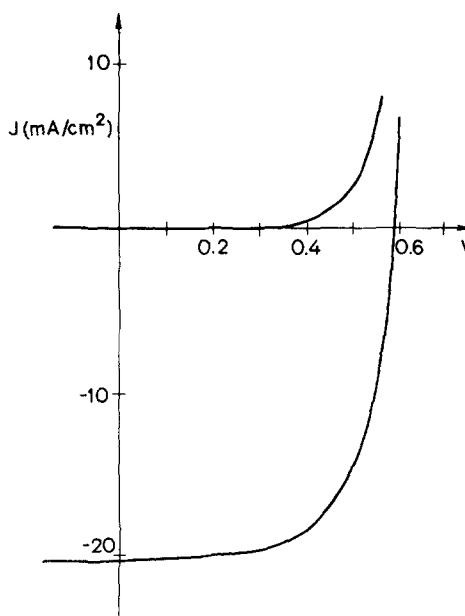


FIG. 3. Light and dark J - V characteristics of a typical ITO/CdTe heterojunction formed on an etched (PE) surface.

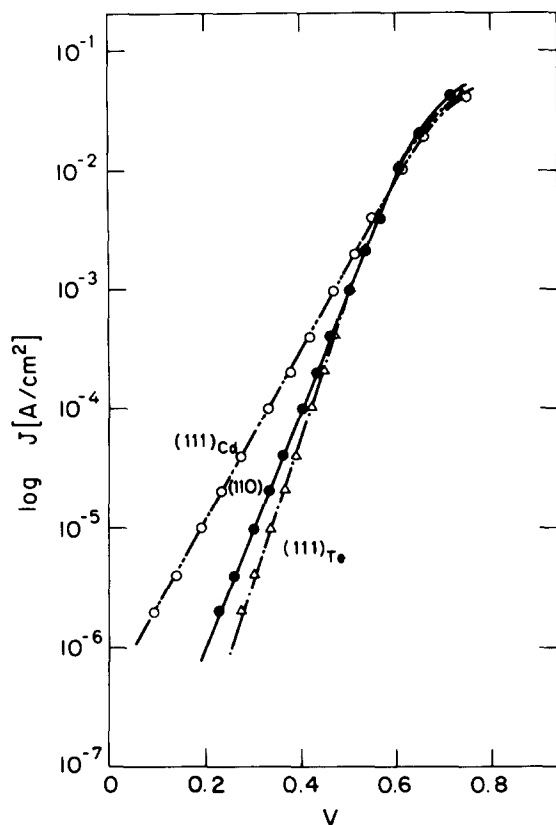


FIG. 4. Dark J - V characteristics of ITO/CdTe junctions formed on differently oriented CdTe surfaces prior to air heat treatment.

for opaque ITO on differently oriented surfaces of CdTe shown in Fig. 4. With opaque ITO, the relationship between the $(111)_{\text{Cd}}$ and the $(111)_{\text{Te}}$ surfaces is similar to that observed for Cr/CdTe junctions as reported elsewhere.¹⁰ For transparent ITO, the orientation effect was less pronounced. The air heat treatment results in junctions with J - V characteristics dominated by the effects introduced during the air heat treatment. The parameters for several cells are summarized in Table II.

The temperature dependence of the dark J - V characteristics was investigated for a typical junction formed on an etched (110) surface. The diode factor of 1.3 showed a temperature independent behavior above 293 K, suggesting a

TABLE II. Solar cell parameters of ITO/CdTe heterojunctions.

Surface orientation	CdTe hole density (cm^{-3})	V_{oc} (V)	J_{sc} (mA/cm^2)	ff	Efficiency ^a (%)
(110)	6×10^{16}	0.56	20	0.65	8.6
$(111)_{\text{Cd}}$	6×10^{16}	0.57	20	0.64	8.6
$(111)_{\text{Te}}$	6×10^{16}	0.59	20	0.63	8.7
$(111)_{\text{Cd}}$	1×10^{16}	0.65	20	0.63	9.6
$(111)_{\text{Te}}$	1×10^{16}	0.65	20	0.61	9.3
(110)	1×10^{15}	0.81	20	0.55	10.5
(110)	1×10^{15}	0.78	19	0.59	10.3

^a For simulated $85 \text{ mW}/\text{cm}^2$ solar illumination.

