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# DOE/SERI Polycrystalline Thin-Film Photovoltaic Research

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**NOTICE**

# DOE/SERI POLYCRYSTALLINE THIN-FILM PHOTOVOLTAIC RESEARCH

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## ABSTRACT

This paper presents recent results, status, and future prospects for the U.S. Department of Energy's (DOE's) Polycrystalline Thin Film Photovoltaic Program, managed by the Solar Energy Research Institute (SERI). The devices being studied most intensively are heterojunctions based on $CuInSe_2$ and on CdTe. Both materials have attained over 10% efficiency in polycrystalline form. The main emphasis is on $CuInSe_2$, for which Boeing has reported an 11%-efficient device (AM1 ELH simulation). Important work is being done on studies of the composition/electronic properties of $CuInSe_2$ and its response to post-deposition annealing. In the CdTe research, ohmic, stable back-contacting and control of p-type doping are being investigated. New efforts to study polycrystalline two-junction stacked cells are underway with two-terminal cells (at IEC) and with four-terminal cells (at SMU). This preliminary work is expected to be expanded, with emphasis on CdTe and other top-cell (high-bandgap) materials. These efforts introduce a number of new research areas (e.g., transparent ohmic contacts to p-CdTe and sub-bandgap light-losses in polycrystalline materials). The aim of the program is to produce stable, high-efficiency (15%), thin-film cells that can be deposited inexpensively by techniques that are scalable to large areas.

## INTRODUCTION

This paper presents recent results, status, and future prospects for the U.S. Department of Energy's Polycrystalline Thin Film Photovoltaic Program, managed by the Solar Energy Research Institute (SERI). (There is substantial work being done at SERI on cell fabrication and analysis, especially in $CuInSe_2$, but SERI in-house work is covered in another article in this publication).

The PV status of the thin-film devices under study is shown in Table 1. All efficiencies are total-area efficiencies. Emphasis is presently on $CuInSe_2$ and CdTe cells.

## COPPER INDIUM DISELENIDE

The 11% $CuInSe_2$ PV cell developed and reported by Boeing under SERI subcontract is the highest-efficiency thin-film, non-single-crystal solar cell in the DOE program.[1] Efficiencies of 10% are commonly reported in the laboratory.

$CuInSe_2$ has demonstrated excellent stability. No degradation has been reported by Boeing in cells subjected to constant AM1 illumination for 9300 hours[2] at temperatures up to 80°C (preliminary studies on three

**TABLE 1. Polycrystalline Thin-Film Program Cell Parameter Status**

| Device Structure | Configuration | Deposition Technique | Efficiency Reported | $V_{oc}$ (volts) | $J_{sc}$ (mA/cm²) | FF | Area (cm²) | Subcontractor |
|---|---|---|---|---|---|---|---|---|
| $(Cd_{0.8}, Zn_{0.2})S/CuInSe_2$ | Backwall | Vacuum Evaporated from CdS, ZnS/Cu,In,Se | 11.00 | 0.4373 | 38.50 | 0.6531 | 1.00 | Boeing |
| $CdS/CuInSe_2$ | Backwall | Knudsen Cell Evaporated | 8.05 | 0.392 | 31.3 | 0.656 | 0.07 | IEC |
| $CdS/CuInSe_2$ | Backwall | Evaporated CdS/Elemental Reactive Sputter Deposited in $H_2Se$ | 4.1 | 0.253 | 28.5 | 0.50 | 0.075 | Telic |
| n-ITO/p-CdTe | Backwall | Ion-Beam Sputtered/$H_2$ Transport CVD | 8.2 | 0.75 | 18.0 | 0.60 | 1.0 | SMU |
| Au/n-CdTe | (Schottky) | Vacuum Evaporated/ $H_2$ Transport CVD | 5.28 | 0.40 | 22.0 | 0.60 | 8.0 | SMU |
| CdS/CdTe | Backwall | Vacuum Evaporated/ Hot Wall Vacuum Evaporated | 4.7 | 0.54 | 16.0 | 0.54 | 0.12 | Stanford |
| $CdS/Cu_{1.8}Se$ | Backwall | Vacuum Evaporated from CdS/Cu,Se | 5.38 | 0.457 | 18.7 | 0.63 | 1.0 | Boeing |
| $Mg/Zn_3P_2$ | (Schottky) | Vacuum Evaporated/ Close-Spaced-Vapor Transported | 4.3 | 0.43 | 16.8 | 0.53 | 1.0 | IEC |

