

Status of High Performance PV: Polycrystalline Thin-Film Tandems

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STATUS OF HIGH PERFORMANCE PV: POLYCRYSTALLINE THIN-FILM TANDEM

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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 photovoltaics (PV) for cost-competitive applications so that PV can contribute significantly to our energy supply and our environment. 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. This work includes bringing thin-film cells and modules toward 25% and 20% efficiencies, respectively; and developing multijunction concentrator cells and modules able to convert more than one-third of the sun's energy to electricity (i.e., 33% efficiency). This paper will address recent accomplishments of the NREL in-house research effort involving polycrystalline thin-film tandems, as well as the research efforts under way in the subcontracted area.

INTRODUCTION

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. 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.

PROJECT GOALS AND OBJECTIVES

The NCPV at NREL directs in-house and sub-contracted research in high-performance polycrystalline thin-film and multijunction concentrator devices. During the project period, extensive collaboration and the work performed to push the research toward established goals should produce significant contributions to the entire PV industry. A roadmap of the High-Performance PV Project

approach is shown covering approximately the next decade (Fig. 1).

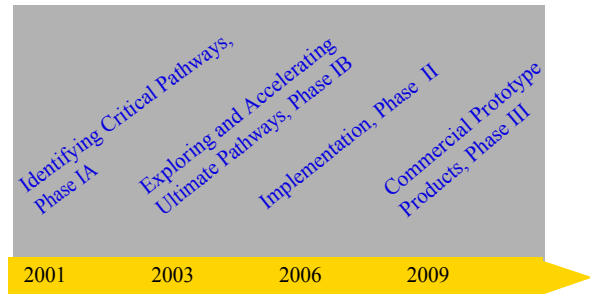


Fig. 1. Roadmap of the High-Performance PV Project.

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 multijunctions 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 1).

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-Bandgap 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

Table 1. Phase IB, “Exploring and Accelerating Ultimate Pathways” Subcontractor Awards

PROJECT GOALS AND R&D FOCUS

Goals

To address HiPerf PV R&D long-term goals of bringing polycrystalline thin-film tandem cells (combining high-bandgap and low-bandgap single junctions) and modules toward 25% and 20% efficiencies, respectively, 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 2). Throughout the project’s term, there will be opportunities to reach established program goals by both disruptive technology advances and/or multiple incremental improvements.

Date	Milestone
2002	10%-Efficient, 1.5 <E _g <1.8 eV Cell (Completed)
2003	Compare Device Design in Terms of Monolithic/Mechanical Structure (Completed)
2004	Assess Research on Exploring Pathways
2005	14%-Efficient Polycrystalline Thin-Film Tandem
2006	15%-Efficient PolycrystallineThin-Film Tandem

Table 2. Near-term Milestones, High-Performance PV Project, Polycrystalline Thin-Film Tandems.

R&D Focus and Advancements

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 bandgap (E_g) in the range of $1.5 < E_g < 1.8$ eV and minimal sub-bandgap absorption. Process compatibility to maintain the performance of the bottom cell is essential.

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 oxides (TCOs) 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.

HIGH-BANDGAP MATERIALS

Several high-bandgap top cell materials have been identified under the project, but they still need further exploration (see Table 3). The table lists several materials that have been highly successful in terms of the operating parameters for the tandem structure.

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/cm², fill factor = 66.8%, and total-area efficiency = 10.2%. CGS is a candidate top cell absorber material. 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)Se₂ 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 Cl(G)S and to minimize defects in the material.

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]. 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 cm². 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/cm², 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/cm², and fill factor = 64. Understanding the growth mechanisms of Cu(InGa)(SeS)₂ films, particularly the incorporation of the chalcogen (S, Se) species, is fundamental to their application in wide-bandgap solar cells. Using deposition conditions that yield uniform through-film Cu(InGa)Se₂ composition and high-efficiency devices, the growth of Cu(InGa)(SeS)₂ or CuIn(SeS)₂ has been shown to form a bi-layer film comprising two different chalcopyrite compositions. A layer at the back of the film is relatively rich in S, and a layer near the surface is S-poor relative to the flux during growth. This results in poor device behavior for the mixed chalcogen (S+Se) films. Although the growth mechanisms that produce this behavior are still under study, this phenomenon appears to be limited by growth kinetics rather than fundamental thermodynamics.