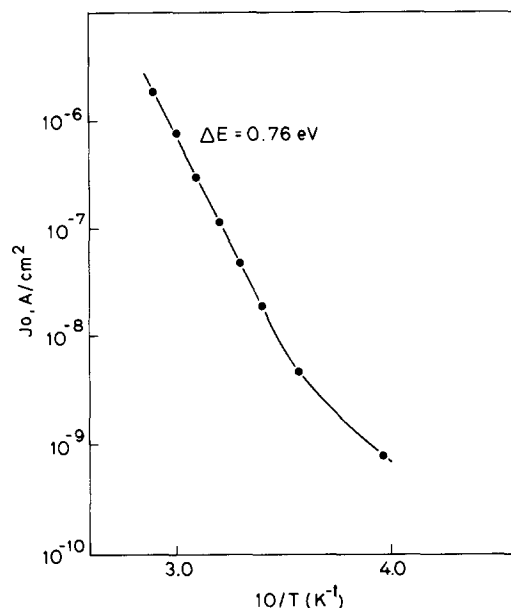


FIG. 5. J_0 as a function of $1/T$ of a typical ITO/CdTe heterojunction formed on an etched (PE) surface.

thermally activated recombination process with J_0 governed by

$$J_0 = J_{00} \exp(-\Delta E/kT). \quad (2)$$

By plotting J_0 as a function of $1/T$, shown in Fig. 5, an activation energy ΔE of 0.76 eV is obtained. Reasonable agreement between the product $A \cdot \Delta E$ of 0.99 eV and the expected diffusion voltage of $qV_D = 1.06 \text{ eV}$ suggests that the recombination mechanism is governed by the interface as proposed by Van Opdorp.¹⁸ The relative independence of J_0 with temperature below 293 K suggests a tunneling mechanism.

The spectral response of this junction, presented in Fig. 6, is characterized by a sharp increase in quantum efficiency at the band gap of ITO, followed by a plateau at a quantum efficiency of about 0.9. This spectral response is indicative of an abrupt junction located close to the surface typical of a genuine heterojunction. Interference effects from light transmission through the ITO film can be observed. For wavelengths above 820 nm, a sharp drop in quantum efficiency is observed at the band gap of CdTe.

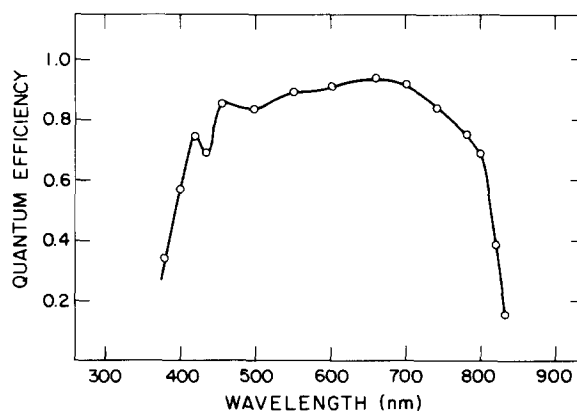


FIG. 6. Spectral response of the quantum efficiency of an ITO/CdTe heterojunction solar cell formed on an etched (PE) (110) surface.

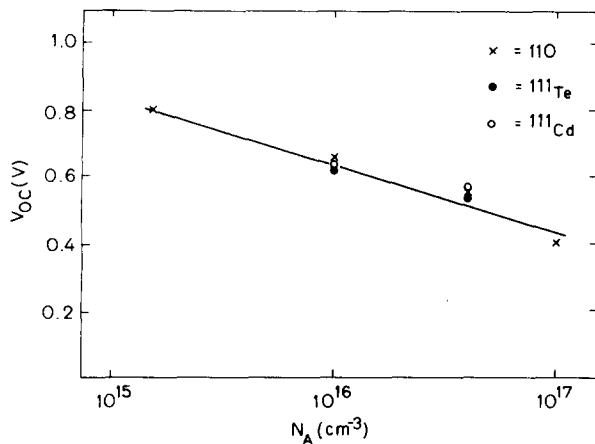


FIG. 7. V_{oc} as a function of CdTe hole density of ITO/CdTe junctions.

