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Progress in High-Performance PV: Polycrystalline Thin-Film Tandem Cells

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PROGRESS IN HIGH-PERFORMANCE PV: POLYCRYSTALLINE THIN-FILM TANDEM CELLS

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Abstract: The High-Performance Photovoltaic (HiPerf PV) Project was initiated by the U.S. Department of Energy to substantially increase the viability of PV for cost-competitive applications. The goal is that PV will contribute significantly to the U.S. and world energy supply and environmental enhancement in the 21st century. The HiPerf PV Project aims at exploring the ultimate performance limits of existing PV technologies, approximately doubling their sunlight-to-electricity conversion efficiencies during its course, to accelerate and enhance their impact in the marketplace. To accomplish this, the National Center for Photovoltaics (NCPV) directs in-house and subcontracted research in high-performance polycrystalline thin-film and multijunction concentrator devices. This paper will describe progress of the subcontractor and in-house R&D on critical pathways for a PV technology having a high potential to reach cost-competitiveness goals: 25%-efficient, low-cost polycrystalline thin-film tandems for large-area, flat-plate modules.
Keywords: Polycrystalline – 1: Thin-film – 2: High-Efficiency – 3

1. INTODUCTION

The HiPerf PV Project aims at exploring the ultimate performance limits of existing PV technologies, approximately doubling their sunlight-to-electricity conversion efficiencies during its course, to accelerate and enhance their impact in the marketplace. Along with other criteria for success (module manufacturing cost and reliability, which are central to other components of the DOE National PV Program), module sunlight-to-electricity conversion efficiency is a key parameter driving the economics of PV-generated electricity. Simply put, raising sunlight-to-electricity conversion efficiency reduces cost per unit of electrical output. The HiPerf PV Project directs Federal resources toward some of the most critical barriers to the widespread use of photovoltaics for energy-significant applications. This addresses one of the highest-priority goals for applied research in the U.S. Photovoltaics Industry Roadmap [1]: "developing high-efficiency, low-cost materials and devices."

This paper will describe progress on exploring critical pathways for a PV technology having a high potential to reach cost-competitiveness goals: low-cost polycrystalline thin-film tandems for large-area, flat-plate modules (Figure 1).

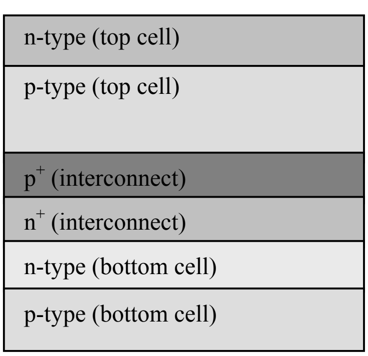

Figure 1: Tandem schematic (monolithic structure)

The concept was introduced to increase efficiency, but its potential for reducing cost also became apparent many years ago [2]. This technology has the potential to reach the installed system cost goal of about \$1/Wp with continued progress in efficiency, reliability, and manufacturing cost.

2.0 APPROACH

2.1 Project Description

The NCPV at the National Renewable Energy Laboratory (NREL) directs in-house and subcontracted research in high- performance polycrystalline thin-film and multi-junction concentrator devices. During the project period and pushing the research toward established goals extensive collaboration should produce significant contributions to the entire PV industry. A roadmap of the High Performance PV Project approach is shown for approximately the next decade (Figure 2).

