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Research on Polycrystalline Thin-Film Materials, Cells, and Modules

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TP-4061

RESEARCH ON POLYCRYSTALLINE THIN-FILM MATERIALS, CELLS, AND MODULES

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ABSTRACT

The U.S. Department of Energy (DOE) supports research activities in polycrystalline thin films through the Polycrystalline Thin-Film Program at the Solar Energy Research Institute (SERI). This program includes research and development (R&D) in both copper indium diselenide and cadmium telluride thin films for photovoltaic applications. The objective of this program is to support R&D of photovoltaic cells and modules that meet the DOE long-term goals of high efficiency (15%-20%), low cost ($50/m^2), and reliability (30-year life time). Research carried out in this area is receiving increased recognition due to important advances in polycrystalline thin-film $CuInSe_2$ and CdTe solar cells and modules. These have become the leading thin-film materials for photovoltaics in terms of efficiency and stability. DOE has recognized this potential through a competitive initiative for the development of $CuInSe_2$ and CdTe modules. This paper focuses on the recent progress and future directions of the Polycrystalline Thin-Film Program and the status of the subcontracted research on these promising photovoltaic materials.

INTRODUCTION

The Polycrystalline Thin-Film Program at SERI is part of the United States National Photovoltaic Program. The objective of this Program is to support research to develop cells and modules that meet DOE's long-term goals by achieving high efficiencies (15%-20%), low cost ($50/m^2), and long time reliability (30 years) [Five Year Research Plan, 1987]. This paper covers the current status of the subcontracted research program in the polycrystalline thin-film area.

The two main materials of interest in this program are copper indium diselenide ($CuInSe_2$) and cadmium telluride (CdTe). Specific research areas include fundamentals, modeling, characterization, measurements, device design, solar cell fabrication, module design and development, module processing, and stability of both $CuInSe_2$ and CdTe [Ullal, 1989].

Both $CuInSe_2$ and CdTe technologies were competing for attention with other materials in the early 1980s. These other materials included ZnP, CuS, CuTe, GaAs and other polycrystalline thin-films. At that time, devices of only around 8%-9% efficiency and about 1 cm^2 were being fabricated from $CuInSe_2$ and CdTe. Cell efficiencies in both technologies rose to about 10%-11% in the mid-1980s. But the potential of these materials remained unrecognized, as there were few increases in performance; processes for making larger areas were in doubt; modules were not being made; and few $CuInSe_2$ or CdTe products were commercially available. Subsequently, both $CuInSe_2$ and CdTe made significant progress (Figure 1), and that progress led to considerable increased attention. One result was an expanded interest in the performance testing and evaluation of these emerging technologies. The Photovoltaics for Utility Scale Applications (PVUSA) project - a joint project between a utility consortium and DOE - has awarded contracts for 20-kW $CuInSe_2$ and CdTe systems to be installed in Davis, California in 1991.

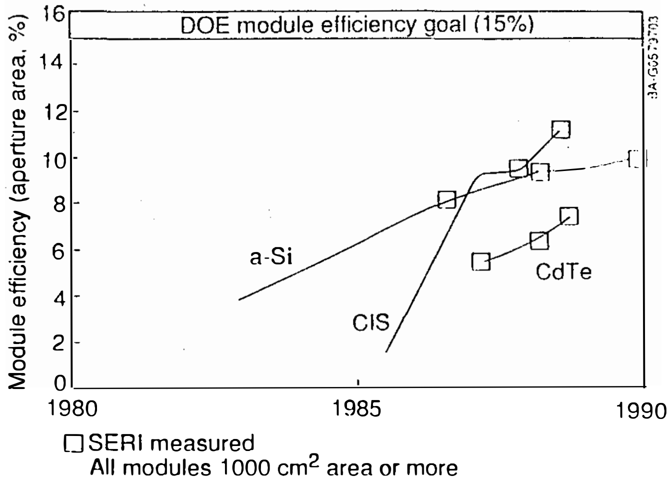

Figure 1. **Progress in thin-film module efficiencies.**

COPPER INDIUM DISELENIDE

During the past decade, progress in $CuInSe_2$ technology has been significant and this photovoltaic material is now considered the leading thin-film candidate in terms of efficiency and long-term reliability [Zweibel and Ullal, 1989]. Theoretical efficiencies for $CuInSe_2$ are as high as 23.5% [Sites, 1988]. Several deposition processes have been used in its fabrication (coevaporation, electrodeposition/selenization, spraying, screening printing, close spaced vapor transport, hybrid evaporation/sputtering, electron-beam-evaporation/selenization, sputtering/selenization, reactive sputtering, sputtering/laser assisted annealing, sputtering/rapid thermal processing, and metal organic chemical vapor deposition) [Zweibel et al., 1990]. These options range from the experimental to those that have been proven at the prototype manufacturing level.