2. Effects of CdTe hole density

Figure 7 shows the dependence of V_{oc} values on the CdTe hole density. Highly doped CdTe results in ITO/CdTe junctions with low V_{oc} . The details of the light and dark J - V characteristics for three differently doped (110) CdTe based junctions are shown in Figs. 8 and 9, respectively. No simple variation of J_0 and A can account for increased V_{oc} values with decreased hole density. However, increased diode factors with decreased hole density suggest a shift from interfacial recombination to recombination in the depletion region. V_{oc} values as high as 0.81 V were observed under 85 mW/cm² of illumination for the junction prepared on CdTe with $p = 1 \times 10^{15} \text{ cm}^{-3}$. Combined with $J_{sc} = 19 \text{ mA/cm}^2$ and a fill factor of 0.55, the efficiency is 10.5%. The fill factor is reduced by series resistance partially arising from the bulk CdTe. These heterojunctions have been stable in a laboratory environment for over 11 months.¹⁹

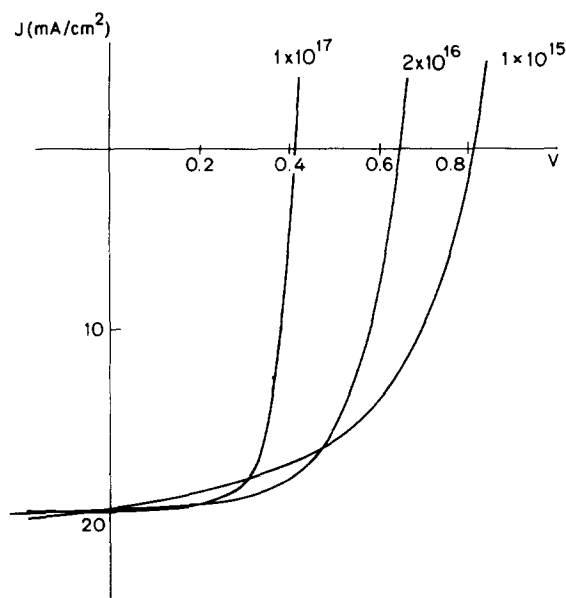


FIG. 8. Light J - V characteristics of ITO/CdTe junctions formed on CdTe of different hole densities.

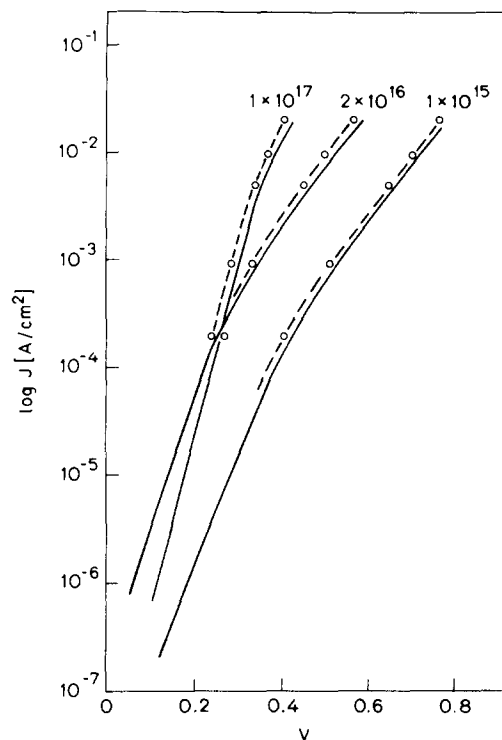


FIG. 9. Dark J - V characteristics of ITO/CdTe junctions formed on CdTe of different hole densities (solid line). Also, J_{sc} vs V_{oc} (dashed line).

