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Polycrystalline Thin-Film Technology: Recent Progress in Photovoltaics

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POLYCRYSTALLINE THIN-FILM TECHNOLOGY: RECENT PROGRESS IN PHOTOVOLTAICS

1992 ASME International Solar Energy Conference, April 4-8, 1992

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ABSTRACT

Polycrystalline thin films have made significant technical progress in the past year. Three of these materials that have been studied extensively for photovoltaic (PV) power applications are copper indium diselenide ($CuInSe_2$), cadmium telluride (CdTe), and thin-film polycrystalline silicon (x-Si) deposited on ceramic substrates.

The first of these materials, polycrystalline thin-film $CuInSe_2$, has made some rapid advances in terms of high efficiency and long-term reliability. For $CuInSe_2$ power modules, a world record has been reported on a 0.4-m^2 module with an aperture-area efficiency of 10.4% and a power output of 40.4 W. Additionally, outdoor reliability testing of $CuInSe_2$ modules, under both loaded and open-circuit conditions, has resulted in only minor changes in module performance after more than 1,000 days of continuous exposure to natural sunlight.

The field of polycrystalline thin-film CdTe module research has also resulted in several recent improvements. Module performance has been increased with device areas reaching nearly 900 cm^2. Deposition has been demonstrated by several different techniques, including electrodeposition, spraying, and screen printing. Outdoor reliability testing of CdTe modules was also carried out under both loaded and open-circuit conditions, with more than 600 days of continuous exposure to natural sunlight. These tests were also encouraging and indicated that the modules were stable within measurement error. The highest reported aperture-area module efficiency for CdTe modules is 10%; the semiconductor material was deposited by electrodeposition. Modules fabricated by screen printing have been reported with an efficiency more than 8% for an area of 1200 cm^2. A thin-film CdTe photovoltaic system with a power output of 54 W has been deployed in Saudi Arabia for water pumping.

The Module Development Initiative has made significant progress in support of the Polycrystalline Thin-Film Program in the past year, and results are presented in this paper. The major U.S. industrial participants in the $CuInSe_2$ portion of this initiative include Siemens Solar Industries, Solarex Corporation, International Solar Electric Technology (ISET), Martin Marietta, and Boeing Aerospace and Electronics. Photon Energy and Solar Cells Inc. are involved in the CdTe research under this initiative. A program is also being addressed for the aggressive development of the thin silicon film on ceramic substrates by AstroPower. The major players in polycrystalline thin films outside of the United States are BP Solar in the United Kingdom, Matsushita Battery in Japan, and Microchemistry in Finland. All three of these groups are developing thin-film CdTe power modules. Additionally, as part of the Photovoltaics for Utility-Scale Applications (PVUSA) project, several emerging thin-film technologies are planned for field testing at Davis, California, during 1992. This paper will summarize the current status and technical results under the subcontracted research in the PVUSA program. In addition, the NREL-funded competition for university research subcontracts will be discussed.

INTRODUCTION

The Polycrystalline Thin-Film Program at the National Renewable Energy Laboratory (NREL), formerly the Solar Energy Research Institute, is part of the United States Department of Energy (DOE) National Photovoltaics Program. The objective of this program is to support research and development on 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) [National Photovoltaic Five Year Research Plan, 1987].

