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Research on Stable, High-Efficiency Amorphous-Silicon Multijunction Modules

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Preface

This report describes work done at Advanced Photovoltaic Systems in Phase II of the NREL contract ZM-2-11091-1, "Research on Stable, High-Efficiency Modules." The goal of the contract is the development of same bandgap, amorphous silicon, tandem-junction modules. The research involved a coordinated effort among two groups in the Research Department of Advanced Photovoltaic Systems: the Device Group and the Module Group. The approach taken was to develop improved semiconductor materials properties in the Device Group. The results of their efforts were then transferred to the Module Group. The Module Group is responsible for scale up of the small-area device results and for optimization of module processing techniques.

Executive Summary

OBJECTIVES

The principal objective of the research described in this report is the development of stable, high-efficiency two-terminal, similar-bandgap, amorphous silicon multijunction photovoltaic modules. Related objectives involve developing cost-effective fabrication processes for these modules and obtaining data on the reliability of the modules. The major goal of the overall research program is the demonstration of a stable, aperture-area efficiency of at least 8.5% for two-terminal, similar-bandgap, amorphous silicon multijunction modules having an aperture area greater than 900 cm^2. The major goal of the second phase of the contract was demonstration of aperture-area efficiency of 7.5% for such modules.

APPROACH

The approach taken involved deposition and characterization of individual semiconductor and non-semiconductor films, single- and tandem-junction devices, and tandem-junction modules. Deposition techniques used were Plasma Enhanced Chemical Vapor Deposition for the various silicon films and layers and magnetron sputtering for the non-semiconductor films and layers. Individual layers were characterized by dark- and photoconductivity (where appropriate), by thickness uniformity measurements, and by optical characterization (where appropriate). Devices were characterized for initial and light-soaked characteristics using I-V and quantum efficiency measurements. Modules were characterized by I-V measurements.

SUMMARY OF RESULTS

Stabilized efficiencies of 8.27% for 1 cm^2 devices and 7.13% for 900 cm^2 modules were obtained. Devices using hydrogenated amorphous silicon oxide p layers and buffer layers were found to have slightly higher stabilized efficiencies than devices with the usual hydrogenated amorphous silicon carbide p layers and buffer layers. The thicknesses and deposition parameters of the p layers in modules were found to be critical in the optimization of stabilized efficiencies.

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Semiconductor Materials Research

Introduction

Semiconductor deposition was performed in three different deposition systems. The first of these, the "B" system, is a small-area deposition system which uses 5.1 cm x 5.1 cm (2" x 2") substrates. It was used for depositions of semiconductor layers and single-junction and tandem-junction devices. The "A" system, a small-area two-chamber deposition system was also used for depositions of individual layers and devices on 5.1 cm x 5.1 cm substrates. Typically, a number of 1 cm^2 devices would be prepared from a run in the A or B system. The third system, the "D2" system, can hold four substrates up to 33 cm x 33 cm (13" x 13") in size. This system was also used for depositions of individual layers and modules.

I Layers

The contract goals in Phase II for i layers were the deposition of an approximately 400-nm thick i-layer with a 1.7-1.8 eV optical bandgap over 900 cm^2, with the same quality as required in Phase I for smaller areas, and a thickness uniformity of $\pm$ 7%. The parameters required in Phase I were a photosensitivity > 10^5 and a hole diffusion length > 150 nm. The photosensitivity is the 1 sun photoconductivity divided by the dark conductivity. Conductivity measurements were performed using small Corning 7059 glass substrates placed on the center of the D2 system box carrier electrodes. Thickness uniformity measurements were performed using soda lime glass substrates 900 cm^2 in size. Therefore conductivity and thickness measurements are reported here separately.

Conductivity

The requirement of a photosensitivity > 10^5 was easily achieved for a variety of deposition conditions in the D2 system, as shown in Table 1. The values for dark conductivity and photoconductivity were relatively low. This is an indication of high purity, since traces of air present during the deposition dope the film slightly n type and raise the dark conductivity and photoconductivity. The highest photosensitivities were achieved for deposition conditions which had low hydrogen dilution (3:1) and low pressure (200-250 mTorr). The optical gaps, as determined from Tauc plots, were all in the required range.

Table 1. Dark and Photo-Conductivity of I-Layers Deposited in the D2 system.

