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SILICON MATERIALS TASK OF THE LOW-COST SOLAR ARRAY PROJECT
(PHASE IV)

Effects of Impurities and Processing on Silicon Solar Cells
Twentieth Quarterly Report, July–September 1980

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November 14, 1980

Work Performed Under Contract No. NAS-7-100-954331

Westinghouse R&D Center
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Solar Energy

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Silicon Solar Cells

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November 14, 1980

July 1980-September 1980

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and P. Rai-Choudhury (Westinghouse R&D Center)
and H. C. Mollenkopf (Hemlock Semiconductor Corporation)

Contract No. 954331

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TECHNICAL CONTENT STATEMENT

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NEW TECHNOLOGY

No new technology is reportable for the period covered by this report.

TABLE OF CONTENTS

	Page
1. SUMMARY	1
2. INTRODUCTION.	2
3. TECHNICAL PROGRESS.	3
3.1 Crystal Growth and Analysis.	3
3.1.1 Ingot Preparation	3
3.1.2 Ingot Evaluation.	5
3.2 Evaluation of Experimental Silicon Materials	8
3.3 Evaluation of Impurity Behavior in Polycrystalline and Single Crystal Silicon by Spectral Response Measure- ments.	15
3.4 Silicon Material Evaluation by DLTS.	25
3.5 The Permanence of Impurity Effects in Silicon Solar Cells.	25
3.5.1 Temperature Effects	25
3.5.1.1 Experimental Method.	25
3.5.1.2 Experimental Results	27
3.5.2 Electrical Bias Effects	28
3.6 Impurity Effects in High Efficiency Solar Cells.	48
4. CONCLUSIONS	53
5. PROGRAM STATUS.	56
5.1 Present Status	56
5.2 Future Activity.	56
6. REFERENCES.	58
7. ACKNOWLEDGEMENTS.	59

LIST OF TABLES

Table	Page
1 Ingot Impurity, Structure, and Task.	4
2 Ingot Impurity Concentration.	6
3 Best Estimate of Impurity Concentrations	9
4 Ingot Electrical and Defect Characteristics.	11
5 Ingot Carbon and Oxygen Concentrations	13
6 Lighted I-V Data from the Solar Cells Used for Spectral Response Measurements.	18
7 DLTS Results on Impurity-Doped Ingots.	26
8 Ingots Used in Permanence Studies.	27
9 Silicon Materials Undergoing Electrical Bias Testing	30
10 Calculated Performance for Ohmic Back, Wide-base Solar Cells	51
11 Calculated Performance for Narrow-base Back-field Solar Cells	54

LIST OF FIGURES

Figure		Page
1	The Effect of Cr, Mo, and Ti, on the Spectral Response of Single Crystal Silicon Solar Cells.	16
2	Change in the Spectral Response of Single Crystal Silicon Solar Cells with Ti Impurity Additions.	19
3	Effect of $1.2 \times 10^{14} \text{ cm}^{-3}$ Titanium on the Spectral Response of Single Crystal and Polycrystalline Silicon Solar Cells .	20
4	Effect of $\sim 2 \times 10^{13} \text{ cm}^{-3}$ Titanium on the Spectral Response of Single Crystal and Polycrystalline Silicon Solar Cells.	21
5	Effect of Molybdenum on the Spectral Response of Single Crystal and Polycrystalline Silicon Solar Cells	23
6	Effect of $1 \times 10^{15} \text{ cm}^{-3}$ Chromium on the Spectral Response of Single Crystal and Polycrystalline Silicon Solar Cells. . .	24
7	Relationship of Efficiency to Temperature and Time for a Copper-Doped Ingot.	29
8	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Base Line Cells	31
9	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Fe-doped ($1.06 \times 10^{15} \text{ cm}^{-3}$) Cells	32
10	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Nb-doped ($< 6.44 \times 10^{15} \text{ cm}^{-3}$) Cells	33
11	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Ag-doped ($2.20 \times 10^{15} \text{ cm}^{-3}$) Cells	34
12	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Cr-doped ($1.04 \times 10^{15} \text{ cm}^{-3}$) Cells	35

LIST OF FIGURES (Cont.)