Wagner and co-workers at Bell Labs first fabricated 12% efficient single-crystal $CuInSe_2$ cells in 1974 [Wagner et al., 1974]. Although single crystals are too expensive for practical applications, they did stimulate research in thin-film $CuInSe_2$ devices. Grindle et al. (1980), at the University of Maine were able to make 5% efficient cells by thermally evaporating $CuInSe_2$ on low-cost substrates. This was followed by successful work by Mickelsen and Chen at Boeing Aerospace, who were able to make 10% efficient $CuInSe_2$ cells (with DOE/SERI support) by coevaporation in 1982 [Mickelsen and Chen, 1982].

Following the $CuInSe_2$ research at Boeing, several groups initiated research programs in $CuInSe_2$. Among these were ARCO Solar (now Siemens Solar Industries), the Institute of Energy Conversion (University of Delaware), and SERI. All of these groups were able to quickly reproduce the Boeing results in $CuInSe_2$ cell efficiency by making cells in excess of 10% [Meakin, 1985; Zweibel and Surek, 1985]. ARCO Solar improved their cell design by enhancing the blue response of their devices, improving their cell efficiencies to 12.5% [Potter et al., 1985]. Choudary et al. (1986), proposed replacing the thick CdS layer (2.4 eV) with a "thin CdS" layer (<500 Å) and a wide band gap ZnO (3.2 eV) window layer. This improved the current density by about 15% (6 mA/cm^2). With this novel design (Figure 2), and the addition of a small amount (<10%) of Ga in place of the In, ARCO Solar was able to report achieving a 14.1% active-area efficiency for a 3.5-cm^2 device (Figure 3)[Mitchell and Liu, 1988]. The addition of Ga improved the open-circuit voltage (V_{oc}) of their $CuInSe_2$ devices. During the same period, Boeing also improved their cell performance, reaching 12.5% (1 cm^2 - total area) by adding 27% Ga to make $CuGaInSe_2$ cells (shown in Figure 4)[Devaney et al., 1990]. The V_{oc} of their device was 555 mV due to the increased band gap.

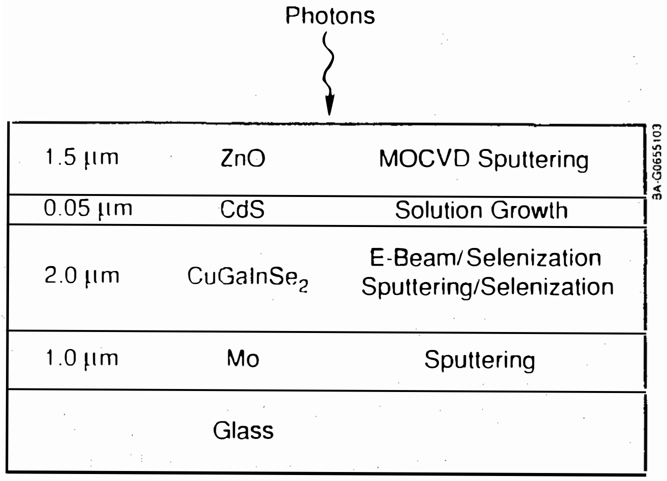

Figure 2. Device structure of a polycrystalline thin-film glass/Mo/$CuInSe_2$/CdS/ZnO solar cell.

International Solar Electric Technology (ISET), another participant in the SERI program, successfully made a 10.9% (active area) device by a low-cost two-stage process [Basol et al.]. In their process, Cu and In layers are deposited by E-beam evaporation on Mo-coated glass substrates. The Cu-In layer is then exposed to a Se-bearing gas that reacts to form high-quality $CuInSe_2$. A thin layer of dip-coated CdS is then deposited on the $CuInSe_2$ for heterojunction formation. Using a similar approach, the Institute of Energy Conversion has also demonstrated a $CuInSe_2$ cell with a short-circuit current (J_{sc}) of 42 mA/cm^2 [Birkmire et al., 1989].