Run	SiH_4 flow rate sccm	H_2 flow rate sccm	Temp. °C	Pres. mTorr	rf Power Dens. mW/ cm^2	E_g eV	Dark cond. S/cm	Photo-cond. S/cm	Photo-sens.
D2-237	50	150	203	200	9.2	1.75	1.5×10^{-12}	7.3×10^{-6}	4.9×10^{6}
D2-239	75	150	217	200	4.6	1.73	2.2×10^{-12}	7.3×10^{-6}	3.4×10^{6}
D2-306	7	35	209	500	18.3	1.76	4.7×10^{-12}	3.7×10^{-6}	7.8×10^{5}
D2-324	10	200	211	500	9.2	1.77	2.5×10^{-12}	2.2×10^{-6}	8.8×10^{5}
D2-326	75	150	209	250	9.2	1.72	1.1×10^{-12}	7.1×10^{-6}	6.4×10^{6}

Table 2. Uniformity and Deposition Rate of I-Layers Deposited in the D2 system.

Run	SiH_4 flow rate sccm	H_2 flow rate sccm	Temp. °C	Pres. mTorr	rf Power Dens. mW/ cm^2	Avg. thick. nm	Dep. Rate nm/s	std. dev. nm	non-unifor.
D2-311	7	35	212	500	4.6	100	0.06	4	8 %
D2-319	40	200	212	500	18	180	0.10	9	10 %
D2-326	75	150	209	250	9.2	477	0.10	30	13 %
D2-329	7	35	211	500	9.2	544	0.08	26	10 %

Uniformity

The goal of 7% non-uniformity (defined as twice the standard deviation divided by the average) in thickness, over 900 cm^2, was not achieved. The best result was a non-uniformity of 8%. It was achieved with a hydrogen-to-silane ratio of 5, a pressure of 500 mTorr, and an rf power density of 4.6 mW/cm^2. Increasing the power density or decreasing the hydrogen dilution increased the non-uniformity and the deposition rate, as shown in Table 2.

N Layers

The contract goal for n layers in Phase II was a uniformity of ±10% over an area of 900 cm^2, using a thickness of 100 nm for diagnostic purposes. Another goal involving n layers was the selection of either an amorphous or microcrystalline n layer for multijunction devices. The choice made will be discussed later in the report.

Amorphous

The uniformity goal was achieved easily for amorphous n layers. The deposition conditions and film properties from the most uniform deposition are given in Table 3. Depositions were performed on glass substrates only, unlike microcrystalline n-layers. The ratio of hydrogen flow to silane flow was 5 and the pressure was 500 mTorr, the same as the conditions which gave the most uniform i layers.

Table 3. Properties of an Amorphous N-layer Deposited in the D2 system.

Run	4% PH_3 in SiH_4 flow rate sccm	H_2 flow rate sccm	Temp. °C	Pres. mTorr	rf Power Dens. mW/ cm^2
D2-277	7.2	37	211	500	18
E_g eV	**Dark cond. S/cm**	**Avg. thick. nm**	**Dep. Rate nm/s**	**std. dev. nm**	**non-unifor.**
1.78	4.2×10^{-4}	159	0.09	2.5	3%

It is not clear why the uniformity of the n-layer deposition described in Table 3 was so much better than the uniformity of the i-layer depositions described in Table 2 under fairly similar conditions.

Microcrystalline

Lateral conductivity measurements on n layers deposited with high hydrogen dilution and at high power showed different results depending on whether the deposition was done on 7059 glass or on a previously deposited i layer, as shown in Table 4. Various concentrations of phosphine in silane were used by either using the phosphine in silane alone or with pure silane as well. It was necessary to go to higher hydrogen dilution and higher power to get good lateral conductivity when the n layer was deposited on an i layer. The goal of < 10% non-uniformity was achieved.

Table 4. Properties of Microcrystalline N-layers Deposited in the D2 System.

Run	SiH_4 flow rate sccm	4% PH_3 in SiH_4 flow rate sccm	H_2 flow rate sccm	Press. mTorr	rf Power Dens mW/ cm^2	Temp. C	substr	cond. S/cm	non-unifor.
D2-175	1.0	0.5	100	1000	28	215	glass	1-2	not measured
D2-195	1.0	0.5	100	1000	28	207	i-layer	1.0×10^{-7}	not measured
D2-196	1.0	0.5	100	1000	28	209	glass	7-11	13%
D2-199	0.0	1.0	200	1000	46	214	i-layer	0.1	not measured
D2-200	0.0	0.8	200	2000	69	215	i-layer	0.2	9%