Figure		Page
13	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Fe-doped ($0.4 \times 10^{15} \text{cm}^{-3}$) Cells.	36
14	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Cu-doped ($65 \times 10^{15} \text{cm}^{-3}$) Cells.	37
15	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Nb-doped ($<0.009 \times 10^{15} \text{cm}^{-3}$) Cells.	38
16	Effect of Electrical Bias on Relative Efficiency as a Function of Stress Temperature--Ti-doped ($0.105 \times 10^{15} \text{cm}^{-3}$) Cells.	39
17	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Fe-doped ($1.06 \times 10^{15} \text{cm}^{-3}$) Cells.	40
18	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Nb-doped ($<0.044 \times 10^{15} \text{cm}^{-3}$) Cells.	41
19	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Ag-doped ($2.20 \times 10^{15} \text{cm}^{-3}$) Cells.	42
20	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Cr-doped ($1.04 \times 10^{15} \text{cm}^{-3}$) Cells.	43
21	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Fe-doped ($6.4 \times 10^{15} \text{cm}^{-3}$) Cells.	44
22	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Cu-doped ($65 \times 10^{15} \text{cm}^{-3}$) Cells.	45
23	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Nb-doped ($<0.009 \times 10^{15} \text{cm}^{-3}$) Cells.	46
24	Effect of Electrical Bias on Relative Efficiency, Normalized to Baseline Cell Behavior, as a Function of Stress Temperature--Ti-doped ($0.105 \times 10^{15} \text{cm}^{-3}$) Cells.	47

LIST OF FIGURES (Cont.)

Figure		Page
25	Internal Recombination Velocity Plots for Ohmic Back Solar Cells.	52
26	Internal Recombination Velocity Plots for Back Field Solar Cells.	55
27	Program Plan	57

1. SUMMARY

The overall objective of this program is to define the effects of impurities, various thermochemical processes and any impurity-process interactions upon the performance of terrestrial solar cells. The results of the study form a basis for silicon producers, wafer manufacturers, and cell fabricators to develop appropriate cost-benefit relationships for the use of less pure, less costly solar grade silicon.

Eight additional p-type silicon ingots were grown during this reporting period; one of these was grown from experimental, dichlorosilane-based poly silicon. Preliminary evaluation of this material shows it to be comparable in behavior to our standard silicon derived from trichlorosilane decomposition.

Spectral response measurements made on single crystal and polycrystalline silicon solar cells containing specific impurities agreed well with measured cell efficiencies. For polycrystalline cells it is shown that both grain boundaries and metallic impurities reduce carrier lifetime, resulting in reduced red response and reduced cell efficiency. Spectral response and DLTS measurements on chromium-doped polycrystalline silicon cells indicate an interaction between chromium and grain boundaries; the nature of this interaction is not yet understood.

Measurements were made to evaluate possible long term effects of copper contamination on solar cell performance. The time-temperature behavior of copper-doped material does not follow a simple Arrhenius model, so it is not possible to predict with certainty what these long term effects might be. Copper behavior is similar to that of iron in this respect.

Nine groups of cells, including a baseline cell group, are undergoing electrical/temperature tests to determine whether electric fields play a role in long term cell degradation. Step-wise testing

at regularly increasing temperatures have not yet conclusively shown an effect which can be attributed to electric field/impurity interactions. These tests are continuing.

A mathematical model for impurity effects in high efficiency solar cells has been developed. The cases of wide base, ohmic back contact cells and of narrow base, back-field cells are treated separately. It is shown that, in both cases, large effective carrier diffusion lengths must be obtained for high efficiency cells. This implies that the concentration of lifetime-killing impurities must be kept low.

2. INTRODUCTION

This is the twentieth quarterly report describing activities conducted under JPL Contract 954331, and is the third report of the Phase IV studies.

In Phase III, "An Investigation of the Effects of Impurities and Processing on Silicon Solar Cells," the effects of thermal processes, impurities, and impurity-process interactions were determined and documented. The development of this data base led to a more precise definition of what constitutes an acceptable "Solar Grade" Silicon. In addition, it provided silicon manufacturers with a rationale for selection of construction materials; it has helped ingot, sheet, or ribbon manufacturers to specify the purity of silicon feedstocks; and it has enabled cell manufacturers to define acceptable wafer purities for cell fabrication and to choose processes which minimize adverse impurity effects. In short, the impurity effect data provide a basis for cost-benefit analysis to producers and users of Solar Grade Silicon.