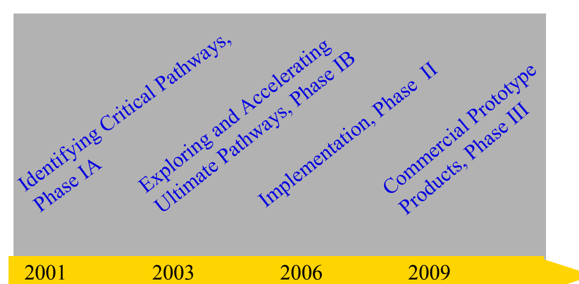

Figure 2: Roadmap of the High Performance PV Project

The first phase of the project is critical because it provides a means to identify, explore, and accelerate the most promising paths for implementation, followed by commercial prototype products. These latter efforts constitute the second and third phases of this planned research program. The first of a two-part phase, "Identifying Critical Pathways," investigated a wide range of complex issues in both the polycrystalline thin-film tandems and III-V multi-junction concentrators. These investigations provided initial modeling and baseline experiments for several advanced concepts to clarify some of the challenges and identify critical paths for the longer-term development and application of high-performance PV technologies. The current Phase IB, "High Performance PV—Exploring and Accelerating Ultimate Pathways," is a continuation of Phase I and addresses exploring and accelerating ultimate pathways to reach the project's long-term goals. It is thought that several promising approaches will be explored in each category during this phase, which will lead to Phase II, "Implementation of Pathways." Seven companies and universities were competitively selected and have received awards for the HiPerf PV Phase IB (see Table I).

Table I: Phase IB, "Exploring and Accelerating Ultimate Pathways" Subcontractor Awards

Subcontractor	**Title**
Georgia Institute of Technology	Thin-Film Si Bottom Cells for Tandem Device Structures
University of Delaware (IEC)	High-Performance PV-Polycrystalline Thin-Film tandem Cells
University of Toledo	Sputtered II-VI Alloys and Structures for Tandem PV
University of Florida	Identification of Critical Paths in the Manufacturing of Low-Cost High-Efficiency CGS/CIS Two-Junction Tandem Cells
University of Oregon	Identifying the Electronic Properties Relevant to Improving the Performance of High Band-gap Copper Based I-III-VI_2 chalcopyrite thin film PV devices
Oregon State	Novel Materials Development for Polycrystalline Thin-Film Solar Cells
Light Spin Technologies	Novel Polycrystalline Thin-Film Solar Cells

3.0 PROJECT GOALS AND R&D FOCUS

3.1 Goals

To address HiPerf PV R&D long-term goals of bringing polycrystalline thin-film tandem cells (combining high-band gap and low-band gap single-junctions) and modules toward 25% and 20% efficiencies, the project investigates a wide range of complex issues and provides initial modeling and baseline experiments of several advanced concepts. Recent work by Coutts *et al*. [3] modeling state-of-the-art thin-film devices has provided critical guidance for the project. A near-term milestone chart of the R&D thin-film polycrystalline tandems is shown by year and will be described here (see Table II). Throughout the projects term, there will be opportunities to reach established program goals by both disruptive technology advances and/or multiple incremental improvements.

Table II: Near-term Milestones, High-Performance PV Project, Polycrystalline Thin-Film Tandems

Date	**Milestone**
2002	10%-Efficient, 1.5 $<E_{gap}<$1.8 eV Cell (Completed)
2003	Compare Device Design in Terms of Monolithic/Mechanical Structure (Completed)
2004	Assess Research on Exploring Pathways
2005	12%-Efficient Polycrystalline Thin-Film Tandem
2006	15%-Efficient PolycrystallineThin-Film Tandem

3.2 R&D Focus

The wide-bandgap top cell material of the tandem is critical; it is anticipated that two-thirds of the tandem cell efficiency originates here. Therefore, R&D is focused on a top cell, which is integrated with the bottom cell via an interconnect junction. Transmission through the top cell is a challenge, requiring an optical band-gap (E_g) in the range of $1.5<E_g<1.8$ eV, and minimal sub-bandgap absorption. High-bandgap alloys based on I-III-VI_2 and II-VI compounds and other novel materials for the top cell are being investigated. Low-bandgap CIS and its alloys, thin Si, and other novel approaches are being considered for the bottom cell.

Integration of the thin-film interconnect with the top cell optically, electrically, and with an eye toward process compatibility is being investigated; this includes the role of defects and how they affect the transport properties of this junction, as well as diffusion of impurities into the bulk. Transparent conducting oxide's (TCO) are able to form a one-sided p/n+ interconnect (shorting/tunneling junction) between the TCO and a non-degenerate p-type absorber [4], playing a strong role in the tandem cell.