Non-Semiconductor Materials Research

Tin Oxide

High-haze, high-transmission, low sheet-resistivity transparent-conducting-oxide-coated glass is essential for achieving high efficiency amorphous silicon devices and modules. One goal of this phase of the contract was achievement of ≥ 82% integrated transmittance and ≤ 5 ohms/square sheet resistance for SiO_2 / SnO_2-coated glass with a resistance non-uniformity ≤ 10% over 900 cm^2 and optimized diffuse transmission. Work toward this goal was discontinued early in Phase II due to budgetary restraints. Consequently, Asahi type "U" tin oxide on 1 mm-thick glass was purchased for tandem module fabrication.

Zinc Oxide

Sputtered zinc oxide was used as a back reflector to increase the red response and hence short-circuit current density of tandem-junction devices. The goal in Phase II of the contract for this activity was the demonstration of a 17% increase in short-circuit current density compared to that obtained with an Al back reflector for a 1 cm^2 tandem-junction cell. The data in Table 5 shows a 21% increase in short-circuit current density resulting from the use of ZnO back reflector.

Table 5. 1 cm^2 Device Parameters with and without Zinc Oxide Back Reflector.

Run Number	back reflector	V_{oc} Volts	J_{sc} mA/cm2	Fill Factor	Efficiency %
B609	Ag/Al	1.563	6.54	0.696	7.12
B609	ZnO/Ag/Al	1.570	7.90	0.686	8.50

Device Research

Tunnel-Junctions

A good "tunnel junction" between photovoltaic junctions is necessary for the proper operation of a multi-junction device. Recombination between the electrons from one junction and the holes of the other junction occur in the tunnel junction. If complete recombination does not occur, the charge carriers from one junction must surmount a large potential barrier in the other junction, resulting in a large loss of energy. Phase II of the contract required the determination of the n1 / p2 tunnel junction properties for the best p2 layer material.

Steve Hegedus at the Institute of Energy Conversion of the University of Delaware measured five cells each of four n-p tunnel junction structures fabricated at APS and reported the results shown in Table 6. The non-linearity was determined by comparing results at +50 mV and -50 mV.

Table 6. Resistivities of Various Tunnel Junction Structures.

n configuration	p configuration	R ohms*cm^2 @ 50 mV	Non-Linearity
"standard" n-µc-Si	p-a-Si:H/p-a-SiC:H	3-6	1%
"standard" n-µc-Si	p-µc-Si/p-a-Si:H/p-a-SiC:H	5-10	1%
n-a-Si:H	p-a-Si:H/p-a-SiC:H	25-50	1%
"Ar diluted" n-µc-Si	p-a-Si:H/p-a-SiC:H	25-60	3-6%
n-a-Si:H	p-µc-Si/p-a-Si:H/p-a-SiC:H	50-1000	2-7%
n-a-Si:H	p-a-SiC:H	150-300	1%

The term "Ar diluted" n-µc-Si refers to n-type microcrystalline silicon deposited from a hydrogen (120 sccm), phosphine in silane (1 sccm), and argon (2 sccm) mixture. Information in the literature indicated that argon ion bombardment might enable attainment of high conductivity with a thinner microcrystalline n layer. The standard microcrystalline silicon is deposited from a hydrogen and silane mixture (50:1). The p-a-Si:H layer used in the p structures is approximately 5 Angstroms in thickness. Its purpose is to serve as a low-bandgap recombination region.

It is clear from the table that the use of a microcrystalline n layer results in better tunnel junction properties than the use of an amorphous n layer, even if there is a microcrystalline p layer adjacent to the n layer. The argon diluted microcrystalline n layer is inferior to the microcrystalline n layer deposited from silane and hydrogen, at least for tunnel junctions.

Tandem-Junction Devices

Phase II of the contract contains a major milestonel of achieving an average >8.5% stable efficiency and an open-circuit voltage of > 1.84 Volts from a 1 cm^2 multijunction cell sampled over an area of 900 cm^2, after exposure to white light (1000 W/m^2) for 600 hours. Tandem junction devices were made in the B system (5 cm x 5 cm) substrates and in the D2 system (900 cm^2 substrates), as diagnostics for the modules.