CdS-CdTe Junction characteristics

As-deposited CdS/CdTe junctions showed J - V characteristics with a different dependence on CdTe surface preparation than ITO/CdTe junctions. Cleaved (C) and heat treated (PEH) surfaces result in junctions characterized by large V_{oc} whereas etched surfaces (PE) produce junctions with lower V_{oc} . This observation is in close agreement with the results obtained for Cr/CdTe junctions formed on the various surfaces.^{9,10} The excess Te produced by etching limits the attainable V_{oc} by resulting in large values of J_0 . Since

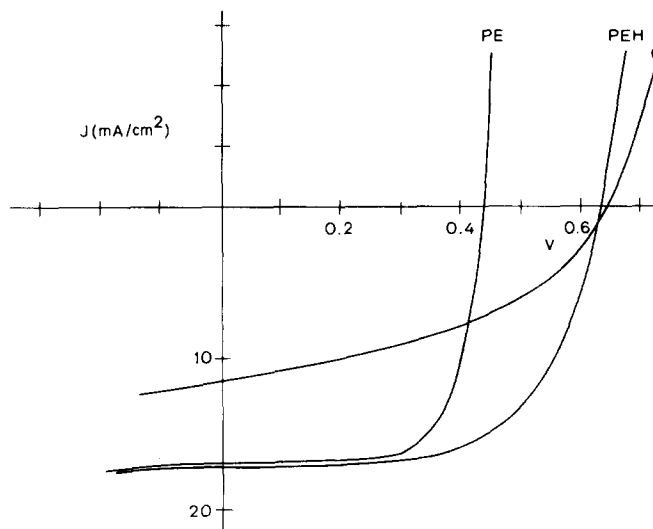


FIG. 10. Light J - V characteristics of CdS/CdTe junctions formed on cleaved (C), etched (PE), and heat treated (PEH) surfaces.

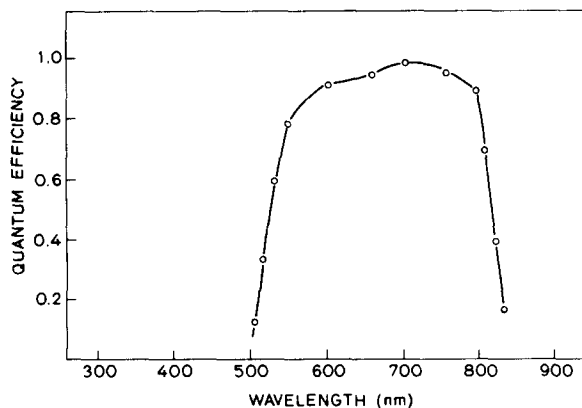


FIG. 11. Spectral response of a CdS/CdTe heterojunction.

CdS film properties depend on the CdTe surface condition, useful analysis was limited; also, although CdS films were highly transparent, resulting resistivities caused reduced fill factors and short circuit currents. Only a brief investigation was carried out for junctions formed on various oriented surfaces, and the study was limited to CdTe with a hole density of about $5 \times 10^{16} \text{ cm}^{-3}$.

The light J - V characteristics for CdS/CdTe junctions formed on the three surfaces are shown in Fig. 10. The (C) and (PEH) surfaces both result in V_{oc} of the order of 0.6 V, whereas the (PE) surface results in a junction with $V_{oc} = 0.45 \text{ V}$. The series resistance effect is especially significant for the junction on the cleaved surface leading to reduced photocurrent and fill factor. A typical spectral response curve is shown in Fig. 11 and is indicative of a genuine CdS/CdTe heterojunction. The band gap of CdS reduces the attainable short circuit currents as compared to ITO. Resulting solar efficiencies (85 mW/cm^2 simulated) reach 6.2% for the junction prepared on the (PE) surface and 7.5% for the junction formed on the (PEH) surface. These CdS/CdTe heterojunctions have been stable for 10 months in a laboratory environment.¹⁹

DISCUSSION

The characteristics of ITO/CdTe and CdS/CdTe heterojunctions have been found to be strongly influenced both by the surface preparation and by the junction formation process itself. They also depend on hole density and orientation of the specific CdTe substrates. ITO/CdTe junctions formed on stoichiometric, cleaved (C) as well as heat treated (PEH) surfaces show poor V_{oc} because of large J_0 values. An air heat treatment, which is necessary for ITO transparency, results in serious degradation of the diode characteristics for junctions formed on these stoichiometric surfaces. Only etched surfaces which are covered with excess Te result in junctions that remain relatively unaltered by the ITO-transparency process. The air heat treatment is accompanied by an amorphous-to-crystalline ITO transformation that is likely to induce defects on the surfaces. Excess Te on the etched surfaces apparently accommodates the amorphous-to-crystalline transformation more readily, especially if this Te layer is in an amorphous state, as suggested by some of the

