

Preparation and Properties of Evaporated CdTe Films Compared with Single Crystal CdTe

Annual Report
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A Subcontract Report

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Stanford University
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Prepared under Subcontract No. XE-2-02081-1



SERI

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ABSTRACT

Variation of CdS/CdTe/graphite thin film solar cell properties was investigated as a function of the temperature for CdS film deposition. A maximum open-circuit voltage of 0.67 V was found for a deposition temperature of 160°C, corresponding to a CdS film resistivity of 150 ohm-cm. The effect is not due to avoidance of higher temperature annealing of the CdTe film in higher temperature CdS film depositions, nor to the diffusion of In from the outermost CdS:In layer. EBIC scans indicate uniform collection over the entire CdS surface area for cells with CdS layers of 2 μm thickness.

The effect of coating the graphite before CdTe deposition with 20A of Au, or 5 to 50A of Cu, was investigated. Although high concentrations of both Au or Cu could be determined after CdTe deposition by microprobe analysis, CdTe films grown on Au- or Cu-coated graphite had lower hole densities than films grown on uncoated graphite. In addition, deposition on a graphite substrate coated with 20A of Au increased the growth rate for the CdTe film by a factor of four. Attempts to deposit CdTe with a partial pressure of Cu to provide Cu doping proved unsuccessful.

Photovoltaic parameters of thin film CdS/CdTe/graphite solar cells were investigated as a function of storage time to check the stability of these cells. Initial degradation of parameters, primarily the fill factor, could be reversed by a heat treatment in hydrogen, with subsequent properties being stable. A cell that was heat treated in hydrogen shortly after cell fabrication showed no degradation effects with time.

Heat treatment of CdS/CdTe/graphite solar cells in air increases the cell resistivity and decreases the fill factor, whereas heat treatment in hydrogen produces the reverse effect. The hole density is not affected by such heat treatments, suggesting that the observed effects are associated with the grain boundaries in the film. These results are consistent with

previously reported results on CdTe/graphite films. A solar cell with a hydrogen passivated CdTe film will be fabricated in the near future.

A computer model has been developed to investigate the bond length changes produced by an impurity atom occupying a substitutional site in the CdTe lattice. One of the applications of the model of particular interest to device applications is the interaction of impurities with native defects through a strain interaction.

CdS/CdTe THIN FILM SOLAR CELLS

Over the last year we have continued the investigation of material improvement and control for p-type CdTe films to be used in CdS/CdTe thin film solar cells. CdTe thin films have had hole densities in the range of 2 to $5 \times 10^{15} \text{ cm}^{-3}$. Empirical relations between growth rate and hole density (without any intentional doping source) have been established and are shown in Figure 1; this effect is under continuing investigation to determine whether the hole density is dependent on Cd or Te over-pressure during growth, as would be expected to be the case if native defects control the hole density.

CdS/CdTe solar cells prepared by the vacuum evaporation of CdS on p-CdTe films deposited by hot-wall vacuum evaporation (HWVE) on graphite substrates, have been shown to have an open-circuit voltage V_{oc} that depends on the CdS film resistivity, with a maximum value of 0.67 V for a CdS resistivity of about 150 ohm-cm , as measured on glass. Other solar cell parameters (e.g., J_{sc} , ff , and collection function) appear to be related to the CdS deposition temperature, but to a lesser extent.

Variation of CdS/CdTe Solar Cell Properties

The CdTe films used in this investigation were deposited at a substrate temperature of 583°C with a CdTe source temperature of 646°C , yielding a hole density of 2 to $5 \times 10^{15} \text{ cm}^{-3}$. The CdTe films were deposited on high purity graphite substrates. Typically the CdS films were deposited in two layers: an initial layer next to the CdTe with a thickness of about $0.8 \text{ }\mu\text{m}$ that was not doped, and an upper layer with a thickness of about 1.2 to $1.7 \text{ }\mu\text{m}$ that was heavily In doped to produce a resistivity of less than 10^{-2} ohm-cm to reduce series resistance.

The open-circuit voltage of these solar cells varied with the

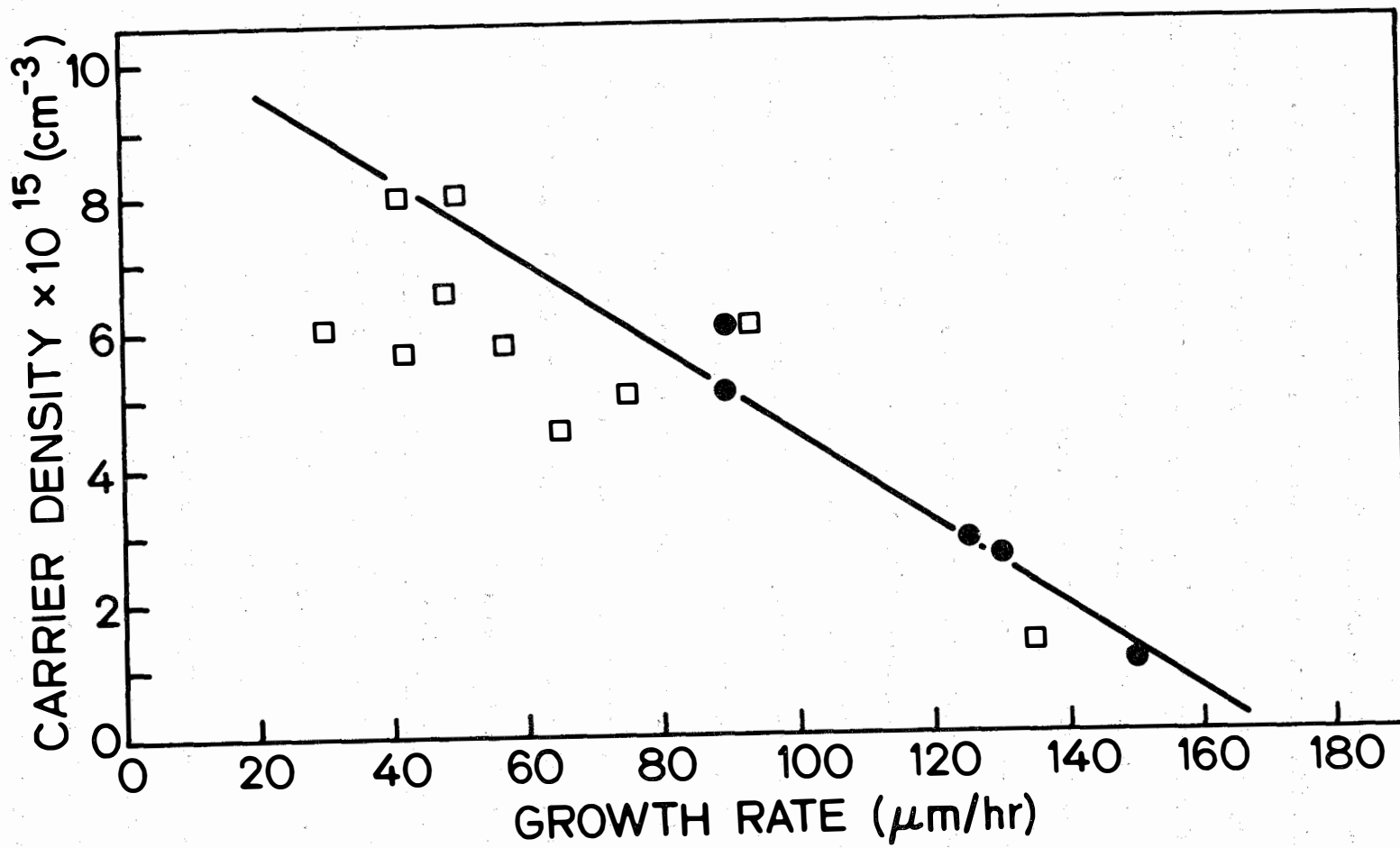


Figure 1. Hole density as a function of growth rate for CdTe deposited by HWVE on high purity graphite substrates. The solid points indicate a single series of films grown sequentially.