The design in terms of a monolithic or mechanical stack is primarily determined by the choice of the high-bandgap top cell material. There are pros and cons to both approaches. For example, with the monolithic approach, only one thick TCO, one grid, and one anti-reflection coating (ARC) would be needed. However, current-matching and temperature-stability issues arise, as well as the necessity of a close thickness tolerance with the tunnel junction. Whereas the mechanical stack design may appear at first glance much simpler than the monolithic design, other issues are involved. For example, more materials (ARCs, TCOs, and glass) would be needed for the overall structure. Regardless of the designs, both structures are being pursued during the project.

4.0 PROGRESS IN HIGH-BANDGAP MATERIALS

Several high-bandgap top cell materials have been identified under the project, but they still need further exploration (see Table III). The table lists several materials that have been highly successful in terms of the operating parameters for the tandem structure. Several of the materials listed are described below.

The Polycrystalline Thin Film PV Group at NREL has demonstrated that a surface-modified CGS cell exhibits the following NREL-confirmed device operating parameters: V_{oc} = 0.823 volts, J_{sc} = 18.61 mA/cm^2, fill factor = 66.8%, and total-area-efficiency = 10.2%. CGS is a candidate top cell absorber material for thin film tandem devices. Its bandgap is ideal at 1.68 eV. This particular device had a bandgap of 1.64 eV. Improving CGS device efficiency has proven to be a challenge over the past several years. The recent understanding of the differences in structural and electronic properties between $CuIn(Ga)Se_2$ and CGS thin films and devices has led to varying the growth process in a way that is likely to make the CGS surface region similar to that of CI(G)S and to minimize defects in the material.
This change led to a gain in the current density of about 3.7 mA/cm^2 versus the previous record cell.

The University of Delaware, Institute of Energy Conversion (IEC) is investigating $Cu(InGa)(SeS)_2$ films and $Cd_{1-x}Zn_xTe$ films of varying compositions and on specific substrates for the top cell of the tandem [5]. Recently $Cu(InGa)(SeS)_2$ films were deposited as part of a set of experiments in which the relative concentrations of sulfur to selenium and gallium to indium were varied to give a fixed bandgap of 1.5 eV. The highest cell efficiencies in these experiments were achieved using absorber layers that contained no sulfur and with a relative gallium composition ratio of Ga/(In+Ga) = 0.75 ± 0.03. This corresponds to an optical bandgap of 1.5 ± 0.03 eV. Solar cells were fabricated at IEC using the structure glass/ Mo/$Cu(InGa)(SeS)_2$/CdS/ZnO/ITO with Ni-Al collection grids and total area, defined by mechanical scribes, of 0.47 – 0.51 cm^2. Current-voltage measurements were completed at NREL on devices from two different depositions. The best cell from one run had efficiency = 10.9 % with V_{OC} = 0.826 V, J_{SC} = 20.4 mA/cm^2, and fill factor = 64.5 %. From the other run, the best cell had efficiency = 10.9 % with V_{OC} = 0.836 V, J_{SC} = 20.4 mA/cm^2, and fill factor = 64.

The Polycrystalline Thin Film PV Group at NREL modified CdS/CdTe devices to assist in early identification of limitations to high optical transparency of high-performance top cells [6]. Initial attempts have produced devices demonstrating ~9 % efficiency. First Solar material was used, and the devices were contacted with ZnTe:Cu/Ti. A project goal is to identify issues limiting Near-Infrared (NIR) optical transmission of the top device, spectrophotometry studies were initiated on similar transparent devices (i.e., incorporating ITO + metal grid contacts), as well as spectroscopic ellipsometry studies of component layers. Preliminary results from these studies have shown that the 9% efficiency cell can be maintained for cells with NIR transparency of ~25%. Although this result establishes a good initial baseline, it also indicates that NIR transmission must be improved considerably for high-efficiency two-junction operation. Numerical modeling studies have shown that much of this NIR absorption can be ascribed to the specific type of TCO incorporated into the First Solar starting materials used in these studies.