TEM studies.¹⁶ Still, the presence of excess Te is likely to give rise to interfacial defects that ultimately govern the interface current transport mechanism. Analysis of the J - V characteristics of the ITO/CdTe junctions indicates that recombination at the interface dominates the J_0 values at temperatures above 293 K. Short circuit currents of the order of 20 mA/cm^2 at 85 mW/cm^2 are indicative of 90% quantum efficiency. Spectral response measurements confirmed the heterojunction nature of the junctions with large response at short wavelengths, indicative of an abrupt junction located close to the surface.

By using low-doped CdTe, V_{oc} was greatly improved. Such a behavior may not be expected in view of reduction in diffusion potential with decreased hole density. However, this improvement was caused by increased diode factors at constant J_0 . For low-doped CdTe, recombination in the depletion region becomes increasingly important whereas recombination at the interface is reduced. Increased diode factors resulted in V_{oc} as high as 0.81 V and a solar efficiency of 10.5%. Reduced fill factors caused partly by the series resistance from the bulk CdTe gives a practical limit to attainable efficiencies with the use of single crystal substrates of reasonable thickness. However, the use of low-doped CdTe to increase V_{oc} by reducing the interface recombination should be a viable approach for all-thin-film CdTe solar cells. Recent results on CSVT grown thin film CdTe solar cells have indeed been reported to have low hole densities (10^{14} cm^{-3}).³

CdS/CdTe junctions showed a dependence on CdTe surface conditions similar to that observed for metal/CdTe junctions.^{9,10} V_{oc} values were of the order of 0.65 V for junctions formed on cleaved (C) and heat treated (PEH) surfaces, whereas V_{oc} values were of the order of 0.45 V for junctions on etched (PE) surfaces. The resistivity of the as-deposited, undoped CdS films showed a strong dependence on the surface condition and orientation. On cleaved surfaces, resulting films were resistive and junctions therefore suffered from reduced fill factors and short circuit currents. No attempt was made to dope the CdS, but it is expected that the effects of series resistance can be eliminated through doping or by coating with a transparent conducting contact. The effects of the three different surfaces on the characteristics of the CdS/CdTe junctions further confirmed the presence of an etch-induced surface layer on the etched surface. Solar efficiencies reached 7.5% for a CdS/CdTe heterojunction prepared on a heat treated (PEH) surface. Previous research on CdS/CdTe has resulted in an 8% efficient heterojunction.⁵ A heat treatment in hydrogen above 400°C was required in the processing of this junction and may have been important in improving the efficiency from 6% as observed for junctions prepared on etched surfaces.

CONCLUSIONS

ITO/CdTe heterojunctions prepared on polished and etched (PE) surfaces exhibit solar efficiencies of about 10% at 85 mW/cm^2 of simulated sunlight. The major losses for these heterojunctions are attributable to reduced fill factors and V_{oc} because of the nonideal nature of the interface region. ITO/CdTe junctions prepared on cleaved (C) as well as

heat treated surfaces (PEH) show poor V_{oc} because of large saturation currents induced by a necessary air-heat treatment for ITO transparency. Etching apparently protects the surface against this effect, but limits final performance achievable.

Future improvements of the solar efficiency of ITO/CdTe heterojunctions call for improved fabrication techniques. Junction properties of as-deposited ITO (prior to air-heat treatment) on (C) and (PEH) surfaces indicate the potential for highly efficient heterojunctions. Even in their present state with efficiency of over 10%, ITO/CdTe heterojunctions prepared on etched (PE) surfaces represent the highest efficiency for any genuine CdTe-based heterojunction to date. These heterojunctions have been stable for over 11 months in a laboratory environment.

CdS/CdTe heterojunctions prepared on (PEH) surfaces showed efficiencies close to 7.5% whereas the (PE) surfaces resulted in 6% efficiency. Results from CdS/CdTe heterojunctions are consistent with the observations from metal/CdTe junctions; (PE) surfaces result in poor V_{oc} values whereas (C) and (PEH) surfaces result in larger V_{oc} values.

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