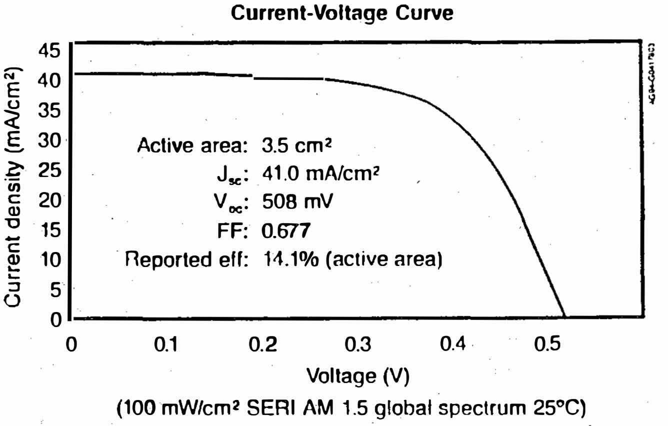

Figure 3. Light I-V characteristics of ARCO Solar's 14.1% efficient $CuInSe_2$ cell.

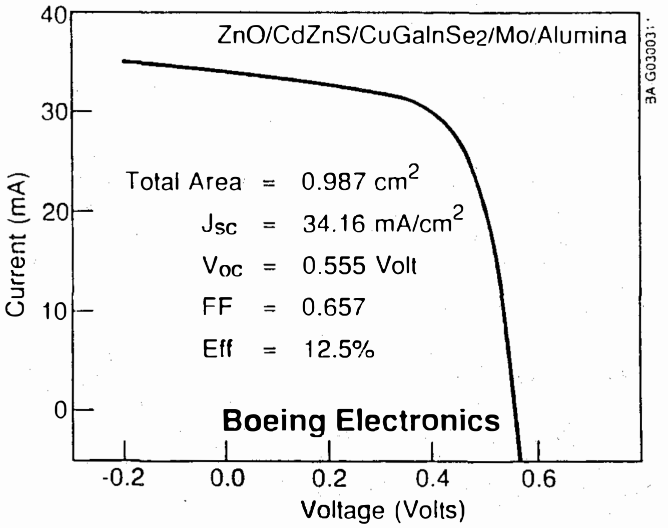

Figure 4. Light I-V characteristics of Boeing's 12.5% efficient $CuInSe_2$ cell.

Some $CuInSe_2$ processes are approaching the stage of scale up for manufacturing. Siemens Solar Industries has successfully scaled up their proprietary process for large-area $CuInSe_2$ module fabrication. A sputtering/selenization method has been described in both U.S. [Love and Choudary, 1984] and European patents and applications [Eberspacher et al., 1989; Ermer and Love, 1989]. Using this process, Ermer and co-workers have reported achieving an 11.1% efficient 938-cm^2 $CuInSe_2$ module [1989, 1990], the most efficient at its size among the various thin films. They have also reported an efficiency in excess of 9.7% for an even larger area (3905 cm^2 - unencapsulated module) with a power output of 37.8 W (Figure 5)[Ermer et al., 1990]. With this success, Siemens Solar Industries was selected as one of the participants in the PVUSA project. They are scheduled to contribute 20 kW of $CuInSe_2$ modules for this project in 1991.

One of the strengths of the $CuInSe_2$ technology is its invulnerability to light-induced instability. Over the last two years, SERI has carried out substantial stability testing on several "zero-th" generation $CuInSe_2$ modules supplied by Siemens Solar Industries. These load and open-circuit outdoor tests have demonstrated excellent reliability (Figure 6) [Mrig and Rummel, 1989]. Excellent outdoor stability is critical to the future acceptance of $CuInSe_2$ modules, and continued tests at SERI should provide important feedback for this requirement.

CADMIUM TELLURIDE

Technical progress in CdTe research has also been significant in the past few years. Based on a band gap of 1.45 eV, which is an optimum match with the solar spectrum, practical CdTe devices could potentially achieve efficiencies over 18%. Theoretical efficiencies are as high as 27.5% [Sites, 1988].

Historically, the CdTe technology was actually under development before a-Si and $CuInSe_2$. It made impressive advances in the 1970s and early 1980s before cell efficiencies temporarily leveled off. Y.S. Tyan and E.A. Perez-Albuerne of Kodak were the first to report reaching 10% efficiency in this thin-film in 1982 [Tyan and Perez-Albuerne, 1982]. Since then, groups such as Ametek, ARCO Solar, BP Solar, Monosolar, ISET, Photon Energy, SOHIO, Southern Methodist University, and Matsushita Battery have all reported efficiencies over 10%.