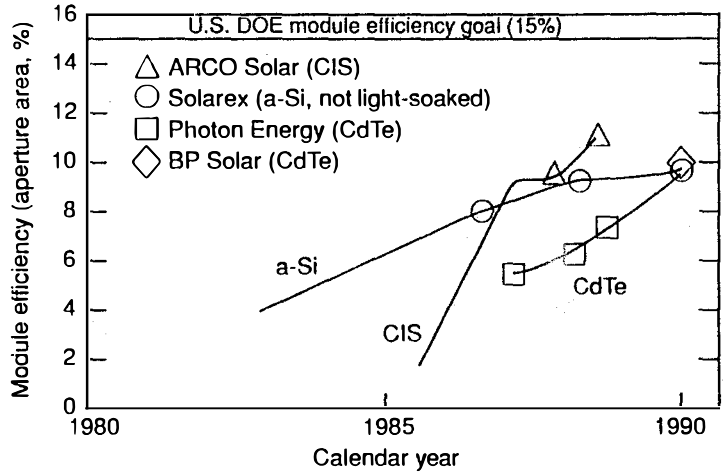
During the last year, researchers in $CuInSe_2$, CdTe, and thin-film x-Si technologies have made significant progress toward achieving the DOE goals. This progress has included (i) the achievement of record thin-film power module (4 ft^2) efficiencies of nearly 10% for $CuInSe_2$, (ii) a breakthrough CdTe cell efficiency of 13.4%, (iii) an accelerated growth of the U.S. industrial infrastructure supporting $CuInSe_2$, CdTe, and thin-film x-Si, and (iv) the continued success of multiyear outdoor stability testing on prototype $CuInSe_2$ and CdTe modules. Many of the technical goals (module efficiency, stability, and cost) associated with attaining truly low-cost PV (under 6 cents/kWh system cost) appear to be achievable by these technologies given existing research trends. Figure 1 shows the relative progress of the thin films toward high-efficiency, 1-ft^2 modules. With theoretical efficiencies of 23.5% for $CuInSe_2$ and 27.5% for CdTe, and ultimate *practical* efficiencies of 16%-18% [Sites, 1988], it is expected that similar progress will continue for these polycrystalline thin films. Correspondingly, module stability also appears to be excellent (see results below), with two Siemens Solar Industries $CuInSe_2$ modules measured at 99.8% and 97.5% of their original efficiencies after almost 1,000 days of outdoor testing at NREL.

U.S. PV INFRASTRUCTURE

Although NREL has maintained a small research program in $CuInSe_2$ and CdTe throughout the 1980s, the funding level was held at only about $3 million per year. This support was allocated to: (i) subcontracted activities in both $CuInSe_2$ and CdTe, (ii) internal Research and Development (R&D) in $CuInSe_2$, and (iii) various analysis and characterization support activities.

Technical achievements in the late 1980s, such as the 11% efficient, 1-ft^2 $CuInSe_2$ module [Mitchell et al., 1988] made by ARCO Solar (now Siemens Solar Industries), led to increased interest in polycrystalline thin films. One of the critical barriers to the progress of these technologies was the narrowness of their industrial base. At that time, the only industrial group making $CuInSe_2$ modules was ARCO Solar, and only a few other corporate entities were capable of making efficient $CuInSe_2$ cells (i.e., ISET and Boeing Aerospace). The state of the CdTe industry was very similar: Ametek had decided to end its participation in the CdTe technology, and the only other viable industrial group making modules, Photon Energy, remained very small and resource-limited. Unlike $CuInSe_2$ however, CdTe was supported by non-U.S. companies of significance: BP Solar and Matsushita. For thin-film x-Si, there was only one small company with a serious commitment, AstroPower.

In 1989, with the success of both the $CuInSe_2$ and CdTe technologies, NREL released a Request for Proposals (RFP) for new subcontracts research to begin in 1990. This RFP represented a 50% increase in the annual polycrystalline thin-film effort. The resulting subcontracts under this program, each 30%-50% cost-shared by industry, included lower-tier subcontracts with university



Notes: All modules 1000 cm^2 area or more; a-Si efficiencies prior to light-induced degradation

Figure 1. Relative progress of thin films toward high efficiency.

participants. This encouraged industrial participants to lead the R&D effort and encouraged collaboration and support between industry and the research efforts at universities. Total funding for the first year of the three-year awards was about $3 million. The objectives of the RFP were to (i) increase module areas and efficiencies toward the long-term DOE goal of 15%; (ii) increase cell efficiencies toward and beyond 15%; (iii) assist in the development of new, lower-cost processes, where appropriate; (iv) assure that all stability issues are addressed; (v) assist the development of a basic understanding of $CuInSe_2$ and CdTe materials and devices; and (vi) assure a U.S. leadership role in commercialization of $CuInSe_2$ and CdTe modules. Additionally, the stabilization and strengthening of the corporate infrastructure of these technologies was a major focus of the DOE/NREL increase in funding for polycrystalline thin films.