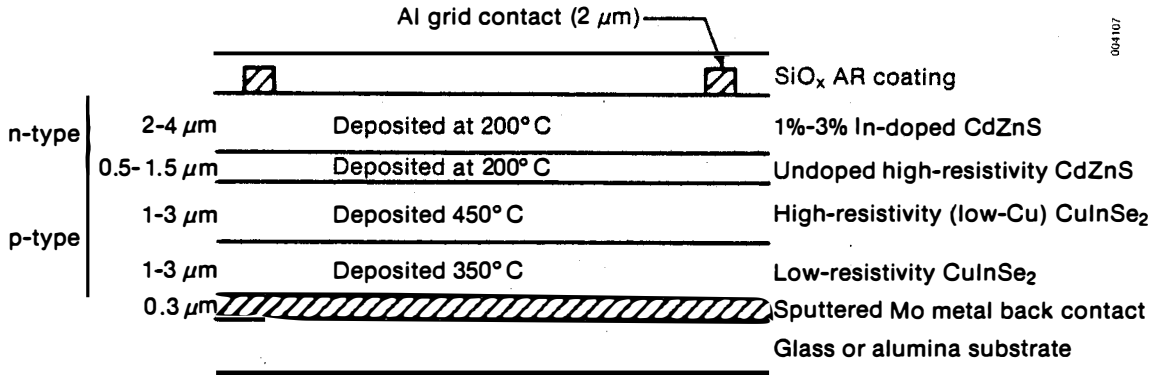


**Figure 1. The Boeing CdZnS/$CuInSe_2$ cell, showing deposition temperatures, film thicknesses, and compositions of the layers during deposition.**

unencapsulated cells in Seattle air). Some performance degradation has occurred in unencapsulated cells (1) when the cells were mechanically damaged during test contacting and (2) at higher temperature (over 80°C) and in moist atmospheres when grid-degradation occurs. Independent Battelle testing of Boeing $CuInSe_2$ cells showed similar results for 6000 hours of exposure. Some contact degradation was reported and attributed to chemical attack on the grids and formation of aluminum chloride. Limited spectral response studies showed the $CuInSe_2$ itself to be undamaged over the course of the test.[3] Early work on the fundamental properties of $CuInSe_2$ has been done by Jaffe and Zunger.[4] They called particular attention to the stability of $CuInSe_2$ to the movement of Cu as compared with that movement in Cu-binary compounds. This is particularly heartening because the movement of Cu atoms in binary semiconductors (e.g., $Cu_2S$, $Cu_2Se$) has led to instabilities such as the formation of Cu nodules that short-circuit the cells. The fundamental stability of Cu-ternaries, suggested by Jaffe and Zunger, gives impetus to the development of Cu-ternary rather than binary compounds, despite their greater complexity.

The highest-efficiency $CuInSe_2$ devices within the DOE program are being deposited at Boeing.[1] Boeing's vacuum-evaporated cell (Fig. 1) is a thin-film polycrystalline heterojunction with a $CuInSe_2$ absorber and a large-bandgap (2.5 eV) (CdZn)S window. The $CuInSe_2$ layer is deposited in a two-layer configuration, but mixing takes place during deposition (Table 2). The final cell layer composition and electronic properties are under close study.[5] Figure 2 suggests that the layers in the final $CuInSe_2$ cell are homogeneous within 0.5 atomic-percent measurement error. However, sub-atomic-percent inhomogeneities (if they exist) could be responsible for significant variations in electronic properties.

Boeing fabricates $CuInSe_2$ films via an open-boat vacuum evaporation system. The elemental flux from the heated boats is measured by electron impact emission spectroscopy, and a feedback loop is used to control the temperature of the boats and the evaporation rates.

Boeing is also a partner in a joint-venture (Sovolco) with Reading and Bates to commercialize their cells. Sovolco has had preliminary success in depositing high-efficiency cells on low-cost substrates.