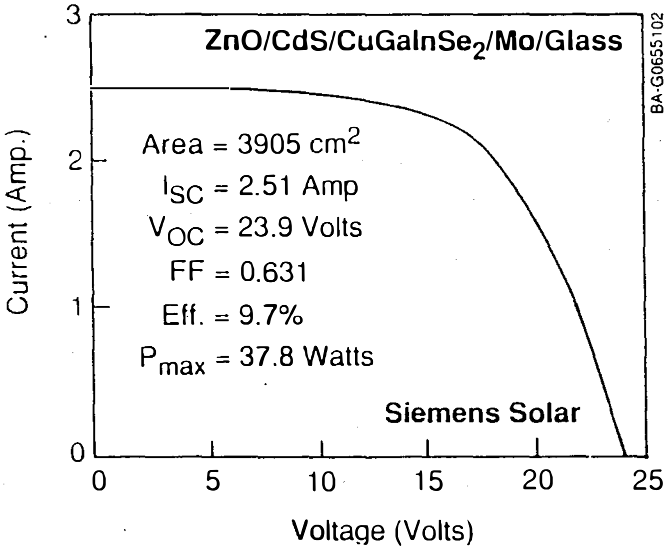

Figure 5. Light I-V characteristics of ARCO Solar's 9.7% efficient, 3905 cm^2, 37.8 W $CuInSe_2$ module.

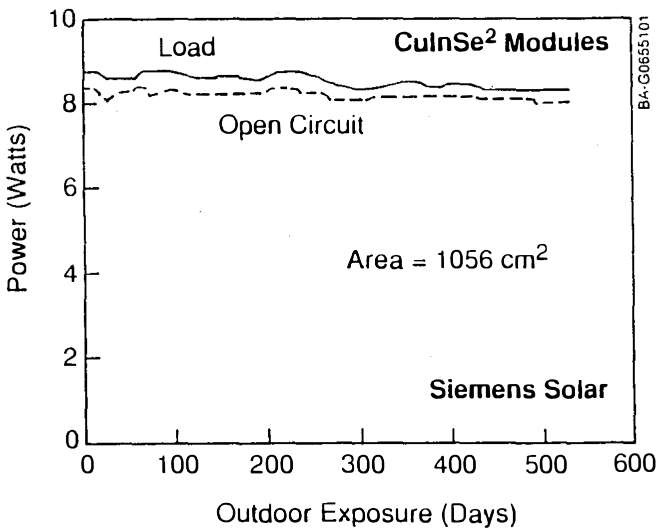

Figure 6. Stability of $CuInSe_2$ modules in outdoor testing, under loading and open-circuit conditions.

A number of methods are used for depositing CdTe thin-films (electrodeposition, spraying, screening printing, close spaced vapor transport, chemical vapor deposition, hot-wall evaporation, ion-assisted evaporation, laser-assisted evaporation, thermal evaporation, sputtering, sputtering/laser assisted annealing, molecular beam epitaxy, and metal organic chemical vapor deposition). At this time, the most promising low-cost approaches are electrodeposition and spraying. Screening printing, a low-cost process, has had limited success due to limitations in module processing.

Electrodeposition is a low-cost, non vacuum method for the deposition of thin-film CdTe. Both Ametek and BP Solar have pursued this approach. BP Solar has reported the fabrication of a 9% efficient 900-cm^2 modules using this deposition technology.

Ametek developed a new CdTe cell design to circumvent one of the key problems encountered in fabricating these thin-film devices - poor contact to the CdTe surface. This has been accomplished through the development of a novel n-i-p cell structure, which is shown in Figure 7. In this design, undoped CdTe acts as an intrinsic layer sandwiched between n-type CdS and p-type ZnTe layers, creating a drift field across the CdTe. (Figure 8 shows the Auger depth profile for this cell structure.) One of the key processing steps in these n-i-p devices is a heat treatment at 400°C for about 20 minutes. This step results in a significant change in film morphology [Ullal, 1988; Zweibel et al., 1988] and is a key to high process yield. During this heat treatment, the polycrystalline grains at the CdS-CdTe interface are believed to coalesce or fuse, thus vastly reducing the density of grain boundary states. Also during this step, a recrystallization occurs over the entire thickness of the CdTe film. Using electrodeposition techniques with this cell design, Ametek has fabricated an 11.2% cell (shown in Figure 9) [Meyers and Liu, 1988]. Ametek recently halted their work in this field and transferred this promising