Those industrial "partners" who were awarded subcontracts under this RFP are listed in Table 1, along with the corresponding focus of their subcontracts. Listed in Table 2 are the ongoing participants in the NREL program, primarily universities. These organizations form a significant portion of the polycrystalline thin-film core program from which the RFP participants and NREL internal researchers have drawn support and collaboration. The subcontractors listed in Table 1 are all working to develop successful prototype thin-film modules. Issues of module design

Table 1. Module Development Goals For Industrial Partners (November 1991)

	$CuInSe_2$	
ISET	Cu & In Sputtering/Selenization	11% efficiency - 900 cm^2
Siemens Solar	Cu & In Sputtering/Selenization	12.5% efficiency - 3900 cm^2
Solarex	Elemental Cu, In, & Se Sputtering	12% efficiency - 900 cm^2
	CdTe	
Photon Energy	Spraying	12.5% efficiency - 3900 cm^2
Solar Cells Inc.	Close-Spaced Sublimation	10% efficiency - 7200 cm^2
	Silicon Films	
AstroPower	Thin-Film x-Si on Ceramic Substrates	12% efficiency - 1200 cm^2

Table 2. Polycrystalline Thin-Film Program Support Participants (November 1991)

Organization	Topics	Output
Institute of Energy Conversion	Selenized & Evaporated $CuInSe_2$ & CdTe	Cells & Device Modeling
University of Illinois	Sputtered/Evaporated $CuInSe_2$	Cells
University of Colorado	Rapid Thermal Processing of Cu/In/Se Films by Electroplating or Sputtering	Cells & Materials Research
California Institute of Technology	Contact Investigation of $CuInSe_2$/Mo Interface	Device Modeling
National Renewable Energy Lab	Growth, Characterization, & Fabrication of $CuInSe_2$ Cells	Cells & Device Modeling
Colorado State University	Characterization & Modeling of CdTe & $CuInSe_2$ Cells	Device Modeling
Purdue University	Modeling of $CuInSe_2$ & CdTe Cells	Device Modeling
University of South Florida	MOCVD & CSS of CdTe & ZnTe	Cells
Georgia Institute of Technology	MOCVD of CdTe & $Cd_{1-x}Zn_xTe$	Cells

and efficiency and prototype processes are the main focus. The annual NREL/DOE funding of these subcontracts is about $4.2 million, with another $1.9 million cost-shared contribution from the subcontractors. Thus, the total investment over the three-year initiative will be about $18 million. In addition to the subcontracts of Table 1, the Polycrystalline Thin-Film Program has one other industrial subcontract with Martin Marietta. The goals of this effort are to investigate deposition by a rotating cylindrical magnetron of copper and indium (the precursors for the selenization of $CuInSe_2$) and cadmium and tellurium (for CdTe), as well as some work in CdTe electrodeposition.

Until this fiscal year, the program was not able to address the infrastructural weakness of polycrystalline thin-film research at universities. However, an RFP for subcontracted research is now under way to support university research in both CdTe and $CuInSe_2$. It is expected that about 6 subcontracts will be awarded under this RFP with each subcontract funded at around $100K/year. In addition, the program has three ongoing university subcontracts (totaling about $740K) at Colorado State University and the Institute of Energy Conversion (IEC) at the University of Delaware, and the University of Toledo. The breakdown of funds in Fiscal Year (FY) 1992 for the Polycrystalline Thin-Film Program research will be about 75% for industry and 25% for universities. In addition, NREL funds an in-house research effort of about $3,300K annually.