CdTe substrate temperature used for CdS vacuum evaporation, as shown in Figure 2. The dependence of V_{oc} on T_{sub} could be attributed to: (1) a change in the CdS resistivity with deposition temperature¹ with a junction mechanism similar to that described by Rothwarf² for CuInSe₂ solar cells, or (2) chemical changes at the interface or at the CdTe surface before CdS deposition due to the substrate temperature, e.g., the formation of a CdS_xTe_{1-x} interlayer as described by Uda et al.³.

A number of experiments were carried out in which T_{sub} was held constant at 160°C, the value at which the highest value of $V_{oc} = 0.67$ V has been observed in this research. Other solar cell parameters are a function of T_{sub} , but to a lesser extent than V_{oc} . A reduced transmission (see Figure 3) of CdS films grown at 160°C (compared to films deposited at 220 and 410°C) is consistent with a lower quantum efficiency in the near CdS bandgap region of the spectrum, as shown in Figure 4, and with a slightly lower short-circuit current.

Table I summarizes solar cell parameters as a function of T_{sub} .

Figure 5 shows the diffuse reflectivity of a CdS/CdTe solar cell (for $T_{sub} = 160^\circ\text{C}$). The reflectivity is approximately 13% and varies by less than 10% for wavelengths between 500 and 850 μm . This was a large area (0.81 cm²) ungridded solar cell.

A CdTe/graphite sample was heated in vacuum of less than 10^{-7} Torr at 300°C for 5 minutes and allowed to cool to 160°C before CdS deposition. The V_{oc} and other solar cell parameters were similar to those of devices made without the 300°C vacuum anneal. Changes that alter solar cell performance do not appear to occur during vacuum annealing alone.

To test whether In diffusing from the outermost CdS:In layer was essential for high V_{oc} at $T_{sub} = 160^\circ\text{C}$, a single undoped CdS layer about 2.0 μm thick was deposited on the CdTe/graphite substrate. The V_{oc} was not affected by the use of the single layer of CdS, although other cell

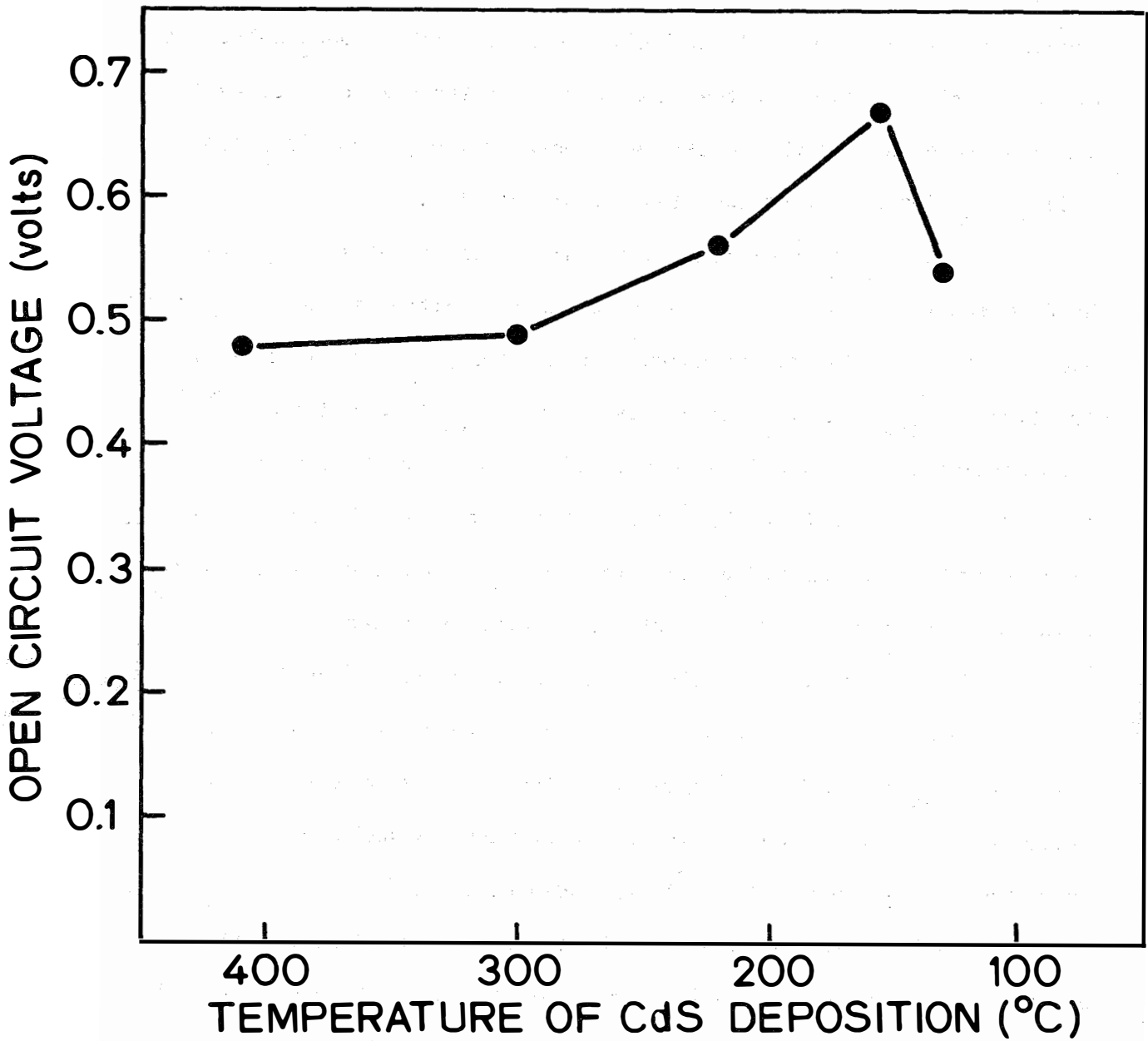


Figure 2. Open-circuit voltage as a function of CdS deposition temperature for In/CdS/CdTe/graphite solar cells with hole density in the CdTe between 2 and $5 \times 10^{15} \text{ cm}^{-3}$.