Table 7. Initial and Light-Soaked Parameters of 1 cm^2 Tandem Devices.

hours of light soaking	V_{oc} Volts	J_{sc} mA/cm^2	Fill Factor	Efficiency %	Series Resistance ohm-cm^2
D2-240-RWM: Asahi "C" tin oxide / p1 (a-SiC) / b1 (a-SiC) / i1 (a-Si) / n1 (μc-Si) / p2 (a-SiC) / b2 (a-SiC) / i2 (a-Si) / n2 (a-Si) / ZnO / Al					
0	1.599	7.79	0.69	8.60	14.8
600	1.554	7.56	0.635	7.47	27.4
B-765: Asahi "U" tin oxide / ZnO / $p1^+$ (a-Si) / p1 (a-SiC) / b1 (a-SiC) / i1 (a-Si) / n1 (μc-Si) / $p2^+$ (a-Si) / p2 (a-SiC) / b2 (a-SiC) / i2 (a-Si) / n2 (μc-Si) / ZnO /Ag / ZnO /Al					
0	1.652	7.87	0.717	9.32	24.3
600	1.58	7.83	0.604	7.47	38.3
B-720: Asahi "U" tin oxide / ZnO / $p1^+$(a-Si) / p1 (a-SiC) / b1 (a-SiC) / i1 (a-Si) / n1 (a-Si) / p2 (μc-Si) / $p2^+$ (a-Si) / p2 (a-SiC) / b2 (a-SiC) / i2 (a-Si) / n2 (μc-Si) / ZnO / Al					
0	1.648	8.44	0.691	9.62	27.8
608	1.624	8.19	0.591	7.85	47.0
673*	1.630	8.36	0.594	8.10	47.7
B-833: MgF / Asahi "U" tin oxide / ZnO / $p1^+$ (a-Si) / p1 (a-SiO) / b1 (a-SiO) / i1 (a-Si) / n1 (a-Si) / p2 (μc-Si) / $p2^+$ (a-Si) / p2 (a-SiO) / b2 (a-SiO) / i2 (a-Si) / n2 (a-Si) / n2 (μc-Si) / ZnO / Ag / ZnO / Al					
0	1.639	8.73	0.682	9.76	30.3
637	1.600	8.55	0.604	8.27	43.8

* MgF anti-reflection coating added at 673 hours(no annealing as the fill factor was unchanged)

The best tandem junction devices made in the B system had p layers and buffer layers in both junctions made of SiO, rather than the usual SiC. Silane and carbon dioxide were used for their fabrication, with the addition of trimethylboron for the p layers. The carbon dioxide to silane flow ratios were 3:1 for the p layers and 3:2 for the buffer layers. Light soaking data for a number of tandem devices with different structures are shown in Table 7.

The devices fabricated in the B system had i layer thicknesses of 59 nm for i1 and 350 nm for i2.The data in Table 7 show that replacement of the SiC p layers and buffer layers by SiO resulted in an increase in stabilized efficiency of 0.17%, from 8.10% to 8.27%. A larger change had been expected, since a similar change in single-junction devices resulted in an increase from 8.0% to 8.5%. Another puzzling result shown in the table is the fact that the tunnel junction structure which gave the best results (n-a-Si:H/p-μc-Si/p-a-Si:H/p-a-SiC:H) is the one which has a relatively high series resistance (see Table 6). However, it should be noted that the tunnel junction structures were deposited in a different deposition system (A system) than the tandem junction devices (deposited in the B system).

The results for 1 cm^2 devices from run D2-240, reported in Table 7, are not representative of the best module results, which were obtained using Asahi type "U" tin oxide, rather than Asahi type "C" tin oxide. Diagnostic 1 cm^2 devices were not made on Asahi type "U" tin oxide, as experiments with the type "U" substrate were concentrated on module fabrication.

Module Research

The major milestone in Phase II of the contract for modules is the achievement of 900 cm^2 module with an aperture area efficiency of 7.5% or more after 600 hours of continuous light soaking. The best result was obtained from run D2-346 from the right-wall (rw) substrate. Note that the D2 system box carrier contains four substrates denoted by position: left wall (lw), left electrode (le), right electrode (re), and right wall (rw).

Module D2-346rw had an aperture area efficiency of 7.13% after 584 hours of light soaking. Its power output at that time was 6.04 Watts and its aperture area is 848 cm^2. The parameters of D2-346rw as a function of light-soaking time are shown in Table 8. The increase in open-circuit voltage with light soaking is an indication that the p layer is quite thin. Another requirement in the major milestone is the achievement of an active area loss due to labor scribing of less than 4%. An active area loss of approximately 4.5% was achieved. Measurements of thin-film thickness non-uniformity for modules, which was required to be less than 10%, were not made in Phase II, as the result was obtained in Phase I.