**TABLE 2. EDAX-Measured Compositions of $CuInSe_2$ Diagnostic Side-Strip Layers Formed during the Deposition of Mixed-Layer Cells[1]**

| | Composition (Atomic Percent) | | |
|---|---|---|---|
| | Copper | Indium | Selenium |
| Layer* | | | |
| 1 | 26.5 | 24.0 | 49.5 |
| 2 | 19.3 | 29.1 | 51.6 |
| 1+2 | 25.1 | 26.3 | 48.6 |
| 1 | 26.2 | 24.8 | 49.0 |
| 2 | 20.0 | 28.0 | 52.0 |
| 1+2 | 24.6 | 26.5 | 48.9 |
| 1 | 25.6 | 26.6 | 47.8 |
| 2 | 20.7 | 28.7 | 50.6 |
| 1+2 | 23.7 | 27.6 | 48.7 |

*Note: This table represents the composition of various diagnostic layers formed during the deposition process. The Cu/In/Se is deposited on nonmetallized alumina side-strips adjacent to the molybdenum-coated alumina substrate used for the composite $CuInSe_2$ absorber used in cell fabrication. Measurement error is about 1%, and the Se values are especially likely to be sensitive. The different layers (1, 2, 1+2) are separately measured on different side-strips. Layer 1 is deposited first, and is made with slightly more Cu than stoichiometric $CuInSe_2$. Layer 2 is the $CuInSe_2$ layer nearest to the (Cd, Zn)S and has significantly less Cu. Layer 1+2 is a composite of both layers 1 and 2 and is most approximate to the composition of the $CuInSe_2$ in the actual cell. Since these side-strips are separate, they have a purely diagnostic role. As pointed out elsewhere (Fig. 2), mixing takes place in the actual film used in the cell, where all the layers are deposited consecutively on a molybdenum-coated substrate.

The Institute of Energy Conversion (IEC) is also making high-efficiency (8%) $CuInSe_2$ devices. IEC uses a feed-forward system of controlled Knudsen cell effusion sources, wherein temperature and aperture are carefully calibrated and programmed to deposit the desired films. The effusion cell design has demonstrated large-area scalability with other solar cell materials, which is important for the future commercial impact of $CuInSe_2$.

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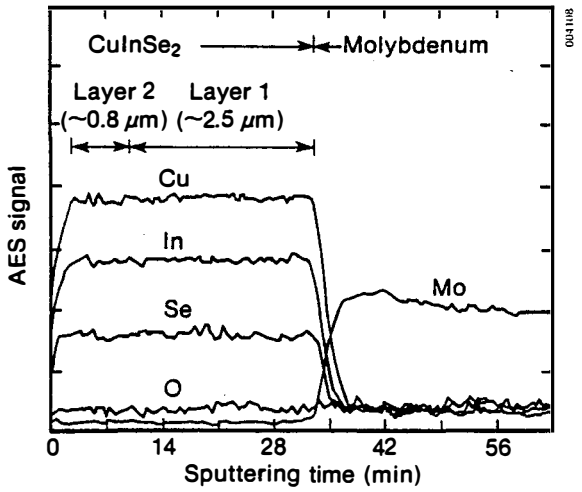


**Figure 2. Compositional uniformity of Boeing $CuInSe_2$ film (SERI).** The Cu, In, and Se concentrations do not vary significantly from 25%, 25%, 50% (within 0.5 atomic % measurement error) over the thickness of the film, showing that the layer homogenizes during deposition. However, the bilayer deposition technique appears to produce more efficient cells. Sub-atomic-percent compositional inhomogeneities may be present: carrier concentrations and conductivity type change very rapidly at near-stoichiometric compositions.

An important and as yet not very well understood phenomenon is that high-efficiency $CuInSe_2$ cells all require post-deposition oxygen heat treatments. Both Boeing and IEC find efficiency enhancements associated with the heating of completed CdS/$CuInSe_2$ devices in $O_2$ at 200°C. Table 3 shows the results of one series of annealings for a Boeing cell. Additionally, some IEC cells start with no appreciable PV performance and reach 6%-7% efficiencies after heat treatment. Treatment can be fast or slow: two months at room temperature can produce similar results to an hour at 200°C. Work is being done by several groups to understand the effects of the heat treatment on the $CuInSe_2$ devices.