In Phase IV of this program, the approaches and techniques developed in Phase III are being extended to several new areas, as well as to developing an improved data base for aging effects and certain process-related phenomena. The Phase IV tasks include (1) evaluation, by previously developed process techniques, of the properties of silicon produced by experimental low cost processes; (2) extending threshold

impurity concentration data to high efficiency cells; (3) measuring the effects of interaction between impurities and grain boundaries in polycrystalline solar cells; (4) evaluating the long term effects of impurities in solar cells; and (5) examining the effects of processes such as ion implantation on contaminated solar cells.

3. TECHNICAL PROGRESS

3.1 Crystal Growth and Analysis

3.1.1 Ingot Preparation

During the third quarter of Phase IV of the Silicon Materials Task of the LSA project effort consisted of monocrystalline and polycrystalline ingot growth. Additional regrowths were made for some polycrystalline samples in order to suppress excessive metal-rich inclusion crystal structure breakdown.

Eight p-type silicon ingots were completed this quarter for microstructural, chemical, electrical and solar cell characterization. Six of the ingots were 4 ohm-cm resistivity while the seventh, made from experimental dichlorosilane - based polysilicon, was backdoped to 1.4 ohm-cm. Each ingot's intentionally added impurity dopant, crystalline structure, and the experimental task for which it was given are indicated in Table 1. All ingots were prepared by Czochralski crystal growth; a description of the method with details of the crystal growth equipment and conditions can be found in earlier reports.^(1,2)

Additional ingot regrowths were necessary for the Fe and Ag doped ingots listed in Table 1 to reduce crystal structure breakdown (constitutional supercooling) caused by excessive heavy metal doping in the first trial growth.

The first silicon ingot was produced this quarter under the experimental silicon task of this program. A first generation maximum gadolinium doped ingot was also grown. Both the experimental silicon and gadolinium doped ingots were monocrystalline. Heavy Mn doping of a

TABLE 1 INGOT IMPURITY, STRUCTURE AND TASK

<u>Impurity</u>	<u>Structure</u>	<u>Task</u>
Fe *	Poly	Poly
Ag *	Single	Permanence
Ni	"	"
None **	"	Experimental Silicon
Mn	"	Permanence
Mn ***	"	Permanence
Cr	Poly	Poly
Gd	Single	Thermochemical Progressing

* Two ingot growths necessary with the second having reduced impurity concentration to suppress constitutional supercooling effects.

** Hemlock Semiconductor's experimental dichlorosilane-based silicon.

*** Single growth prohibited due to excessive impurity doping for permanence studies.

melt designated to produce an ingot for extended permanence studies was also attempted, but monocrystalline growth was prohibited by the excessive metal doping.

3.1.2 Ingot Evaluation

Detailed ingot impurity data are indicated in Table 2 with the listing of ingot identification, intentionally added target impurity concentration, ingot impurity concentration based on melt analysis by atomic absorption (calculated concentration), and ingot seed impurity concentration measured by spark source mass spectrometry.^(2,3)

Ingot W205 was a polycrystalline regrowth with the Fe concentration reduced from 8 to 5×10^{14} atoms/cm³. The reduction in impurity concentration was necessary to reduce heavy metal precipitation and excessive fine grain structure. Similar adjustments were also necessary for ingot W227 where the Cr concentration was reduced from 1 to 0.4×10^{15} atoms/cm³ since heavy metal inclusions were found in at least half of the more heavily doped wafers. These lower impurity concentration adjustments were implemented to eliminate metal inclusions which shunt solar cells made on the metal-rich wafers.

Extensive spark source mass spectrometer analysis (SSMS) of the heavy Ni doped ingot W221 (1×10^{16} atoms/cm³) showed no detectable Ni concentration in ingot seed samples. Tang sample analysis showed Ni concentrations from 5 to 150 ppma signifying clustering of the intentionally added impurity. The inability to detect Ni in the seed samples is due to the low intensity of the Ni⁺⁺⁺ line (Ni⁺ and Ni⁺⁺ have identical lines with silicon at $m/e = 58$ and 29). This problem has been noted before.⁽¹⁾ The growth of single crystal ingot W224 was completed this quarter using silicon produced from Hemlock Semiconductor's experimental dichlorosilane - based reductive chemical vapor deposition process.⁽⁴⁾ Pertinent details concerning the run conditions etc. for producing the dichlorosilane silicon are found in reference 5. The ingot produced from the dichlorosilane silicon was back doped to 1.4 ohm-cm and had p-type conductivity. As shown in Table 2, SSMS analysis indicated heavy metals to be below the sensitivity of the mass spectrometer.