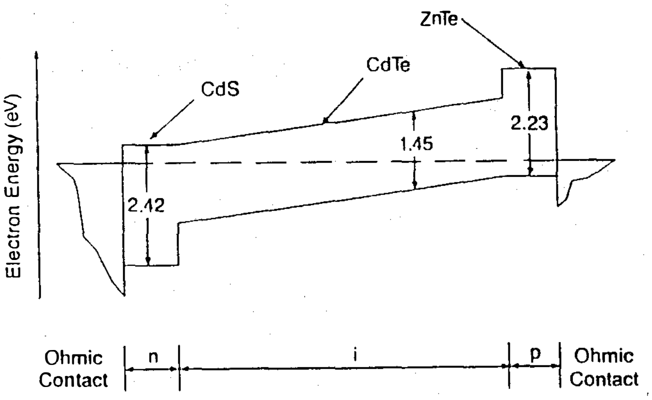

Figure 7. **Simplified band diagram of the Ametek n-i-p thin-film CdTe solar cell.**

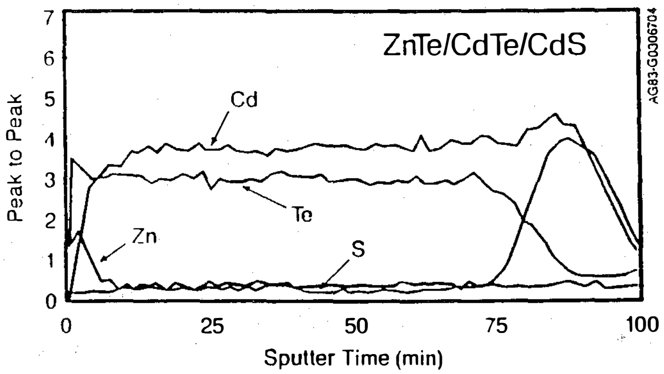

Figure 8. **Auger depth profile of a n-i-p n-CdS/i-CdTe/p-ZnTe polycrystalline thin-film solar cell.**

technology to the Colorado School of Mines. Several potential industry partners are currently investigating opportunities in this technology with this educational institution.

Spray deposition processes have also demonstrated significant promise. Photon Energy has successfully used this technique to fabricate both high-efficiency cells (12.3%, Figure 10) and square-foot modules (7.3%, Figure 11) [Albright et al., 1990]. They have also fabricated the first-ever 4-ft^2 prototype CdTe module. Photon Energy has very aggressive near-term goals to enter photovoltaic production markets. Their cost estimates suggest that they have a potential for being one of the lowest cost module manufacturers in the near future. Due to the simplicity of their process, it does not appear to require the same manufacturing level (i.e. 10 MW/year) to achieve economies of scale. In fact, Photon Energy claims that they will achieve low-cost ($1-$2/W) modules at about 3 MW/year. Photon Energy has been selected to provide 20 kW of CdTe modules for the PVUSA project.

As with CuInSe$_2$, CdTe devices are not vulnerable to light-induced instability. Almost a year of loaded and open-circuit stability testing data from "zero-th" generation CdTe modules at SERI are shown in Figure 12. These tests, carried out on CdTe modules supplied by Photon Energy, have demonstrated excellent reliability [Mrig and Rummel, 1989]. Ametek has also tested their cells and submodules and have reported over 5,000 hours of stability. Continued SERI outdoor testing is expected to provide updated information on the long-term stability of CdTe technologies.