Recently, University of South Florida demonstrated an 80% transparent CdSe device (Figure 3). The CdSe was deposited on a transparent conductor and a ZnSe/Cu contact was added. The highest efficiency reported was 17 mA/cm^2. This is believed to be a record for a thin-film solid-state CdSe device. The challenge for these materials will be to develop p-type window layer contacts with the requisite optical and electronic properties to qualify for use in transparent, high-bandgap II-VI devices.

Table III: High-Bandgap structures and their operating parameters (NREL verified)

Organization	**High Band-Gap Top Cell Structure (eV)**	**Voc (V)**	**Jsc(mA/ cm2)**	**Fill Factor (%)**	**Efficiency (%)**	**comments**
NREL	Glass/Mo/CGS/CdS/ZnO (1.64 eV)	.823	18.61	66.8	10.2	Surface modified CGS
NREL	Glass/Mo/CGS/CdS/ZnO (1.68 eV)	.905	14.8	70.9	9.53	
NREL	Glass/SnO_2/CGS/CdS/ZnO (1.68 eV)	.864	15.36	51.25	6.8	60%-70% transmission
University of Delaware (IEC)	Glass/Mo/Cu(InGa)Se_2/ CdS / ZnO/ITO (1.5 eV)	.826	20.4	64.5	10.9	
NREL	Glass/SnO_2/CdS/CdTe/Zn Te:Cu (1.5 eV)	.785	17.5	64.9	8.93	First Solar CSS CdTe, 25% transmission
NREL	Glass/$CdSnO_4$/$ZnSnO_4$ /CdS/CdTe/CuTe (1.5 eV)	.776	24	68.21	12.7	50% transmission
University of South Florida	Glass/TC/CdSe (1.7eV)	.250	14	.53	1.9	80% transmission

*Not NREL verified

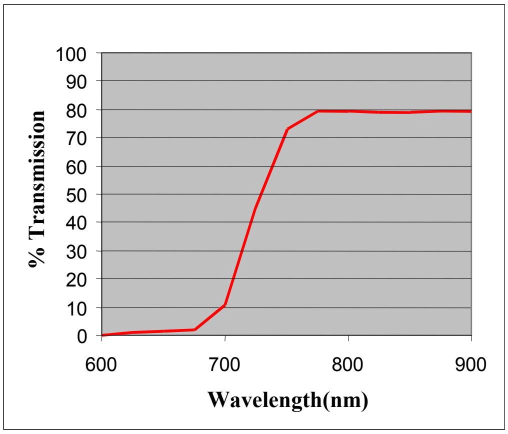

Figure 3: University of South Florida device transparency for 2-micron glass/TC/CdSe

5.0 PROGRESS IN TANDEM SOLAR CELLS

Several polycrystalline thin-film tandem structures have been developed and demonstrated under the HiPerf project, but they still need further exploration (Table IV). The table lists several structures, both mechanical and monolithic designs, in terms of the operating parameters. Several of these novel devices will be described below.

The NREL High-Performance Polycrystalline Thin-Film Tandem Group demonstrated a prototype monolithic tandem using Si and CdS/$CuGaSe_2$ (CGS) as the bottom and top cell absorbers, respectively [6]. Figure 4 shows a schematic of the device. The bottom cell is an NREL-grown, crystalline Si cell, with a diffused Al back contact and a p/n^+ junction. The CGS top cell was grown by elemental evaporation following the NREL-patented 3-stage process. The interconnect junction consists of an n^+ indium tin oxide layer on n+ c-Si, which had been shown earlier to produce a reasonably good transparent back contact to CGS[7]. The non-official measurement shows excellent voltage addition of about 1.3 V, a short circuit current of 9 ma/cm2, and a fill factor of 43%, with an overall efficiency of about 5.1%. Witness CGS cells grown on Mo showed Voc of about 730 mV, despite the non-ideal Cu/Ga ratio of ~0.99.