Oregon State has worked on several new compositions, and films are being developed for use as conductive windows, tunnel junctions, and absorbers in tandem solar cells. The material BaCuTeF has been identified as a new p-type semiconductor with a bandgap of 2.3 eV.

Measurements on pressed pellets reveal an electrical conductivity of 8 S/cm and a Seebeck coefficient of +25 μV/K; the temperature dependence of the electrical conductivity is consistent with degenerate behavior. Thin films of the material are being processed via pulsed laser deposition; these films should interface well with CdTe. In the family of compounds MCuSnQ₄ (M = Ba, Sr; Q = S, Se), compositions have been systematically varied to identify the material BaCuSnSe₂S₂ (direct E_g = 1.77 eV) as a candidate absorber for top cell applications

PROGRESS IN TANDEM SOLAR CELLS

Several polycrystalline thin-film tandem designs have been developed and demonstrated under the HiPerf project, but they still need further exploration. Table 4 lists several of these structures, which include both mechanical and monolithic designs, in terms of the operating parameters. Several of these novel devices will be described below.

Researchers at the Georgia Institute of Technology are looking to make Si-based tandem cells compatible to the processing conditions required to fabricate CuInSe₂-based solar cells using conventional crystalline Si cell structures. They have shown that c-Si devices remain intact after exposure to Cu(InGa)Se₂ growth. However, Ag diffusion from the metallization of the Si cell was observed on the Cu(InGa)Se₂ surface. This increased the resistance of the Si cell and reduced its fill factor.

Organization	High Band-Gap Top Cell Structure (eV)	Voc (V)	Jsc (mA/cm ²)	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/Cd ₂ SnO ₄ /ZnSnO _x /nano-CdS:O/CdTe/Cu _x Te(1.5 eV)	.806	24.97	69.22	13.9	60%-40% transmission

Table 3. High-Bandgap Top Cell Structures and their Operating Parameters (NREL verified).

Organization	Tandem Structure	Voc (V)	Jsc (mA/cm ²)	Fill Factor (%)	Efficiency (%)	Comment
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	Topcell: Glass/Cd ₂ SnO ₄ /ZnSnO _x /nano-CdS:O/CdTe/Cu _x Te Bottom cell: glass/Mo/CIS/CdS/ZnO Mechanical stack	.786 .357 1.14	25.5 6.059	68.9 68.01	13.8 1.47 15.3	4-terminal device
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	

Table 4. Tandem Structures and their Operating Parameters.

The NREL Polycrystalline Thin-Film Group recently demonstrated a 4-terminal polycrystalline thin-film tandem cell consisting of a CdTe-based top cell and a CIS-based bottom cell, officially measured at NREL with efficiency of 15.3%. The top CdTe-based cell structure is: Corning glass/Cd-stannate/Zn-stannate/CdS:O/CdTe/Cu_xTe/ITO. The transparency of this structure is about 50%. The device parameter of this cell measured at: V_{oc}=0.786 volts, J_{sc}=25.55 ma/cm², FF=68.9, efficiency = 13.8%. The CIS bottom cell structure is: glass/Mo/CIS/CdS/ZnO. Its efficiency as measured under the transparent CdTe cell is 1.5%. The performance of the CIS cell degraded somewhat during the processing of the stacks into a measurable structure. This achievement represents a benchmark and a first step for this device; work is ongoing to design material improvements and develop a more optimal structure.

CONCLUSIONS

Both monolithic and mechanical tandems have been developed under the project; they are listed in Table 4. These devices used high-bandgap alloys based on I-III-VI₂ and II-VI compounds. To date, the highest efficiency reported on a 4-terminal structure is 15.3%; thin CdTe is used for the high-bandgap top cell.

The developments under the High-Performance PV Project reported here are progress toward 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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