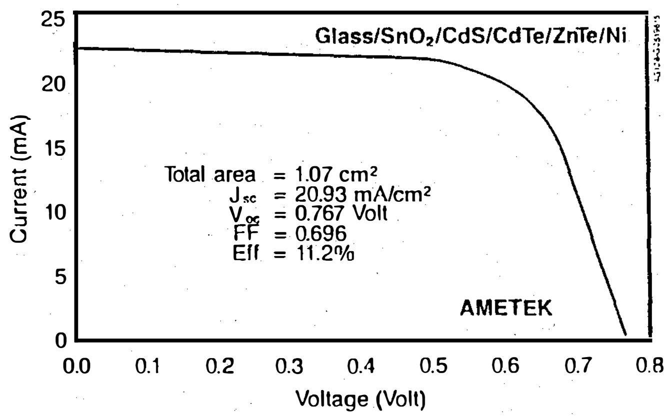

Figure 9. **Light I-V characteristics of Ametek's 11.2% efficient CdTe cell.**

CASCADE (TANDEM) CELLS

Cascade cell configurations with polycrystalline thin-films are also investigated under the SERI program as small efforts at the University of South Florida (USF) and Georgia Institute of Technology (GIT). These research efforts are directed at making "top cells" for these cascade devices with a CuInSe$_2$ bottom cell structure. Band gaps of interest for the top cells in these studies are in the 1.7-2.0 eV range. USF is working on HgZnTe cells deposited by metal organic chemical vapor deposition, where the band gap can be varied through variations in the Hg content. GIT is fabricating "top cells" with band gaps near 1.7 eV based on the addition of Zn to form CdZnTe.

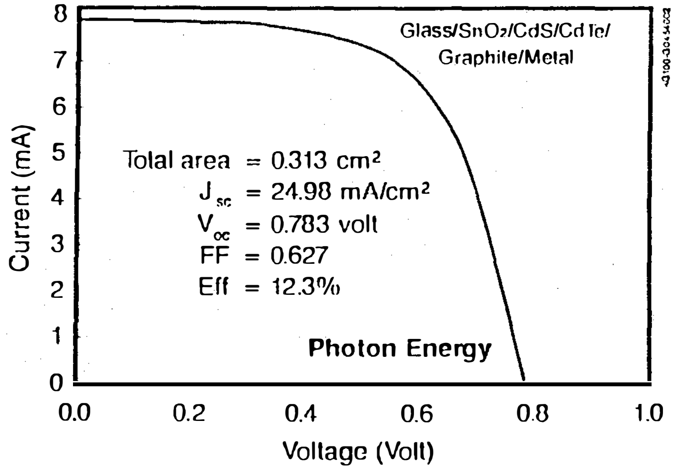

Figure 10. **Light I-V characteristics of Photon Energy's 12.3% efficient CdTe cell.**

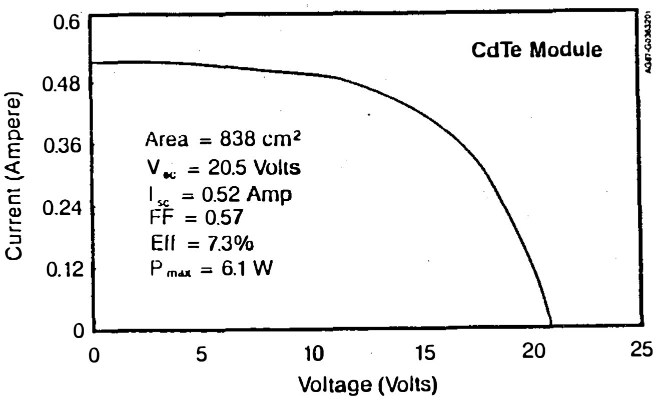

Figure 11. Light I-V characteristics of Photon Energy's 838 cm^2, 6.1 W CdTe module.

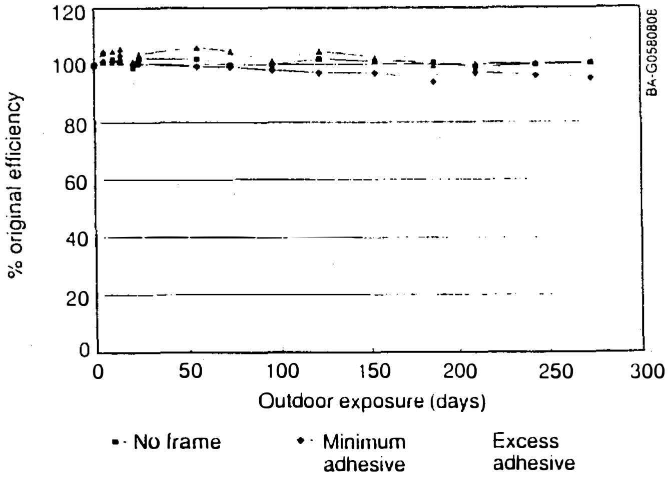

Figure 12. Stability of CdTe modules in outdoor testing, under loading and open-circuit conditions.