**TABLE 3. Heat Treatment of a Boeing CdS/$CuInSe_2$ Solar Cell in Oxygen at 200°C**

| Cumulative Bake Time | Efficiency (%) | $V_{oc}$ (V) | $I_{sc}$ (mA) | FF |
|---|---|---|---|---|
| Unbaked | 4.19 | 0.312 | 32.55 | 0.412 |
| 10 min. | 6.77 | 0.377 | 35.35 | 0.509 |
| 20 min. | 7.20 | 0.379 | 35.59 | 0.537 |
| 30 min. | 7.33 | 0.384 | 35.68 | 0.536 |
| 40 min. | 7.54 | 0.388 | 35.60 | 0.546 |

## CADMIUM TELLURIDE

CdTe is a prime PV candidate due to its ideal direct bandgap (near 1.5 eV), high optical absorption coefficient (greater than $3 \times 10^4$ $cm^{-1}$ at the band edge energy), and relative ease of deposition. Homojunction devices based on single-crystal CdTe reportedly have exhibited efficiencies in excess of 13%,[6] and a privately funded Kodak group has reported a polycrystalline thin-film CdS/CdTe device with 10.9% efficiency (AM2). The CdS and CdTe in this latter case were deposited by close-spaced sublimation. Electrodeposited CdS/CdTe thin-film heterojunction devices have been reported by Ametek, Inc., with about 8% efficiencies and by Monosolar, Inc., with above 9% efficiencies (measured at SERI). A Japanese company, Matsushita, reports an active-area efficiency of 12.8% for small backwall n-CdS/p-CdTe cells made by screen printing. However, due to the placement of electrodes, the actual total-area efficiency is substantially less.

The DOE/SERI program has been supporting research in which CdTe is formed by chemical vapor deposition (CVD), by hot wall vacuum evaporation (HWVE), and by electrodeposition. CVD is a scalable deposition process, and SERI-sponsored research at Southern Methodist University has demonstrated a reported 8.2%-efficient ITO/CdTe device in which the CdTe was deposited by CVD and the ITO was ion-beam sputtered. HWVE p-CdTe/n-CdS devices have been studied at Stanford University and in-house at SERI. The most recent results from Stanford have shown 4.7%-efficient CdS/CdTe heterojunctions in which both materials were deposited by HWVE.

A major objective in the SERI/DOE program has been to find a way to make a stable ohmic contact to p-CdTe. One possible solution is use of an antimony-doped CdTe interlayer incorporated between the p-CdTe and the back contact. Interface resistance has been reduced to about 3.5 ohm-$cm^2$ using this method.[7]

Additionally, a new electroless method for the deposition of p-CdTe films has been developed. The electroless method is based upon short-circuiting the thin-film substrate to an easily oxidizable redox component (e.g., Al, Cd) in the electrolytic bath. This latter method shows promise for routine film deposition, since it avoids the instrumentation for potentiostatic or galvanostatic control.

## FUTURE RESEARCH

The DOE/SERI program is aimed at developing solar cells that can economically displace other forms of energy production. To do this, high-efficiency devices (15% and above) and scalable production deposition techniques are needed. Several new research directions are envisioned (Figure 3):

- Optimization of single-junction cells by investigating $CuInSe_2$, CdTe, and selected alloys of each.
- New research into complex cell geometries such as polycrystalline cascade devices for flat-plate applications.
- The development of alternative heterojunction-partner ("window") materials, including n-ZnO, n-CdO, n-$SnO_2$ and new p-type materials.
- Advanced deposition research, including fundamental studies and studies of area-scalable device deposition techniques such as sputtering or electroplating.

### Single-Junction Cells

The fabrication of 15%-efficient single-junction polycrystalline cells requires significant progress. Both $CuInSe_2$ and CdTe are above 10%, while efforts in other materials ($Zn_3P_2$, $Cu_2Se$) have not been as successful.

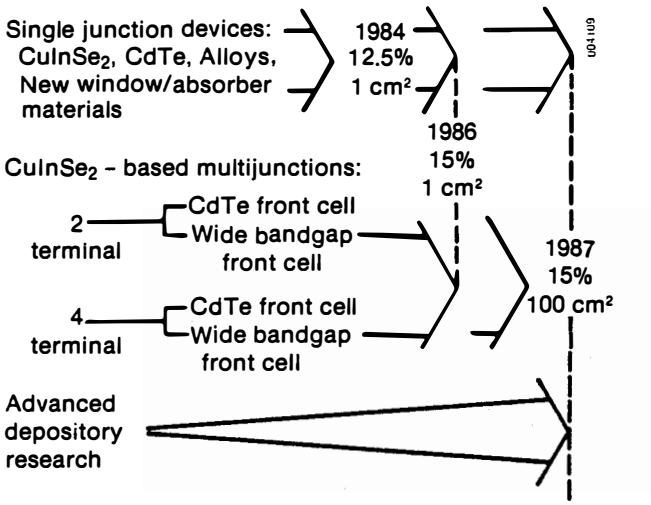


**Figure 3. Opportunities in polycrystalline thin-film solar cells.**

Current program priorities indicate that the bulk of future work should be focused on $CuInSe_2$ and CdTe. Each has special problems as well as areas of likely improvement.