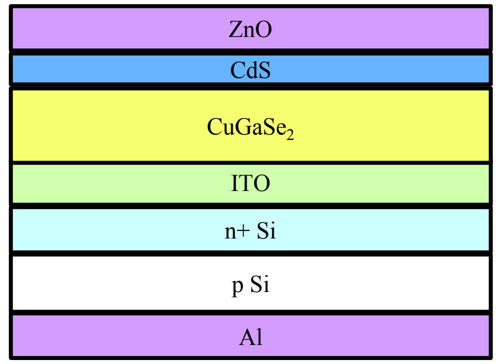

Figure 4: Schematic of CGS, c-Si tandem

Quantum efficiency results reveal that the top cell is still far from ideal.

The NREL High-Performance Polycrystalline Thin-Film Tandem Group demonstrated a prototype mechanical stack tandem using CGS and CIS for the top cell absorber and bottom cell, respectively [7]. The CGS top cell was grown by elemental evaporationfollowing the NREL-patented 3-stage process. The transmission data are shown to be 70%-80% for SnO_2 /CGS/CdS/ZnO device (Figure 5). This was a mechanical stack with an official NREL measurement of 9.7% efficiency and a Voc of 1.29 V with an AR coating.

University of Toledo (UT) under a High-Performance PV subcontract, "Polycrystalline Thin-Film Tandem. Photovoltaic Cells," demonstrated a two-terminal CdTe-HgCdTe cell (Figure 6). The tandem cell was fabricated in the superstrate structure starting with a sputter-deposited top cell [SnO_2:F/CdS/CdTe (1.8-2.0 μm)] with UT's standard processing and $CdCl_2$ treatment condition P-type ZnTe:N and n-type ZnO: Al layers were used to

Table IV: Tandem structures and their parameters (NREL verified)

Organization	Tandem Structure	Voc (V)	Jsc (mA/cm 2)	Fill Factor (%)	Efficiency (%)	Comments
NREL	Top cell: glass/ SnO_2//CGS/CdS/ZnO Bottom cell: glass/Mo/CIS/CdS/ZnO Mechanical stack	.864 .456 1.29	15.36 12.46	51.25 69.17	6.8 3.9 9.7	4-terminal device
NREL	Monolithic structure: ZnO/CdS/CGS/ITO/n+Si/pSi/ Al	1.32	9	43	5.1	
University of Delaware (IEC)*	Monolithic structure: ZnO/ITO/CdS/Cu(InGa)Se_2 /ZnO/CdS/CIS/Mo/glass	.688	10.4	52.8	3.8	
University of Toledo	Monolithic structure: SnO_2:F/CdS/CdTe/ZnTe:N/ZnO:Al/CdS /HgCdTe	.960	2	62	1.2	

*Not NREL verified

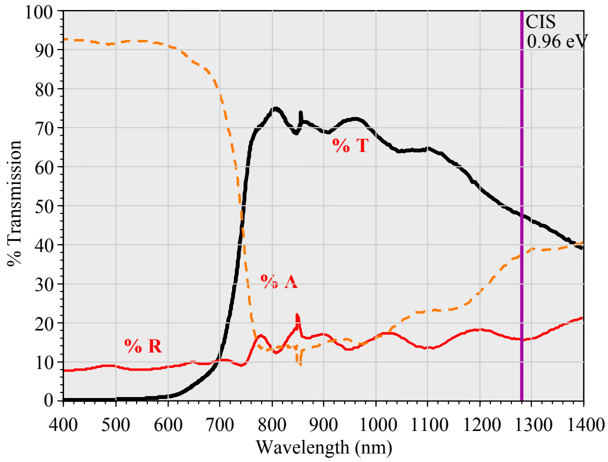

Figure 5: Transmission for SnO_2/CGS/CdS/ZnO Device

form a recombination junction [8,9]. The bottom cell, CdS/HgCdTe (1.2-1.5µm), was also sputter-deposited with a bandgap of HgCdTe of about 1.0-1.15 eV. A second $CdCl_2$ treatment was given before finishing the cells with UT's standard evaporated Cu/Au back contact.