The thin film structure used in module D2-346rw is: Asahi "U" tin oxide / p-a-SiC:H / i-a-SiC:H / i-a-Si:H / n-μc-Si / p-a-SiC:H / i-a-SiC:H / i-a-Si:H / n-a-Si:H / ZnO / Al. The i-a-SiC:H layers between the p-a-SiC:H and i-a-Si:H layers are buffer layers.

Table 8. Parameters of Module D2-346rw as a Function of Light-Soaking Time.

hours of light soaking	V_{oc} / cell Volts	J_{sc} mA/cm^2 aperture area	Fill Factor	Efficiency
0	1.587	7.07	0.696	7.81
47	1.619	6.81	0.662	7.29
160	1.602	6.88	0.652	7.18
584	1.641	6.90	0.630	7.13

Run 346 was part of a series of runs in which the p1, i1, and i2 deposition conditions were varied. The device structure was the same for all of the runs. Initial and 600 hour efficiencies for the series are shown in Table 9. Different deposition conditions are separated by double lines. The best results were obtained with a p1 deposition pressure of 150 mTorr, a p1 power density of 4.6 mW/cm^2 and i1 and i2 layer thicknesses of 66 nm and 360 nm, respectively. Higher values for the p1 pressure and power were then tested, to improve the stability of the glow-discharge.

However, as seen in the table, these changes resulted in lower initial and stabilized efficiencies. The p1 deposition time was then reduced, as it is believed that the greater thickness of the p1 layer is responsible for the decrease in efficiency. The reduction in p1 deposition time brought the initial efficiency back up to around 8%. The stabilized efficiency is not yet known, as the modules are still being light soaked.

Table 9. Initial and 600 hour Efficiencies for a Series of 900 cm² Modules

Module #	p1 dep. time secs	p1 dep. pres. mTorr	p1 dep. power dens. mW/cm²	i1 thick nm	i2 thick nm	initial eff. %	600 hour eff. %	drop %
346rw	70	150	4.6	66	360	7.81	7.13	8.7
346lw	70	150	4.6	66	360	8.04	7.1	11.7
348lw	70	150	4.6	66	400	8.22	6.89	16.2
345lw	70	150	4.6	66	320	7.09	6.38	10.0
342rw	70	150	4.6	72	430	7.78	6.58	15.4
344rw	70	150	4.6	72	360	7.53	6.30	16.3
350lw	70	300	4.6	66	360	7.67	6.39	16.7
352lw	70	300	4.6	60	360	7.04	6.34	9.9
355lw	55	300	9.2	66	360	7.77	6.15	20.8
355rw	55	300	9.2	66	360	7.70	5.83	24.3
357lw	55	300	9.2	60	360	7.65	6.04	21.0
378lw	45	300	9.2	60	360	8.16		
381lw	45	300	9.2	60	360	8.14		

The data in Table 9 show that the decrease in module efficiency caused by the 600 hours of light soaking varied over a large range for the different deposition conditions. Figure 1 shows that the level of decrease is strongly correlated with the initial open-circuit voltage per cell of the module. Note that the center of the data label (the module number) corresponds to the point.

The relationship shown in Figure 1 is consistent with our previous experience with single-junction devices, that p layer deposition conditions that give larger values of open-circuit voltage result in more light-induced degradation. The modules with the highest values of stabilized efficiency, 346rw, 346lw, and 348lw, fall below the general trend in Figure 1.