A 15% $CuInSe_2$ device necessitates enhancing fill factor from about 65% to 75% and improving open-circuit voltage ($V_{oc}$) from 0.44 V to about 0.57 V. A major research direction that can be pursued to reach these goals is to produce $CuInSe_2$ alloys that have bandgaps of 1.2-1.4 eV. If materials properties (defects, composition) can be maintained or bettered, a higher $V_{oc}$ should be produced. Given the high absorption coefficient of selected $CuInSe_2$ alloys, current should remain near optimal. With a large bandgap window (perhaps wider than the 2.5-eV gap of CdZnS), current densities could be about 35 $mA/cm^2$ or more.

Progress with CdTe requires a different strategy. Major problems with CdTe are poor contacting and high bulk resistivity. High resistivity is thought to be due to electrically active grain boundaries and low carrier concentrations in the p-type material. Significant progress should be expected if these problems can be solved or reduced. One approach is the introduction of Hg to produce higher p-type conductivity and better contacting. This also lowers the bandgap, forcing a trade-off between reduced $V_{oc}$ and greater current.

## Two-Junction Cells

Multijunction polycrystalline cells have not been adequately investigated. Recently, IEC successfully demonstrated a two-terminal CdTe/$CuInSe_2$ two-cell stack, showing both voltage addition (over 1 V) and several $mA/cm^2$ current density.

Two- and four-terminal stacked polycrystalline cells have trade-offs similar to single-crystal analogs. In two-terminal devices, current must be matched, and deposition processing limitations may be significant. In four-terminal devices, the cells must be separately contacted, inter-cell reflection may need to be reduced, and extra insulating layers may be required.

For four-terminal devices, where current-matching is not necessary, the total achievable efficiency of a two-cell device is the efficiency of the top cell plus a fraction of the normal AM1 efficiency of the bottom cell. To a first approximation (if the spectral response of the bottom cell is flat), the efficiency of the bottom cell is its normal (AM1) efficiency times the fraction of the sunlight that reaches it when it is under the top cell. For instance, at AM1 a $CuInSe_2$ cell beneath a CdTe cell can receive about 35% of the solar photons compared to an uncovered CdS/$CuInSe_2$ cell.* Thus, the achievable efficiency of a CdTe/$CuInSe_2$ four-terminal device would be the AM1 efficiency of the CdTe cell plus 35% of the AM1 efficiency of the $CuInSe_2$ cell. If the bandgap of the top cell could be raised to near 1.75 eV without sacrificing efficiency, about 50% of the light could reach the bottom cell, raising the combination's total efficiency substantially. Figure 4 shows a simple estimate of the practical, achievable efficiencies of CdTe/$CuInSe_2$ four-terminal devices. Note that efficiencies over 15% are within reach given separate cells that are 11% efficient.

Two-terminal devices are limited by the need to equalize the currents generated in each cell to achieve optimal performance. One might consider that allowing 50% of the spectrum to reach the bottom cell would equalize currents, but $CuInSe_2$ is such a good current generator

*This is a conservative estimate based on counting photons between given intervals (the various bandgaps) in the AM1 direct normal solar spectrum. The actual percentage of photons reaching the $CuInSe_2$ cell may vary between 35% and 45% depending on spectral content. At AM1.5, similar "photon counting" suggests a 40% spectral fraction.