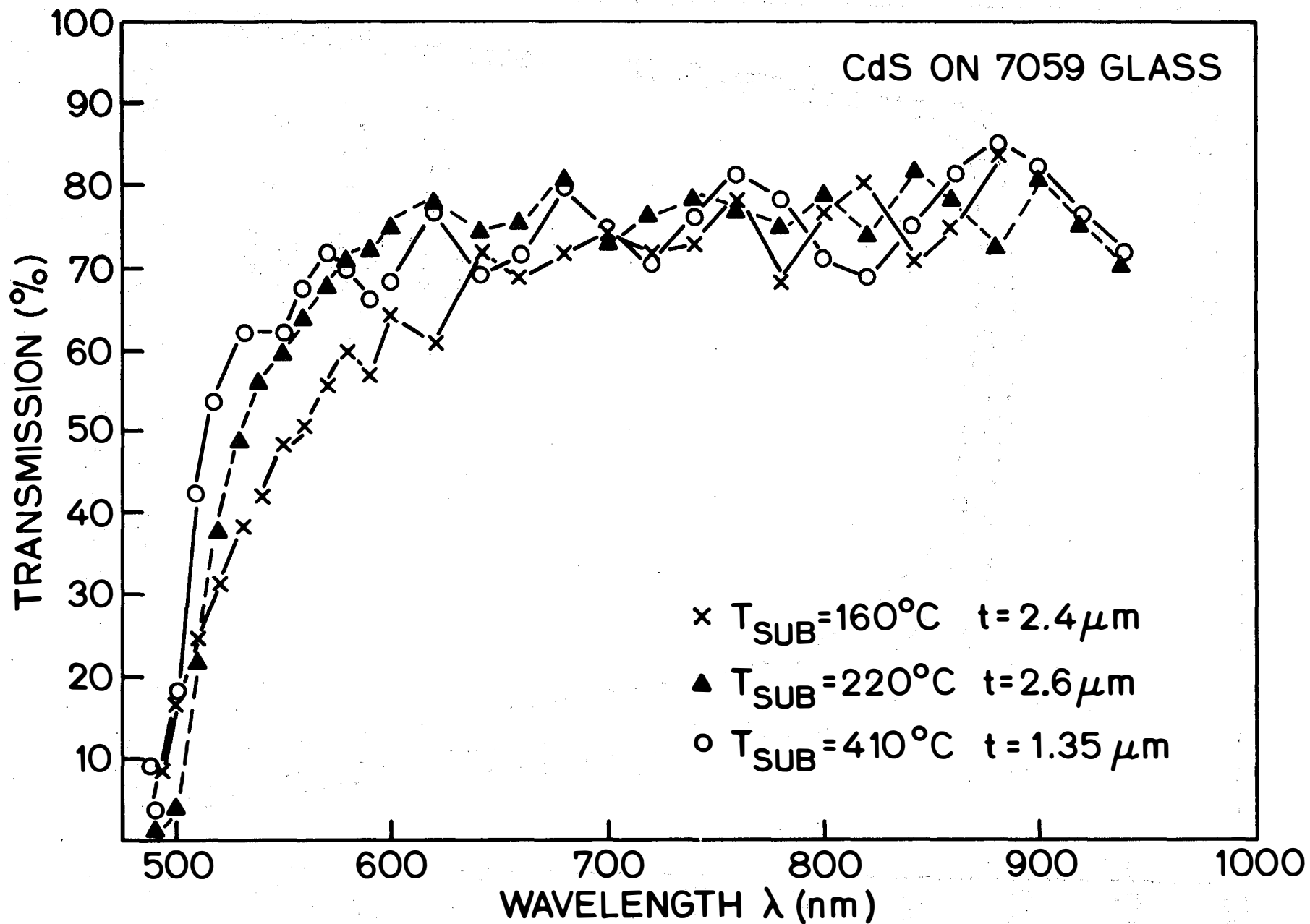


Figure 3. Optical transmission of CdS films for CdS deposited on glass at substrate temperatures of 160, 220 and 410°C.

CdS/CdTe CELLS

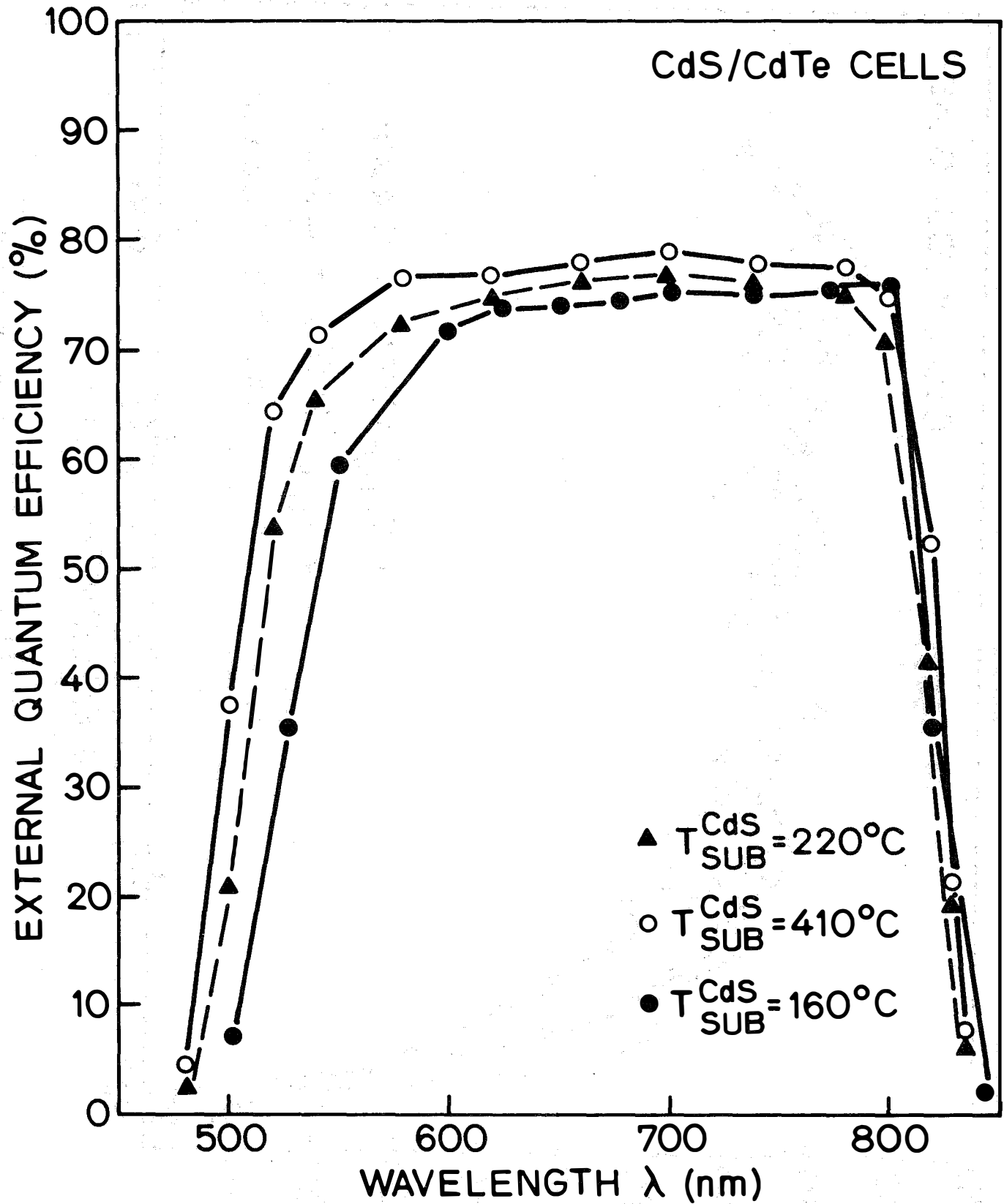


Figure 4. Spectral response for CdS/CdTe/graphite solar cells prepared with substrate temperatures for CdS deposition of 160, 220, and 410°C.

TABLE I

Thin Film CdS/CdTe Solar Cells^a

T _{sub} (CdS)	Area (cm ²)		V _{oc} , V	ff	J _{sc} (mA/cm ²) (active area)	Efficiency, % (active area)
	<u>Active</u>	<u>Total</u>				
130	0.12	0.14	0.54	0.25	2.5	less than 1%
160	0.12	0.14	0.67	0.47	12.7	4.3
160	0.72	0.78	0.65	0.46	13.2	4.3
220	0.12	0.14	0.56	0.52	16.2	5.2
300	0.089	0.11	0.48	0.57	16.1	4.8
410 ^b	0.12	0.14	0.48	0.49	15.8	4.5
410	0.12	0.14	0.62	0.45	6.9 ^c	2.1

^a Under simulated sunlight at 90.9 mW/cm².

^b Undoped CdS layer deposited at 410°C; doped CdS:In layer deposited at 300°C.

^c Apparent formation of buried homojunction.