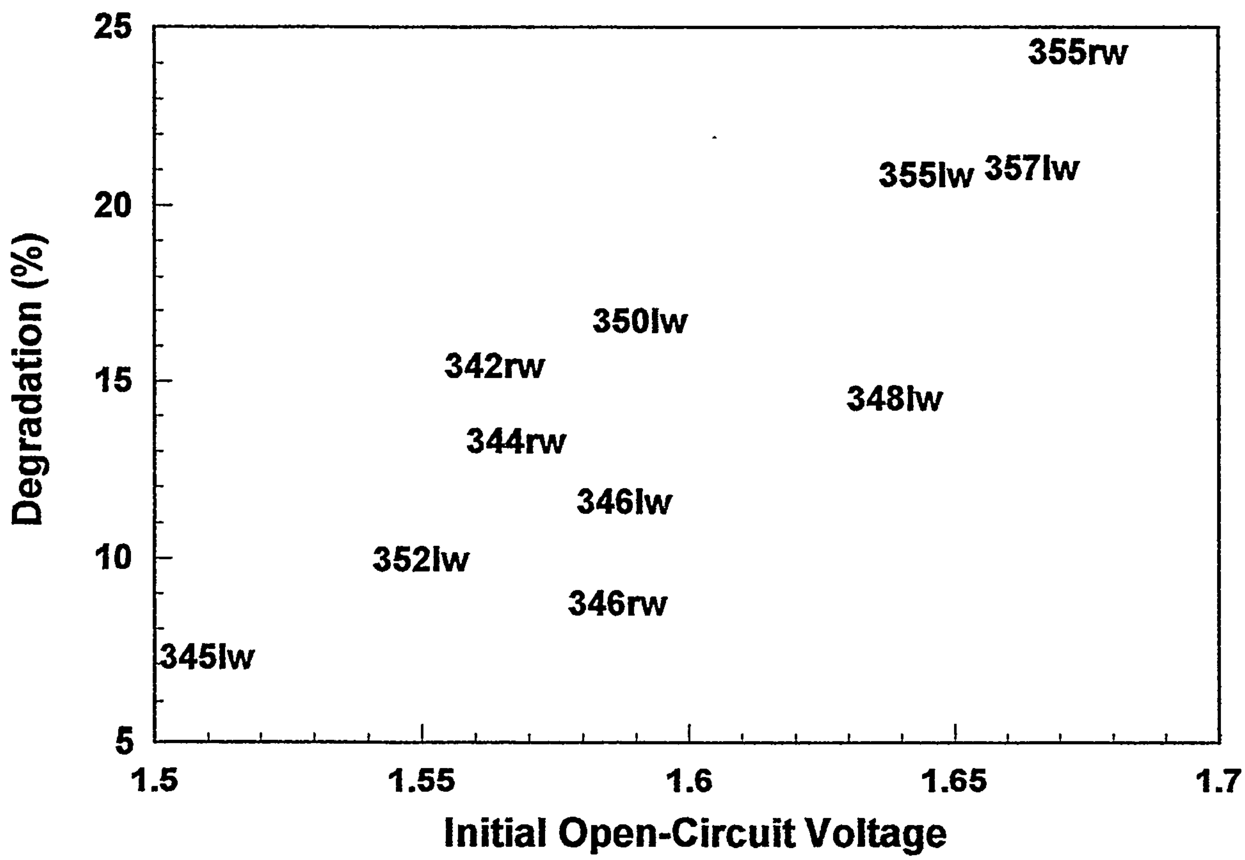

Figure 1. Relationship between module open-circuit voltage and module light-induced degradation.

Four light-soaked modules were encapsulated, for longer-term and outdoor light soaking. The encapsulation process, which is done at 150 C, appears to have partially annealed the modules. The highest efficiency measured was 7.32% for module 348lw, which was 6.89% before encapsulation. The encapsulated modules will be sent to NREL for measurement and light soaking.

Work will continue in Phase III on the optimization of the p1 layer deposition parameters before proceeding to optimization of other aspects of the thin film structure. A study of the firing voltage for the p-layer gas mixture as a function of pressure will be made first to determine the optimum pressure for initiating and maintaining the discharge. Following that, a systematic study (using designed experiments) of the effect of the p-layer deposition conditions will be made. The goal of these studies will be to increase open-circuit voltage and short-circuit current (initial and stabilized) while maintaining or reducing the level of degradation in fill factor. Optimization of the other critical parts of the tandem-junction structure, such as the tunnel junction, will then be undertaken.

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13. ABSTRACT *(Maximum 200 words)*

This report describes work performed on same-band-gap, amorphous-silicon, tandem-junction 1-cm^2 devices and 900-cm^2 modules. The same-band-gap, amorphous-silicon, tandem-junction structure is of interest because it suffers from less light-induced degradation than the single-junction structure. The tandem-junction structure is more complex than the single-junction structure in that the interface between the two junctions, the so-called "tunnel junction," must enable the efficient recombination of holes from one junction with electrons from the other junction without absorbing a significant amount of light. The topics covered in this report include the deposition of the various amorphous silicon thin-film layers with the required electrical properties and uniformity for modules, deposition of nonsemiconductor layers for the front and back contact with the requisite optical properties, preparation and characterization of tunnel junctions and small-area devices, and preparation and characterization of modules.

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