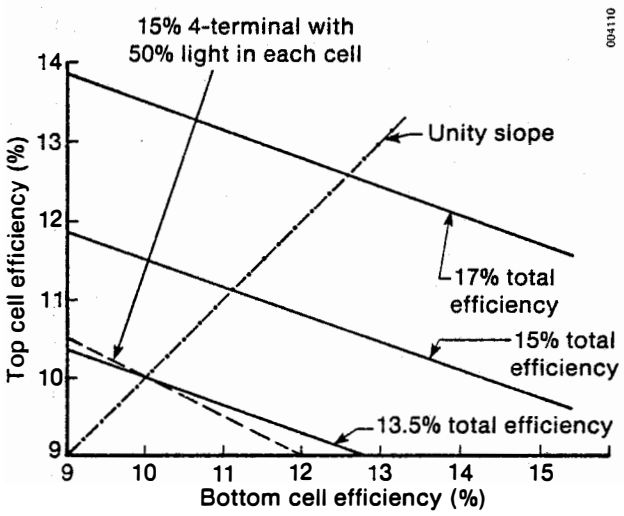


**Figure 4. A simple estimate of the expected efficiencies of four-terminal CdS/CdTe on CdS/$CuInSe_2$ optically stacked cells for various separate cell efficiencies, assuming 35% of the sunlight is reaching the bottom cell. For comparison, the combined efficiency of a pair of cells in which 50% of the light reaches the bottom cell is also given.**

(compared to potential top-cell candidates) that something less than 50% might be appropriate. Even the simplest calculations for expected efficiencies from two-terminal devices are complex in that they depend on the actual quantum efficiencies and physical properties of the separate cells. However, some simple projections can be made for the CdTe/$CuInSe_2$ device. A two-terminal version with $V_{oc}$ about 1.25V, $J_{sc}$ about 16 $mA/cm^2$, and FF about 0.75 would be 15% efficient. All of these parameters seem possible for a cascade cell: (1) 0.8 V and 0.45 V have been achieved individually for CdTe and $CuInSe_2$ cells; (2) at a 40% spectral fraction at least 16 $mA/cm^2$ should be available from $CuInSe_2$; and (3) FF rises in $CuInSe_2$ devices for lower currents.*

In practice, initial cells are likely to be limited by light losses in the top cell. Sub-bandgap light that should theoretically pass through the top cell may still be absorbed, scattered, or reflected. These losses may occur because of (1) below-bandgap absorption in gap states near the band edge, (2) scattering by impurities and grain boundaries, and (3) interfacial reflection. Another required research task is the optimization of a transparent back contact for use in top-cell devices, especially for p-CdTe. Further, optimization of the individual cells for use in cascade devices should provide opportunities for performance enhancements.

**New Window Materials**

Cadmium sulfide has been the most frequently used heterojunction partner with thin-film polycrystalline absorbers. Zinc has been alloyed to CdS to make $Cd_{1-x}Zn_xS$, raising the bandgap. However, resistivity rises rapidly as Zn is added, and materials have not been made with Zn/Cd greater than 0.2 and bandgap over 2.5 eV. There are substantial reasons for developing new n-type heterojunction partners ("window" materials):

1. To optimize single-junction cells by letting the maximum number of solar photons reach the junction, and to better match various absorbers in terms of interface properties, if possible. Figure 5 shows several selected highly conductive, highly transparent window materials currently under investigation. Note the substantial fraction of photons lost by using CdS as the heterojunction partner.

2. To allow the maximum amount of light to reach the high-bandgap top cell of a cascade device, especially where the top cell is the one limiting the current.

3. To match new windows to bottom cells in cascade designs, where bandgaps can be as low as that of the top-cell absorber. For instance, n-CdSe or n-CdTe might be potential bottom-cell windows for p-CuInSe beneath a CdTe top cell.

In addition, it is important to develop p-type windows to match n-type absorbers (a) in four-terminal devices, where the polarity of top and bottom cells do not have to be the same or (b) for single-junction cells, to match with several alternative n-type absorbers that have not yet been optimized.

*Preliminary outdoor test measurements at SERI on (Cd, Zn)S/$CuInSe_2$ cells show an increase in FF at lower light intensities.

The use of p-type window materials would be useful in a number of specific cell designs. For example, n-CdSe might be usable as a good top-cell absorber at 1.7 eV if a high-conductivity, high-bandgap p-type window could be developed. Also, a substantial number of n-type absorbers (n-CdTe, n-$CuInSe_2$) exist and have not been exploited because of a lack of a p-type window. Use of n-type absorbers may be advantageous, because they are frequently easier to contact. For example, the use of a p-type window for n-CdTe would be valuable because (1) contacting would be substantially easier for n-type rather than p-type CdTe and (2) high-quality n-CdTe has already been deposited by large-area methods such as electrodeposition.

## CONCLUSION

Polycrystalline thin-film devices have demonstrated over 10% efficiencies. We see potential optimizations that should bring these single-junction devices to the 15% range. Further, the development of two-junction, stacked solar cells suggests that ultimate, practical efficiencies in the 15%-20% range are possible.