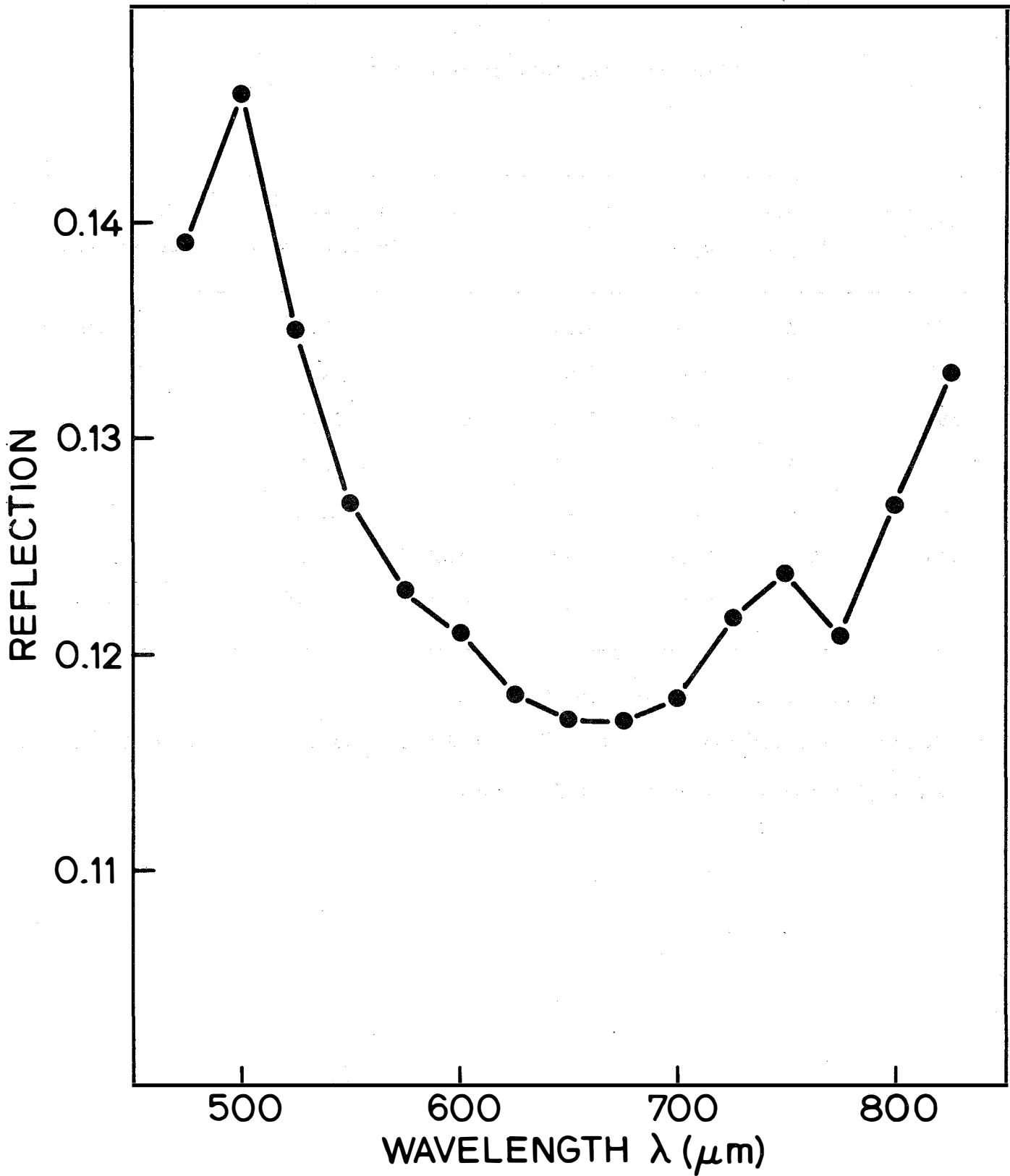


Figure 5. Diffuse reflectivity of CdS/CdTe/graphite solar cells with CdS deposited at a substrate temperature of 160°C.

parameters were adversely affected by the increased series resistance, as expected.

To test the effect of CdS resistivity ρ , a single CdS:In layer with $\rho < 10^{-2}$ ohm-cm was deposited at $T_{\text{sub}} = 160^{\circ}\text{C}$ directly on the CdTe/graphite substrate, resulting in a solar cell with a small $V_{\text{oc}} = 0.38$ V. The preparation of this cell was the same as those with intervening layers of insulating CdS that show high V_{oc} --except for the presence of the In. This suggests that the V_{oc} depends on the resistivity (and hence the carrier density) of the CdS at the interface, rather than a chemical interaction resulting in the formation of compounds such as $\text{CdS}_x\text{Te}_{1-x}$ or CdTeO_x .

The resistivity of CdS deposited at 130°C is lower by a factor of about two than the resistivity of films deposited at 160°C , but have lower $V_{\text{oc}} = 0.54$ V. A summary of all cells deposited at $T_{\text{sub}} = 160^{\circ}\text{C}$ is given in Table II.

EBIC scans (see Figure 6) show the uniform collection over the entire CdS surface typical of solar cells made with CdS layers about $2 \mu\text{m}$ thick. An attempt was made to increase the transmission of light in the short wavelength region by using a very thin CdS film (300A undoped + 300A CdS:In). The CdS layer appears to be discontinuous in the EBIC scan shown in Figure 7, and the light current is very small.

Deposition of CdTe on Au- and Cu-Coated Graphite Substrates

Either 5 to 50A of Cu, or 20A of Au, was vacuum evaporated on $6 \times 12 \text{ mm}^2$ graphite substrates. CdTe was simultaneously deposited on coated and uncoated graphite substrates for $T_{\text{sub}} = 583^{\circ}\text{C}$ and $T_{\text{CdTe}} = 646^{\circ}\text{C}$, conditions that normally produce hole densities in the 2 to $5 \times 10^{15} \text{ cm}^{-3}$ range. CdTe deposited on the uncoated graphite substrates had hole densities within this range in all cases.

Microprobe analysis of the CdTe film grown on 20A of Au on graphite indicates a Au density of 0.03 at.%. The density of the Au is consistent with the Au being dispersed throughout the film, since the microprobe samples to depth of $0.5 \mu\text{m}$ and the film exceeded $35 \mu\text{m}$ in thickness. The growth rate on the Au-coated graphite was over three

TABLE II

Properties of CdS/CdTe/graphite Solar Cells^a

Cell No.	Sample Preparation	V_{oc} , V	J_{sc} , mA/cm ²	ff	Effic., %	A	J_o , A/cm ²	ρ_{CdS} , ohm-cm
UD36	Standard	0.66	12.1	0.47	4.3	1.9	7×10^{-9}	149
UD63A	Single un-doped CdS layer	0.64	7.0	-	-	2.0	7×10^{-9}	149
UD65A	Single doped CdS layer	0.38	1.2	-	-	1.2	8×10^{-8}	0.01
UD65B	300°C vacuum anneal before CdS deposition	0.65	13.0	0.44	4.5	1.84	3×10^{-8}	149
UD36	Completed cell H ₂ annealed 5 min, 300°C	0.60	12.5	0.48	-	1.75	3×10^{-9}	-
UD44B	Large cell	0.65	12.2	0.42	4.3	1.9	1×10^{-8}	149
UD75	300A undoped CdS + 300A CdS:In (dis-continuous CdS film)	0.65	-	-	-	2.1	1×10^{-8}	149

^a All CdS films deposited at a CdTe/graphite substrate temperature of 160°C.

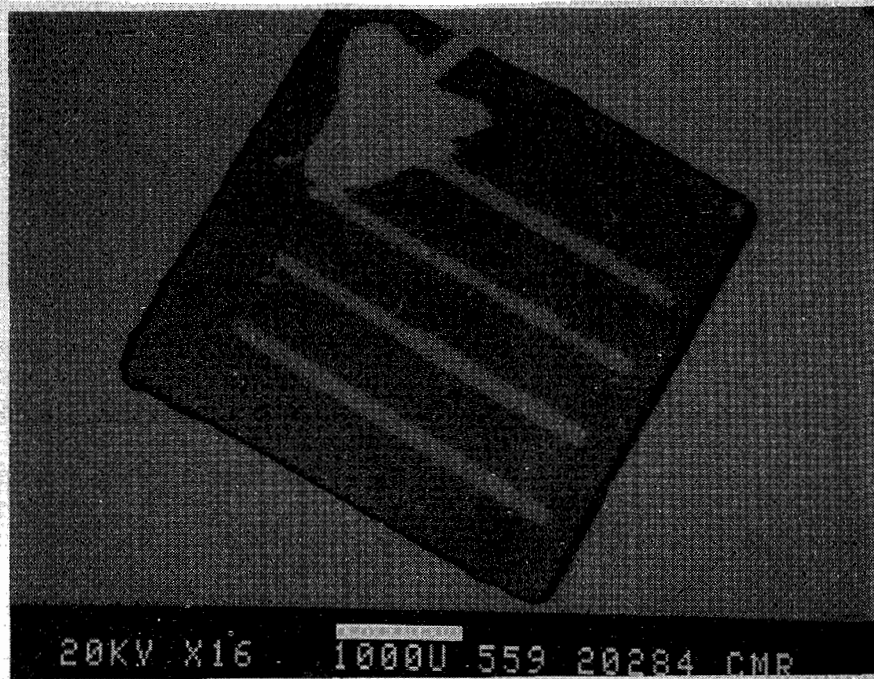


Fig. 6. EBIC scan of CdS/CdTe solar cell with a CdS layer $\sim 2 \mu\text{m}$ thick (magnification 16X). Collection appears to be uniform over the whole device, except for the grid lines and Ag paint contact. (Negative image: dark areas show greatest collection).