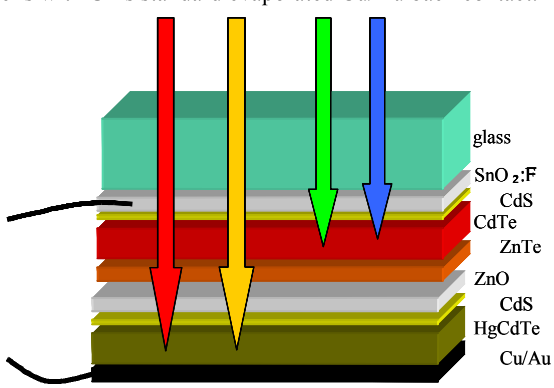

Figure 6: Schematic of the University of Toledo SnO2:Fe/CdS/CdTe/ZnTe:N/ZnO:Al/CdS/HgCdTe Device

Researchers from UT believe that better deposition control and the optimization of post-deposition treatment of HgCdTe films will lead to better cell performance for both single-junction HgCdTe cells and tandem cells. The cell area was 0.06 cm^2. The I-V measurement shows V_{oc} of 960mV, and J_{sc} of 1.92 mA/cm^2 and efficiency of 1.2%. The QE characterization shows that the photocurrent was limited by the HgCdTe cell current due to the bandgap being too large. This is an excellent first demonstration of a two-terminal CdTe-HgCdTe.

Georgia Institute of Technology, under a High Performance PV subcontract, is investigating the compatibility of c-Si-based bottom cells with CGS top cell materials. As a component of a thin-film tandem solar cell, the Si bottom cell is an excellent candidate for surviving the fabrication of the top cell. This will be demonstrated by fabricating monolithic tandem structures. To date, it has been found that the junction quality of the Si bottom cell remains intact after CIGS deposition. Cu(InGa)Se_2 reduces the long wavelength QE response of the Si cell, which is likely due to sub-bandgap absorption in the Cu(InGa)Se_2 film.

6.0 CONCLUSIONS

Phase IB, "Exploring and Accelerating Ultimate Pathways," of the HiPerf PV Project is underway with in-house and subcontracted research efforts in high-performance polycrystalline thin-film tandems.

Our investigations have led to several wide-bandgap materials that are now world-record efficiencies. These high-bandgap materials are beginning to meet requirements in terms of Voc, Jsc and transmission for polycrystalline thin-film tandems.

Both monolithic and mechanical tandems have been developed under the project, they are listed in Table IV. These devices used high band-gap alloys based on I-III-VI_2 and II-VI compounds. The developments under the High-Performance PV Project reported here are progress towards achieving long-term DOE-goals [1]. The project is focused to assure that tandem thin-film polycrystalline modules reach efficiency levels consistent with cost-competitive goals.

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14. ABSTRACT *(Maximum 200 Words)*

The High-Performance Photovoltaic (HiPerf PV) Project was initiated by the U.S. Department of Energy to substantially increase the viability of PV for cost-competitive applications. The goal is that PV will contribute significantly to the U.S. and world energy supply and environmental enhancement in the 21st century. The HiPerf PV Project aims at exploring the ultimate performance limits of existing PV technologies, approximately doubling their sunlight-to-electricity conversion efficiencies during its course, to accelerate and enhance their impact in the marketplace. To accomplish this, the National Center for Photovoltaics (NCPV) directs in-house and subcontracted research in high-performance polycrystalline thin-film and multijunction concentrator devices. This paper will describe progress of the subcontractor and in-house R&D on critical pathways for a PV technology having a high potential to reach cost-competitiveness goals: 25%-efficient, low-cost polycrystalline thin-film tandems for large-area, flat-plate modules.

15. SUBJECT TERMS
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