COPPER INDIUM DISELENIDE

During the past decade, progress in $CuInSe_2$ technology has been significant. This PV material is now considered the leading thin-film candidate in terms of efficiency and long-term reliability [Zweibel and Ullal, 1989]. Theoretical efficiencies for $CuInSe_2$ have been reported as high as 23.5% [Sites, 1988], resulting in this material being viewed as having significant potential as a low-cost, thin-film PV material.

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 [MOCVD]) [Zweibel et al., 1990]. These options range from the experimental to those that have been proven at the prototype manufacturing level.

$CuInSe_2$ Power Module - Siemens Solar Industries

Of the numerous advances in the last year, perhaps the most significant was the achievement of a near-10%, encapsulated, $CuInSe_2$ power module by Siemens Solar Industries. Because thin-film modules come in many sizes, it is somewhat difficult to develop perspective about the relative importance of their efficiencies and areas. However, in overall technical progress, it is probable that the Siemens Solar $CuInSe_2$ power module is the most significant achievement in thin-film PV. No other module of this size has even come close in efficiency. Figure 2 shows the NREL-measured I-V curve of this module with an aperture-area (3883 cm^2) efficiency of 9.7%, P_{max} = 37.7 W; V_{oc} = 24 V; J_{sc} = 2.44 A; and FF = 64.4%.

Improved $CuInSe_2$/Mo Adhesion - ISET

ISET has been successful in fabricating $CuInSe_2$ cells by selenization of Cu/In precursor films with a total-area efficiency of 11.5%. Analysis by Sites (1991) has suggested that the junction properties of ISET $CuInSe_2$ cells (in terms of diode quality factors and recombination currents) are as good or better than those of any other $CuInSe_2$ devices available. Their properties are comparable to those needed for $CuInSe_2$ cells to achieve a 15% efficiency. Like other organizations involved in $CuInSe_2$ fabrication, especially those that use selenization, ISET has encountered adhesion problems at its $CuInSe_2$/Mo interface. In July 1991, ISET was granted a U.S. Patent [Basol and Kapur, 1990] on an innovative solution to the adhesion problem: the deposition of a very thin tellurium layer (10-500 Å) between the Mo and the $CuInSe_2$. ISET has claimed in its patent that this layer reduces adhesion problems significantly and also results in a device with improved performance. By adding the Te layer, ISET has been able to increase the flexibility with which it adds subsequent Cu and In (and Ga, if desired) layers. For example, ISET researchers prefer to deposit In and then Cu, the reverse of the conventional order.

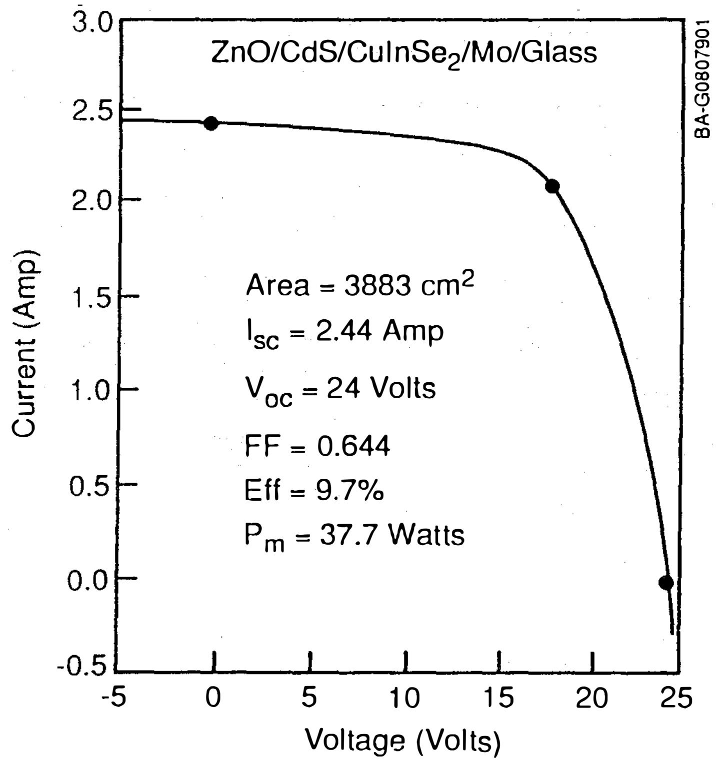


Figure 2. NREL-measured I-V curve of ARCO Solar's 9.7%-efficient, 37.7-W, $CuInSe_2$ module, showing its aperture area (3883 cm^2).