Fig. 7. EBIC scan of a CdS/CdTe/graphite cell with a very thin CdS layer (2 layers of 300 Å each) (magnification 136X). The CdS coverage appears to be discontinuous and collection occurs only around the In contact strip. (Positive image: light areas show greatest collection).

times larger than the growth rate on the uncoated substrate grown simultaneously, as summarized in Table III.

In diodes were applied to both the CdTe on Au-coated graphite and the CdTe on uncoated graphite. Hole densities measured by C-V measurements indicate that the hole density is lower on the Au-coated substrate than on the uncoated substrate. J vs V measurements (see Figure 8) indicate a larger series resistance for the sample grown on the Au-coated graphite, consistent with a lower hole density. Other diode characteristics for the Au-coated graphite substrates were remarkably unaffected by the presence of Au, as indicated in Table III.

Microprobe analysis of the CdTe films grown on Cu-coated graphite indicates as much as 1 at.% Cu present (for CdTe grown on 50A Cu). The hole density was lower than for films grown on uncoated graphite, but to a lesser extent than for the films grown on Au-coated graphite. Microprobe analysis of CdTe grown on 50A-Cu-coated graphite indicates a uniform surface distribution as shown in Figure 9. The concentration indicated is somewhat large, but the quantitative accuracy of the microprobe is insufficient to permit detailed conclusions.

A 100A Te overlayer was applied to a 50A-Cu-coated graphite substrate to enhance the possibility of Cu_2Te formation, which could lead to reduced Cu diffusivity. Te-coated Cu-coated graphite substrates did not appear to be different from non Te-coated samples.

The diffusion coefficient and solubility of Cu (and Au) in CdTe is high,⁴ but the electrical activity is critically dependent on the growth conditions - in particular, the Cd partial pressure.⁵ Attempts will be made to anneal and deposit CdTe on Cu-coated substrates under conditions favorable to produce electrically active species.

CdS/graphite Junctions

CdS was deposited on a high purity graphite substrate at 350°C.

TABLE III

Properties of CdTe Films Deposited on Au-coated or Cu-coated Graphite

Coating	A (In diode)	N_A , cm^3	Growth Rate, $\mu\text{m/hr}$
None	1.7	$2.2 - 2.8 \times 10^{15}$	31
20A Au	1.7	3×10^{14}	115
5A Cu	1.7	1.6×10^{15}	48
50A Cu	-	1×10^{15}	21.5

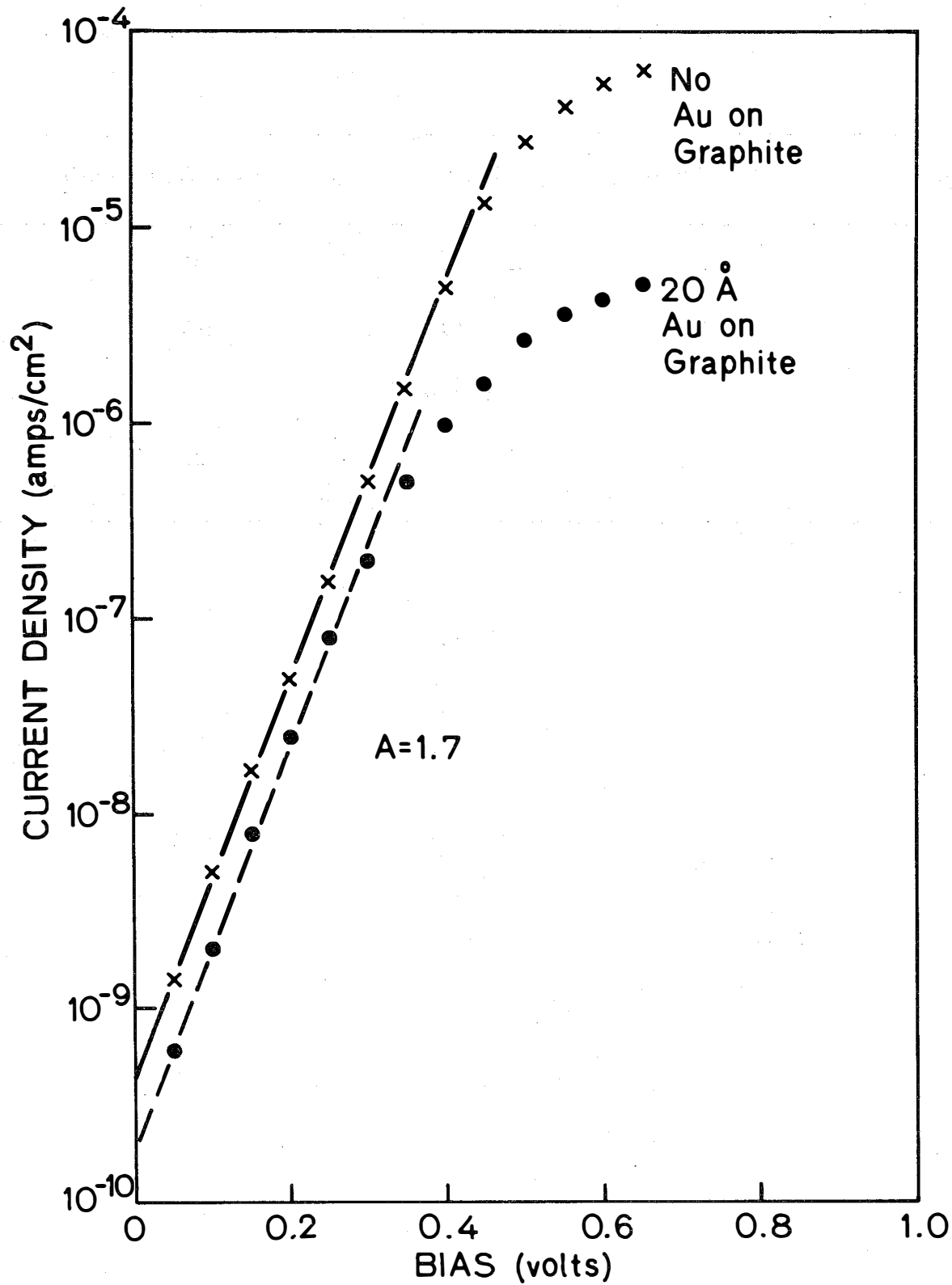


Figure 8. Current as a function of voltage for In/CdTe diodes on CdTe films deposited on uncoated graphite and on graphite coated with 20Å of Au.