THE POLYCRYSTALLINE THIN-FILM MODULE DEVELOPMENT INITIATIVE

SERI maintained a small research program in CuInSe$_2$ and CdTe throughout the 1980s. The funding level for this research was held at about $3 million annually, funding subcontracted activities in both of these technologies, internal R&D in CuInSe$_2$, and various analysis and characterization support activities.

In response to the success of both the CuInSe$_2$ and CdTe technologies, SERI released a request for proposals (RFP) in 1989 for new subcontracts to begin in 1990. This RFP represented a 50% increase in the polycrystalline thin-film effort in 1990. These subcontracts, each 30%-50% cost-shared by industry, include lower-tier university participants. In this way, research awards encourage industry to lead the R&D effort while encouraging collaboration and support between industry and the research efforts at universities. Total funding for the first year of the three-year awards is about $3 million. The objectives of the RFP were to: increase module areas and efficiencies toward the long-term DOE goal of 15%; increase cell efficiencies toward and beyond 15%; assist in the development of new, lower-cost processes, where appropriate; assure that all stability issues are addressed; assist the development of a basic understanding of CuInSe$_2$ and CdTe materials and devices; and assure a U.S. leadership role in commercialization of CuInSe$_2$ and CdTe modules.

Those organizations who were awarded subcontracts under this RFP are listed in Table 1, along with the corresponding focus of each for their subcontracts. Also listed are ongoing participants in the SERI program. Those have been universities, for the most part, and form a significant core program from which the RFP participants and SERI internal researchers will draw support and collaboration.

The Polycrystalline Thin-Film initiative described above was a direct response to the increased success of the CuInSe$_2$ and CdTe technologies. Continuation of this research support will be required to realize their potential.

CONCLUSIONS

Polycrystalline thin-film CuInSe$_2$ and CdTe cells and modules have made rapid advances and are now recognized, in terms of efficiency and stability, as the leading thin-films for photovoltaics. They have attained the highest cell efficiencies (14.1% for CuInSe$_2$ and 12.3% for CdTe), the highest module efficiencies (11.1% - CuInSe$_2$ on 1 ft^2; 9.7% - CuInSe$_2$, 4 ft^2); the best stabilities (CuInSe$_2$ - 500 days, CdTe - 300 days, without degradation), and are made by the lowest cost processes (spraying, electrodeposition, sputtering, and selenization). We can be optimistic about both of these technologies achieving the DOE long-term goal of 15% module efficiency. Both CuInSe$_2$ and CdTe are now moving out of the lab and into demonstrations in the form of PVUSA projects. This is demonstrated through the 20-kW systems of CuInSe$_2$ (to be supplied by Siemens Solar Industries) and CdTe (to be supplied by Photon Energy), soon to be installed in Davis, California, for field testing and evaluation.

ACKNOWLEDGMENT

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Table 1. Participants in the Polycrystalline Thin-Film Program (October 1990)

Research Organization	Topics
Institute of Energy Conversion	Selenized and Evaporated $CuInSe_2$ and CdTe; Cells & Device Modeling
International Solar Electric Technology	E-beam/Sputtering of Cu,In Layers and Selenization; Submodules
University of Illinois	Sputtered/Evaporated $CuInSe_2$; Cells
University of Colorado	Rapid Thermal Processing of Cu/In/Se Films by Electroplating or Sputtering; Cells and Materials Research
California Institute of Technology	Contact Investigation of $CuInSe_2$/Mo Interface; Device Modeling
Solar Energy Research Institute	Growth, Characterization and Device Fabrication of $CuInSe_2$; Cells and Device Modeling
Colorado State University	Characterization and Modeling of CdTe and $CuInSe_2$ Cells; Device Modeling
Purdue University	Modeling of $CuInSe_2$ and CdTe Cells
Photon Energy	Spray Deposition of 900 to 3900 cm^2 CdTe; Modules
University of South Florida	MOCVD and Close Space Sublimation of CdTe & ZnTe; Cells
Georgia Institute of Technology	MOCVD of CdTe & $Cd_{1-x}Zn_xTe$; Cells
University of Toledo	Laser Driven Physical Vapor Deposition of CdTe; Cells & Submodules
Solarex	$CuInSe_2$ Submodule Fabrication of 900 cm^2 by Sputtering
Boeing Aerospace and Electronics	Coevaporation of High-Efficiency $CuGaInSe_2$ Solar Cells
Siemens Solar Industries	$CuInSe_2$ Module Fabrication of 900 to 3900 cm^2

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