ISET believes that the presence of a Te layer allows for better deposition of the In layer. Without this layer, the In tends to "ball up" and causes a degradation of uniformity in the final film. The progress at ISET in finding a workable solution to the $CuInSe_2$/Mo adhesion problem, and the subsequent improvement in its cell efficiencies, a significant contribution to the progress of $CuInSe_2$ technology.

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, this material has theoretical efficiencies as high as 27.5% [Sites, 1988]. Given this significant potential, it is reasonable to expect that CdTe devices may achieve practical efficiencies of 18%.

Several 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 MOCVD). 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 its limitations in module processing.

Advances in CdTe Cell Efficiencies - the Chu Breakthrough

Several record CdTe cell efficiencies were reported during the last year [Sites, 1991]. In some cases, however, these reports were not validated by independent measurements. In specific cases, for instance, questions concerning excessive current densities suggest that the reported efficiencies were inaccurate. On the other hand, NREL did measure cells from Photon Energy and then from T.L. Chu (a lower-tier subcontractor of Photon Energy at the University of South Florida) that surpassed previously verified results.

In May 1991, Photon Energy produced a small-area (0.3 cm^2) cell with exceptionally high current density (26.2 mA/cm^2). This current density, which is about 90% of the theoretical maximum for CdTe, was achieved by using an optically thin CdS layer. This glass/TCO/CdS/CdTe cell had a quantum efficiency of 80% at the 400-nm wavelength (Figure 3), a strong indication that much of the improved current was coming from a reduction in the absorption of photons with energies above the CdS band gap.

Three weeks later, Chu substantially surpassed the Photon Energy result. Chu achieved a 13.4%-efficient CdS/CdTe cell with an area of 1.2 cm^2, V_{oc} = 840 mV, and FF = 72.6%. This was reached by again achieving very high voltages and fill factors (shown in Figure 4). Another Chu cell having a 12.6% efficiency demonstrated the highest known CdTe fill factor to date, 74.6%.

These results were achieved using an innovative deposition process for the solution growth of CdS with both Chu's own CdTe (made by close-spaced sublimation) and with Photon Energy's CdTe materials (demonstrating up to 890 mV). The consistency of these high voltages suggests that much of the improved efficiency can be attributed to the solution-grown CdS rather than to the CdTe.

However, there is still room for improvement in these cells because the current densities of the Chu devices were moderate - about 20-22 mA/cm^2. The nature of the results of this CdS layer research suggests that further progress on these devices will be relatively easy, especially because the solution-grown CdS layer is not yet optimized for optical thinness. It is expected that this optimization and the achievement of cell efficiencies over 15% will be a focus in upcoming months.

OUTDOOR TESTS AND STABILITY ISSUES

Although intrinsic device stability appears good with all polycrystalline thin films, there are issues at the module level that