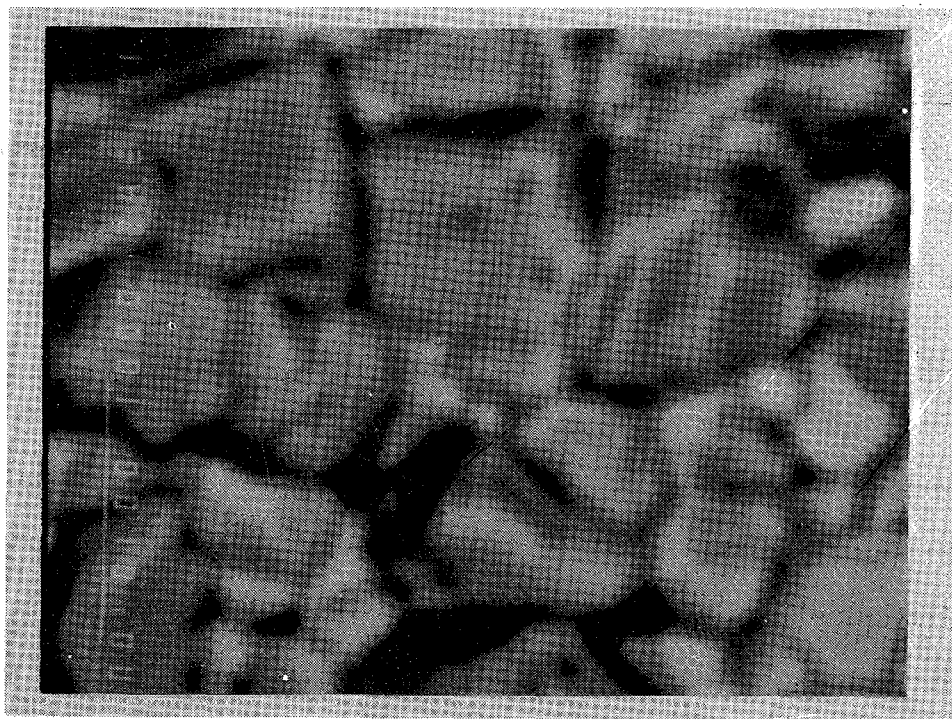
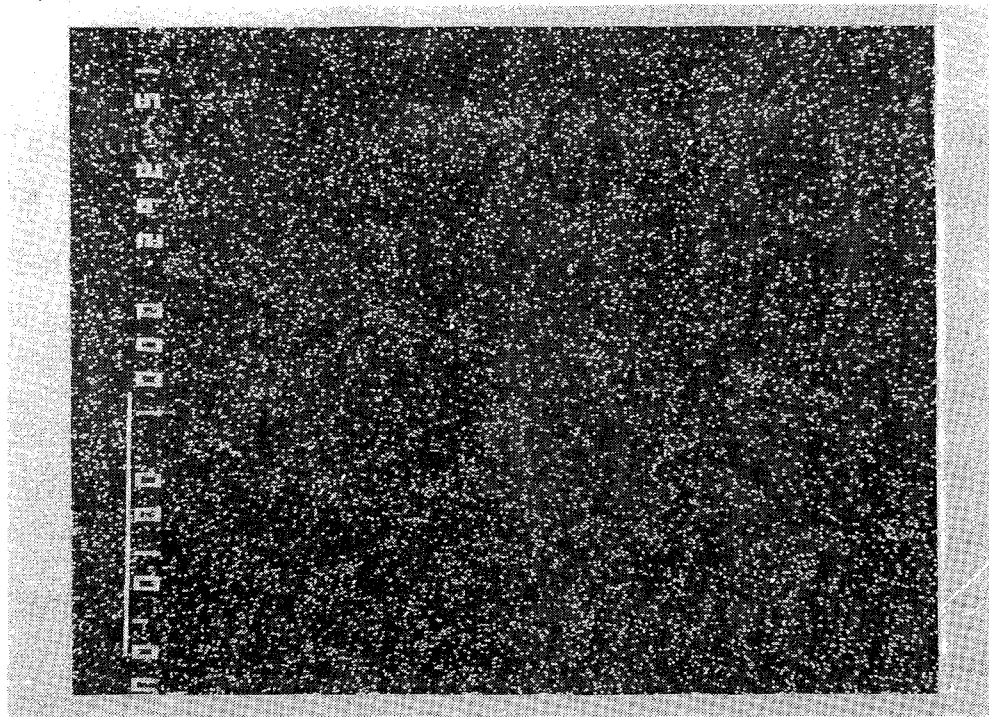


Fig. 9. (Top) Distribution of Cu on the surface of a p-CdTe film deposited on a graphite substrate previously coated with 50 Å of Cu. Measured by electron microprobe. (Bottom) SEM image of the same sample. The line at the left of each image indicates 10 μm .

In and Au dots were deposited by vacuum evaporation. Both metals appeared to make ohmic contact to the CdS. Two-point measurements using either metal dot (0.0256 cm^2 in area) and the graphite substrate indicated a rectifying junction between the graphite and the CdS with $A = 1.8$ and $J_0 = 3 \times 10^{-5} \text{ A/cm}^2$. Initial C-V measurements suggest an electron density of 10^{18} cm^{-3} , which is unusually high. Further research will be directed toward the use of the CdS/graphite junction as an analytical tool.

Growth of CdTe with a Cu Source

Initial attempts to deposit CdTe with a partial pressure of Cu to provide Cu doping, proved unsuccessful. Cu source temperatures of 800°C failed to produce an increase in the hole density in CdTe films. Higher source temperatures are not possible with the present HWVE apparatus.

RGA Installation

Initial RGA tests and installation are now complete. Modification of the HWVE heat shields is also completed, and initial growths monitored by the RGA should occur shortly.

Stability of CdS/CdTe Thin Film Solar Cells

The photovoltaic parameters of CdS/CdTe solar cells as a function of time are given in Figure 10. For Cell #UD37 the undoped and doped CdS layers were deposited at 220°C ; for Cell #UD41A the undoped CdS interlayer was deposited at 410°C and the CdS:In layer was evaporated at 300°C . The cells were stored in the dark in a laboratory environment and were heat treated as indicated in hydrogen at 300°C for 5 minutes at different times.

The values of the short-circuit current J_{sc} are based on active area and were measured under simulated sunlight of 90.9 mW/cm^2 . Negligible changes of J_{sc} with time and hydrogen heat treatment were observed.