## ACKNOWLEDGEMENTS

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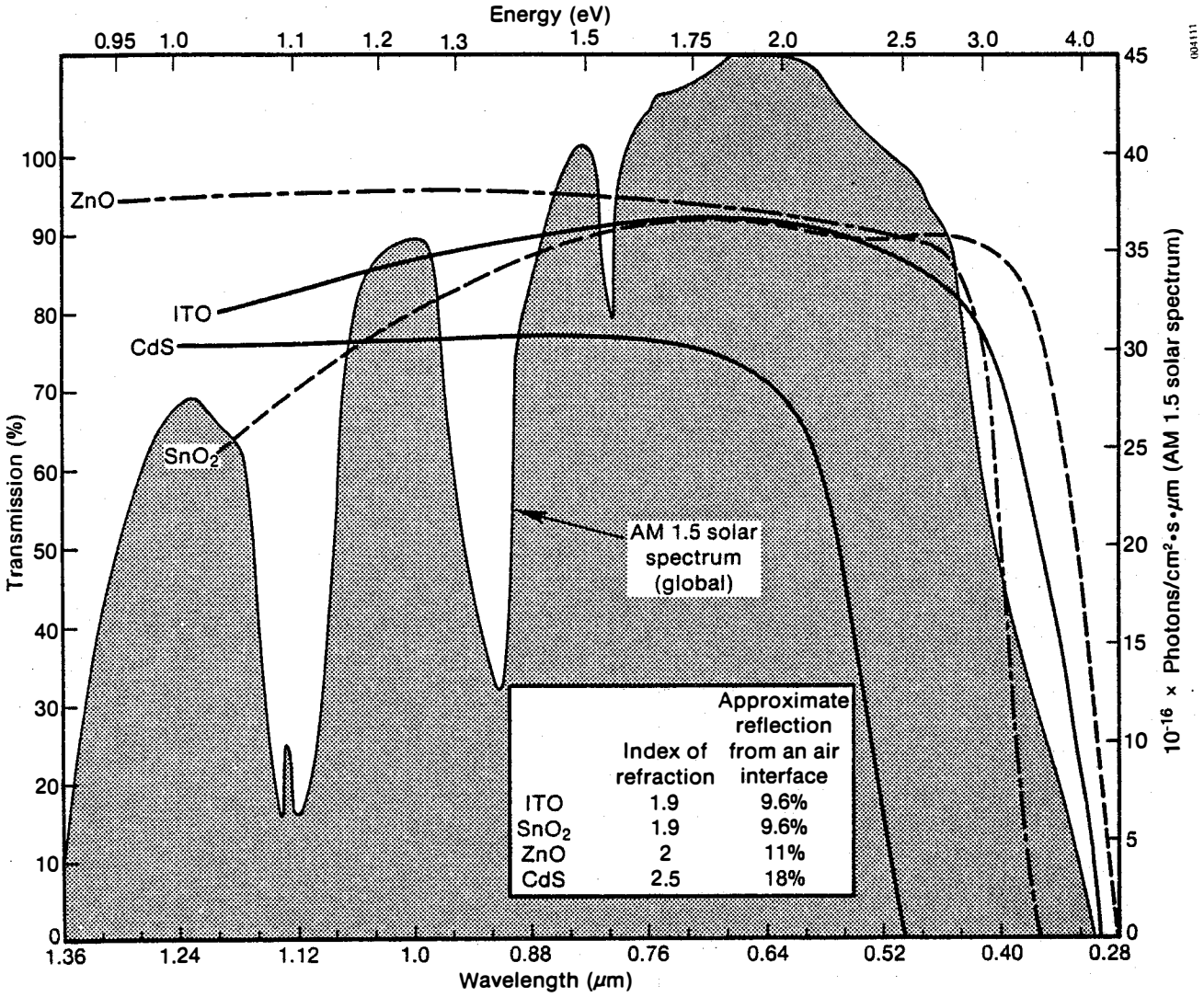


**Figure 5. Window materials: transmission of selected high-bandgap heterojunction partners**

Notes: Transmission curves are smoothed to suppress interference effects, which depend on thickness. Film thicknesses are about (qualitatively) one micron. Carrier concentrations are greater than $10^{20}/cm^3$.

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