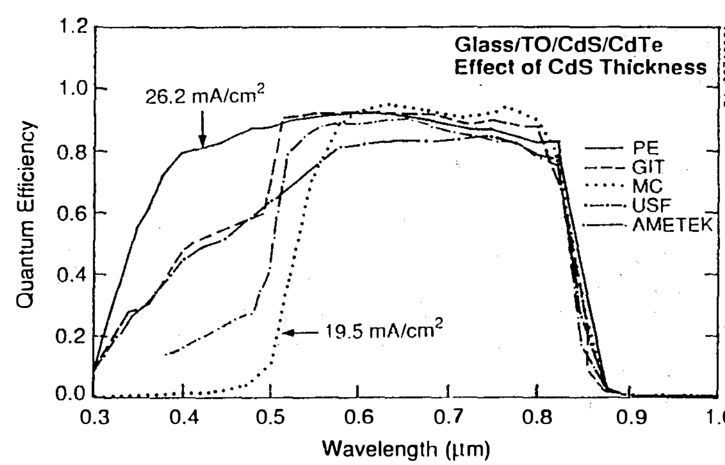


Figure 3. Quantum Efficiency curve for Photon Energy small-area (0.3 cm^2) cell having a 26.2-mA/cm^2 current density.

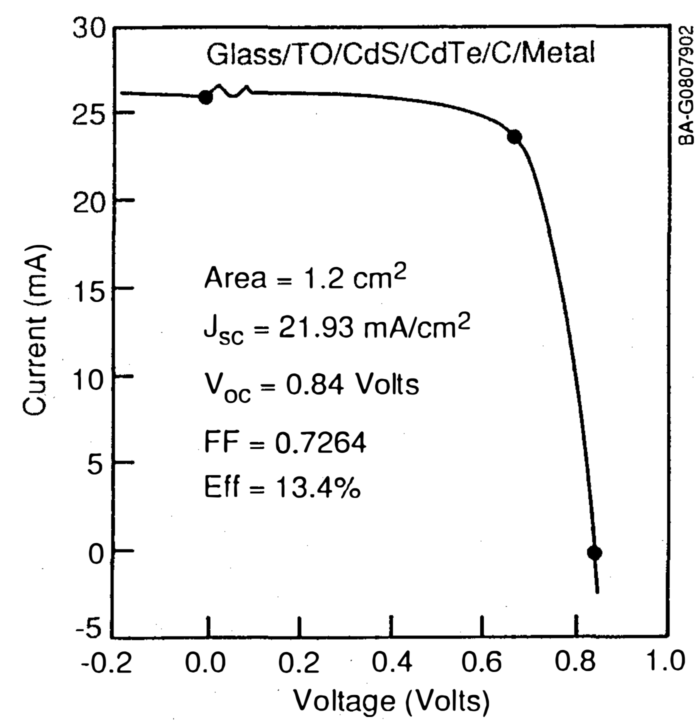


Figure 4. Light I-V characteristics of Chu's 13.4%-efficient CdTe cell (Area = 1.2 cm^2, V_{oc} = 840 mV, and FF = 72.6%).

require attention as these technologies move into the marketplace. For instance, $CuInSe_2$ and CdTe are sensitive to chemicals used in module fabrication. Therefore, various approaches need to be developed for minimizing chemical interactions. Additionally, CdTe cells and modules are sensitive to water vapor, requiring careful encapsulation. Issues with $CuInSe_2$ modules tend to be associated with specific layers, such as undesirable MoSe layers at the back contact that impede current flow, and defects and adhesion issues associated with Mo and $CuInSe_2$ interfaces. Therefore, the achievement of a 30-year life for these technologies appears to require attention to specific processing details and careful module design and encapsulation.

To this date, the results of almost three years of outdoor tests on Siemens Solar $CuInSe_2$ modules are very promising. NREL has conducted these tests on only two of these $CuInSe_2$ modules; however, the most recent measurements indicate that both of these modules are producing power at 99.8% and 97.5% of their original levels (Figure 5). The aperture-area efficiencies of the modules were near 8% with measurements taken at *near*-standard conditions. This accounts for some of the scatter in the data. Those data appear to indicate that the *intrinsic* stability of these modules is not an issue for $CuInSe_2$. Any remaining stability issues associated with this material are expected to be process- and design-specific.

The outdoor test results for Photon Energy CdTe modules are also promising; however, some issues remain. As can be seen from Figure 6 (outdoor tests, *near* standard conditions), several Photon Energy modules have shown good outdoor stability over reasonably long periods. However, other modules (not shown) have exhibited some slight degradation. Photon Energy believes that the deficiency in these modules is associated with encapsulation and edge-sealing techniques. Photon Energy modules are expected to improve as encapsulation issues are dealt with successfully.