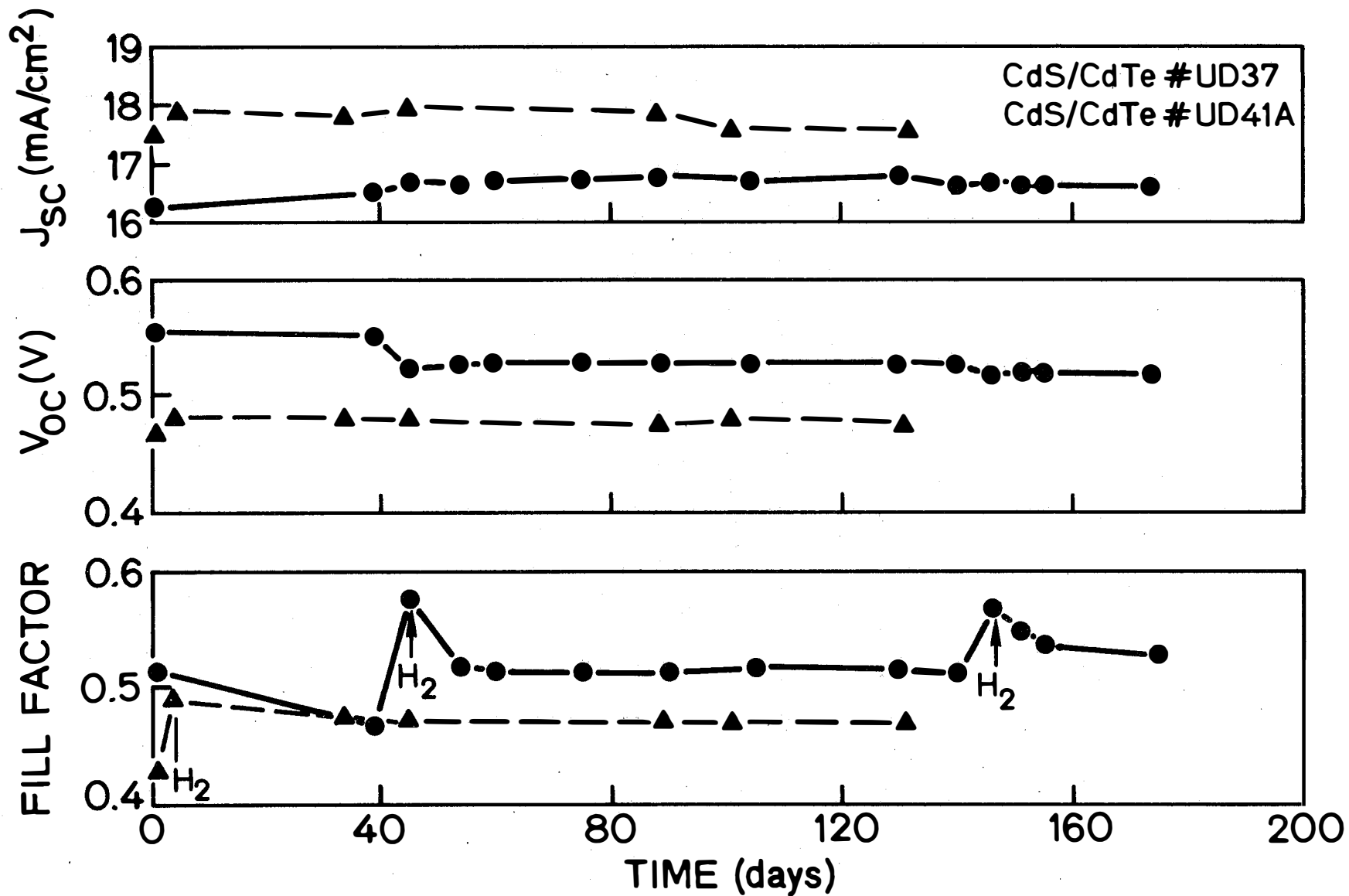


Figure 10. Photovoltaic parameters of CdS/CdTe/graphite thin film solar cells as a function of time. The cells were stored in the dark in a laboratory environment. The cells were hydrogen heat treated at 300°C for 5 min as indicated.

The open-circuit voltage of Cell #UD37 is slightly reduced by the hydrogen heat treatment. The as-deposited cell had a value of $J_0 = 2.5 \times 10^{-8} \text{ A/cm}^2$ and $A = 1.77$. After the first hydrogen heat treatment the J-V curve can be fitted by a simple diode relation with $J_0 = 9.6 \times 10^{-9} \text{ A/cm}^2$ and $A = 1.55$. For Cell #UD41A no change in the diode parameters with hydrogen heat treatment was found.

The largest effect of the hydrogen heat treatment is on the fill factor of our CdS/CdTe heterojunctions. The fill factor of the non-heat treated cell (#UD37) decreased by 11% over a period of 39 days. A hydrogen heat treatment increased the fill factor from 0.47 to 0.57. Fifteen days after the heat treatment the fill factor of this cell was about identical with the value measured immediately after cell deposition. However, the fill factor is subsequently stable over a period of three months after the heat treatment. A similar behavior was observed after a second hydrogen heat treatment. The Cell #UD41A, heat treated in hydrogen shortly after cell fabrication, showed no degradation effects with time.

The effects of heat treatments in air and hydrogen on the hole density, light resistance and fill factor were analyzed for the CdS/CdTe Cell #UD37. All heat treatments were for 5 min. These three parameters are plotted as a function of the heat treatment temperature in Figure 11. The value of the hole density was obtained from C-V measurements on the CdS/CdTe junction. The light resistance was estimated from the slope of the light J-V curve at V_{oc} .

For temperatures greater than 150°C , the air heat treatment significantly increased the light resistance and consequently decreased the fill factor. The hydrogen heat treatment reduced the resistance under illumination to a value of 7.1 ohm-cm^2 and improved the fill factor. The heat treatment in different atmospheres did not change the hole

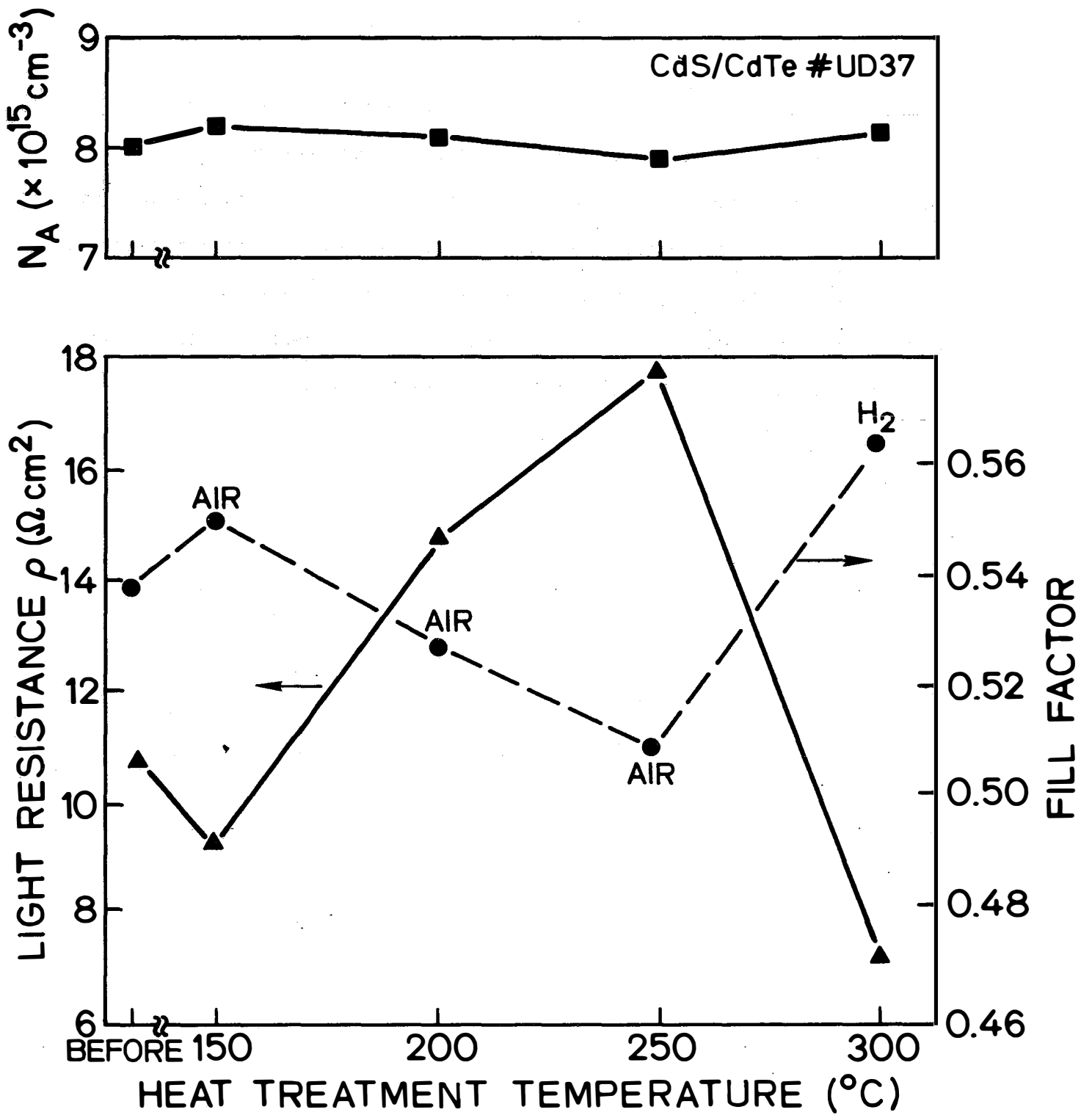


Fig. 11. The hole density N_A , light resistivity ρ , and fill factor of CdS/CdTe/graphite cell #UD-37 as a function of the heat treatment temperature in different atmospheres.

Average optical cross sections for boundary defect states and majority carrier capture coefficients were also determined. These values were obtained using the results of the above-mentioned photoconductivity decay experiments, with additional information obtained from photocapacitance vs light intensity measurements. Optical cross sections were typically around 10^{-16} cm^2 , and capture coefficients ranged from $10^{-8} \text{ cm}^3/\text{sec}$ to $8 \times 10^{-10} \text{ cm}^3/\text{sec}$. The experiments indicated that thermal terms are negligible in a generation-recombination balance of the kinetics of an illuminated grain boundary at 300°K .

Grain boundary passivation experiments indicated that H_2 and Li effectively passivate boundaries in p-type material, although the effect is not stable with time. Only air has been found to be effective in passivating grain boundaries in n-type CdTe.

In an effort to produce more stable passivation effects in p-type material, several CdTe films were annealed at 500°C in a hydrogen plasma. The plasma was produced by evacuating a small glass chamber, admitting molecular hydrogen to a pressure of about 1 Torr, and discharging a Tesla coil across the chamber. Instantaneous effects on film resistivity were similar to those obtained with molecular hydrogen, but an unknown surface reaction occurred, indicated by surface discoloration. Stability of the passivation appeared to be somewhat greater than that found for molecular hydrogen, but is still definitely a transient effect. Figure 12 shows the variation of film resistivity vs time for a film annealed in a hydrogen plasma.

Finally, an experiment was performed to determine whether a CdS/CdTe solar cell could reasonably be fabricated that incorporated the hydrogen passivation effects of CdTe grain boundaries. Since it is necessary to heat CdTe films in vacuum in order to deposit CdS, it was thought that the heating process might considerably accelerate the

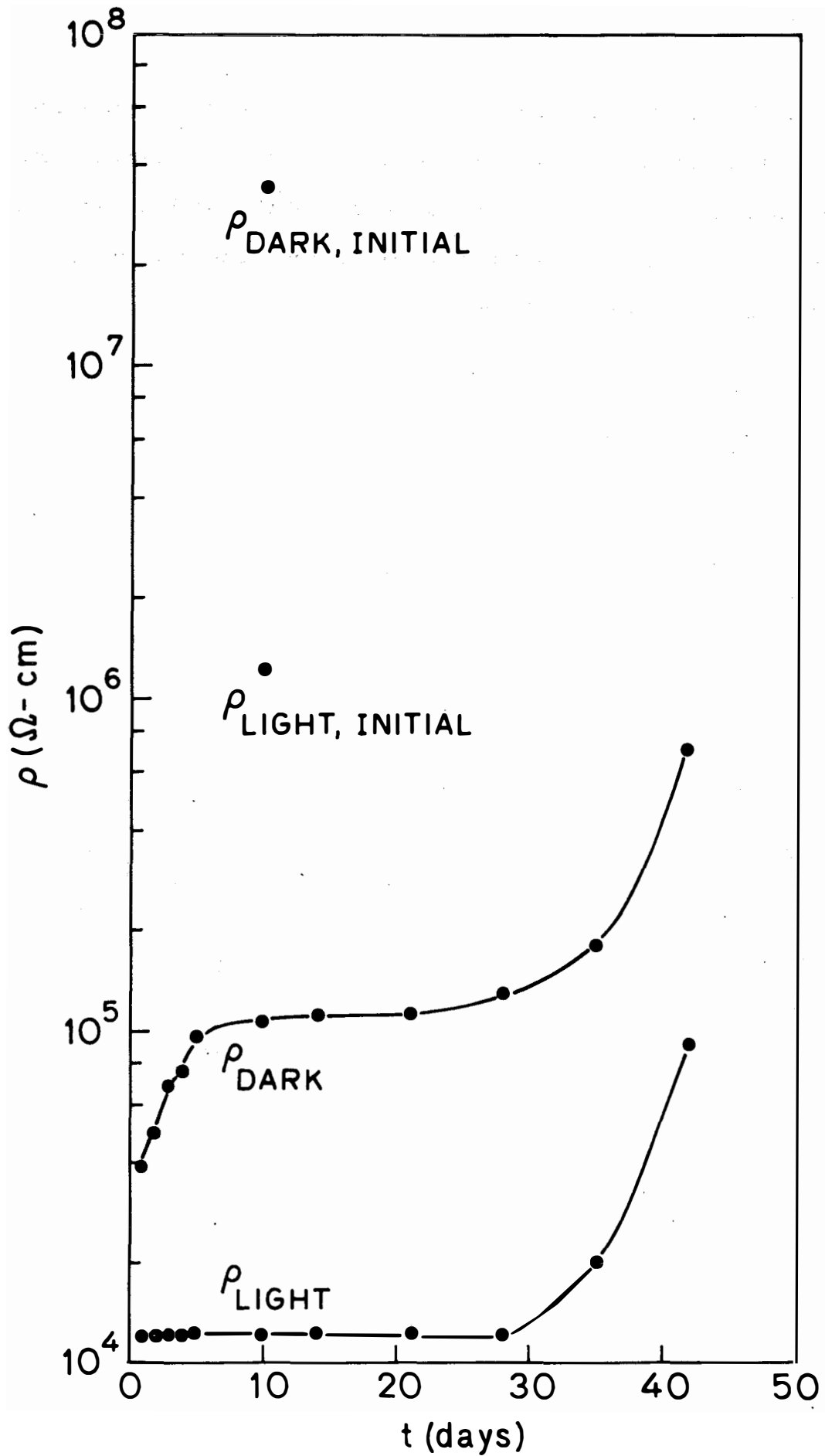


Figure 12. Dark and light resistivity of a CdTe/graphite film before annealing in a hydrogen plasma at 500°C , and after annealing as a function of time.

decay of passivation effects, and thus eliminate any beneficial alteration of cell characteristics. It was found, however, that passivation effects remained substantial after heating in vacuum at 160°C, a viable temperature for CdS deposition. A solar cell with a hydrogen passivated CdTe film will be fabricated in the near future.

BOND LENGTH CHANGES RESULTING FROM IMPURITY INCORPORATION

A computer model has been developed to investigate the bond length changes produced by an impurity atom occupying a substitutional site in the CdTe lattice, assuming that the interaction between the impurity and the lattice extended to first and second nearest neighbors only, and using the tight binding theory developed by Harrison⁹ to determine the equilibrium bond lengths that result from impurity incorporation.

One of the applications of the model of particular interest to device application is the interaction of impurities with native defects through a strain interaction. Native defects (e.g., Cd vacancy, Te vacancy, Cd interstitial) produce pronounced lattice strains. Crystals grown with an excess Cd (typically producing n-type material through either Te vacancy or Cd interstitial donors) cause the lattice to contract with the lattice constant changing from 6.482Å to 6.480Å. Crystals grown with excess Te (producing p-type material most likely through Cd vacancy acceptors) cause the lattice constant to increase to 6.488Å.¹⁰ It is a natural consequence of equilibrium dynamics that factors that decrease the formation energy of native defects, e.g., through strain energy interaction, result in higher concentrations of that defect. Interactions that increase the formation energy result in lower concentrations.

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16. Abstract (Limit: 200 words) Variation of CdS/CdTe/graphite thick film solar cell properties was investigated as a function of temperature for CdS film deposition. A maximum open-circuit voltage of 0.67 V was found for a deposition temperature of 160°C, corresponding to a CdS film resistivity of 150 ohm-cm. The effect is not due to avoidance of higher temperature annealing of the CdTe film in higher temperature CdS film depositions nor to the diffusion of In from the outermost CdS: In layer. The effect of coating the graphite before CdTe deposition with Au or Cu was also investigated. Although high concentrations of both Au or Cu could be determined after CdTe deposition, CdTe films grown on this coated graphite had lower hole densities than films grown on uncoated graphite. Photovoltaic parameters of thin-film CdS/CdTe/graphite solar cells were investigated as a function of storage time to check the stability of these cells. Initial degradation of parameters (especially fill factor) could be reversed by heat treatment in hydrogen, with subsequent properties being stable. Heat treatment of CdS/CdTe/graphite solar cells in air increases cell resistivity and decreases fill factor; heat treatment in hydrogen produces the reverse effect. The hole density is not affected by these heat treatments, suggesting that effects are associated with grain boundaries in the film.			
17. Document Analysis			
a. Descriptors			
b. Identifiers/Open-Ended Terms			
a. Photovoltaic Cells, Cadmium Telluride Solar Cells, Solar Cells.			
b. Cadmium Telluride Films, Cadmium Telluride Single-Crystals			
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