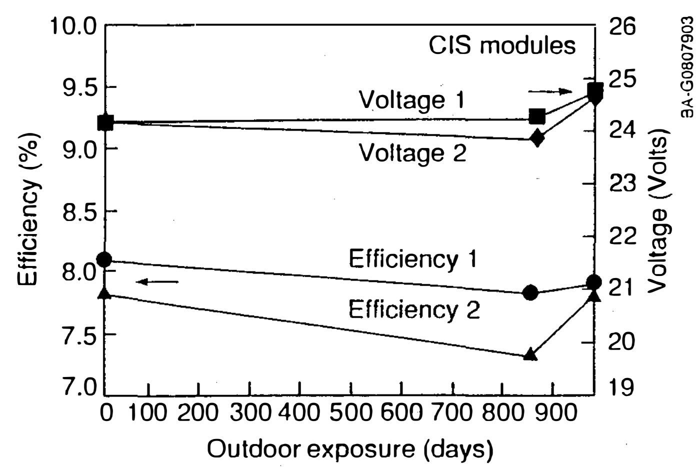


Figure 5. Results of three years of NREL outdoor tests on Siemens Solar Industries $CuInSe_2$ modules, showing power production at 99.8% and 97.5% of original levels.

OTHER HIGHLIGHTS

Other key advances sponsored by the NREL program during the last year include progress at Georgia Institute of Technology (GIT) (MOCVD CdTe cells), the University of South Florida (USF) (MOCVD CdTe cells), AstroPower (thin-film x-Si cells), IEC ($CuInSe_2$ devices by H_2Se selenization), and in-house NREL research (precursors/selenization for $CuInSe_2$).

GIT and USF have both improved the performance of their thin-film CdTe devices deposited by MOCVD techniques. Cell efficiencies in the 10%-11% range have been reached when subjected to $CdCl_2$ chemical treatments and heat treatments of 420°C for 20 minutes. Further improvements are expected from reducing the thickness of the CdS film and improving the contacts to the high-resistivity CdTe absorber layers.

AstroPower's "Product I" is currently in the manufacturing stage with a capacity of 0.5 MW for FY 1991. AstroPower's plans include the expansion of this effort to 2-3 MW in 1992. In the subcontracted portion of the DOE/NREL Polycrystalline Thin-Film program, AstroPower is developing elements of its "Product II" and

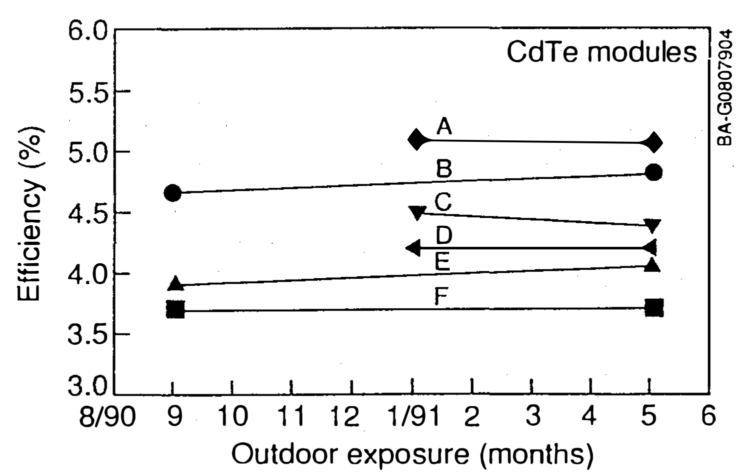


Figure 6. Results of NREL outdoor tests on Photon Energy CdTe modules, showing good stability over reasonably long periods.

"Product III" technology. Product II essentially uses a metallurgical barrier deposited on a low-cost conducting ceramic substrate. This barrier also serves as an optical reflector to enhance the short-circuit current and improve the cell efficiency. In addition, the thickness of the Si-film in Product II will be reduced to less than 50 microns. Product III incorporates a monolithically integrated 1,200-cm^2 module fabricated on an insulating ceramic substrate.

IEC has fabricated 10%-efficient small-area $CuInSe_2$ devices by the selenization method using H_2Se. IEC was also successful in fabricating 7%-efficient small-area devices by depositing Cu, In, and Se layers and heat-treating them in an atmosphere of excess Se. IEC also carried out modeling studies in support of both the $CuInSe_2$ and CdTe research. From this effort IEC has concluded that (i) the V_{oc} of $CuInSe_2$ solar cells cannot be solely described by a Shockley-Read-Hall (SRH) recombination mechanism, (ii) CdTe solar cells operate as p-n heterojunctions with their current dominated by SRH recombination in the junction region, and (iii) ZnTe:Cu contacts in an n-i-p CdTe structure are more stable than either Au or Cu-Au contacts.

The NREL in-house emphasis over the last year has been on the understanding of the phase behavior and microstructure of Cu/In precursors used in selenization processes for $CuInSe_2$. In these processes, the fabrication of a precursor structure (containing mainly Cu and In deposited onto Mo-coated substrates) precedes the actual selenization step. A correlation of the Cu/In precursor microstructure with the post-selenized $CuInSe_2$ film and device characteristics should identify techniques for enhancing the quality of $CuInSe_2$ films for optimum device efficiency. Additional research has also been carried out on the compositional and substrate temperature dependencies of co-evaporated $CuInSe_2$ films. The goal of this work is to optimize the fabrication process by deliberate modification and better control to achieve higher-quality $CuInSe_2$ films.

TRANSITION TO MANUFACTURING

Three of the participants in the NREL Polycrystalline Thin-Films Project (Siemens Solar Industries, Photon Energy, and AstroPower) were winners in the most recent (1990) PVUSA Emerging Technology competitions. However, the developmental work that exists between the achievement of an excellent prototype module and an actual manufactured module is significant. The three technologies in question ($CuInSe_2$, CdTe, and thin-film x-Si) have reached an excellent level of laboratory success; however, the transition to true production has only recently begun. NREL is supporting these technologies during this transition, and we recognize that delays and unexpected problems are natural. This delay in the maturing of the technology has resulted in a postponement in delivery of the 20-kW systems required from both of these organizations under this program.

The problems associated with the final maturity of polycrystalline thin-film module products require that specific problems be addressed by each manufacturer. These include (i) identifying all process steps for cost-effective module production, (ii) completing the design of successful encapsulation schemes, (iii) confirming module reliability with outdoor and accelerated tests, and (iv) developing a market acceptance for these untried polycrystalline modules. In addition, each organization will have to fully address environment, safety, and health (ES&H) issues such as plant safety, plant waste disposal, and related matters. In parallel, the technologies must still progress toward higher efficiencies (10%-15% modules) if they are to make the kind of impact on global energy production that the DOE/NREL program believes is possible.

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 13.4% for CdTe), the highest module efficiencies (11.1%-$CuInSe_2$ on 1 ft^2; 9.7%-$CuInSe_2$, 4 ft^2); the best stabilities ($CuInSe_2$ - three years without degradation, CdTe - two years without degradation), and are made by the lowest-cost processes (spraying, electrodeposition, sputtering, and selenization). Both $CuInSe_2$ and CdTe are now moving out of the lab and into module demonstrations in the form of PVUSA projects, and we can be optimistic about both of them achieving the DOE long-term goal of 15% efficiency for their modules. However, the strengths of the polycrystalline thin-film materials remain somewhat obscure because progress in the laboratory and at the prototype module level still needs to be translated into success with manufactured products. In order to achieve a production yield for 10%-15% efficient modules, further required research should be focused on identifying all required process steps, successful encapsulation methods, market acceptance schemes, and all ES&H issues.

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