TABLE 2. INGOT IMPURITY CONCENTRATION

<u>Ingot Identification</u>	<u>Target Concentration $\times 10^{15}$ atoms/cm³</u>	<u>Calculated Concentration $\times 10^{15}$ atoms/cm³</u>	<u>Mass Spec. Analysis $\times 10^{15}$ atoms/cm³</u>
W-198-00-000	None	N/A	None
W-199-00-000	None	N/A	None
W-200-V-004-Poly	0.4	0.38	18.5*
W-201-Mo-007-Poly	0.005	0.003	77*
W-202-Ti-013-Poly	0.02	0.018	<0.25
W-203-V-005-Poly	0.04	0.053	<0.15
W-204-Cr-008-Poly	1.0	0.82	1322*
W-205-Fe-009-Poly	0.5	0.61	<1.5
W-206-V-006	0.02	0.026	<0.15
W-207-Mo-008	0.002	0.002	<0.5
W-208-Cr-009	0.2	0.19	0.6
W-209-Ti-014	0.02	0.024	<0.25
W-210-Ti-015	0.08	0.10	<0.25
W-211-Cu-007	1.0	1.0	2.6
W-212-Cu-008	10	12.5	27
W-213-Pb-001	Max. Conc.	Non Detectable	<0.10+
W-214-V-007-Poly	0.20	0.30	0.55**
W-215-Mo-009-Poly	0.0025	0.002	<0.5**
W-216-Cr-010-Poly	0.80	0.64	2.2**
W-217-Ta-005	0.00015	0.0003	<0.5
W-218-Ta-006	0.000065	0.0001	<0.5
W-219-V-008	0.007	0.009	<0.15

TABLE 2. INGOT IMPURITY CONCENTRATION (continued)

Ingot Identification	Target Concentration $\times 10^{15}$ atoms/cm ³	Calculated Concentration $\times 10^{15}$ atoms/cm ³	Mass Spec. Analysis $\times 10^{15}$ atoms/cm ³
W-220-W-005	0.0008	0.0007	<0.15
W-221-Ni-005	10	8.2	Processing
W-222-Ag-002	4.5	3.2	6.0
W-223-Ni-006	1.0	1.1	<1.5
W-224-HSC/DCS057	NA ⁺⁺	None	$\leq 0.2^{+++}$
W-225-Mn-009	1.0	1.5	5.5
W-226-Mn-010	4.0	***	***
W-227-Cr-011-Poly	0.4	Processing	Processing**
W-228-Gd-001	Max.	Processing	Processing

* Ingots contain metal-rich inclusions due to constitutional supercooling.

** Ingots regrown to remove metal-rich inclusions due to constitutional supercooling.

+ Pb dopant vaporized on two separate ingot growths.

++ No intentional impurity

+++ Heavy metals $\leq$ sensitivity of SSMS.

*** Single growth prohibited due to excessive impurity doping for permanence studies

A best estimate⁽³⁾ of the impurity concentrations for Phase IV ingots is listed in Table 3. Additional spark source evaluations of W212 were completed this quarter to check the possibility that initial high spark source values were indicative of impurity clustering. Subsequent averaged evaluation results yielded better agreement between the calculated and spark source results. Additional analyses are also warranted for W225 to determine the nature of the discrepancy between a calculated Mn concentration of $1.5 \times 10^{15} \text{ cm}^{-3}$ and a value of $5.5 \times 10^{15} \text{ cm}^{-3}$ determined by mass spectrometry. This will be completed early in the next quarter.

The resistivity and etch pit data for all of Phase IV ingots are indicated in Table 4. Etch pit analysis is not applicable to the polycrystalline samples.

The carbon and oxygen concentrations of each odd-numbered ingot were measured by infrared absorption at room temperature using Fourier Transform Infrared Spectroscopy. The amplitude of the absorption peaks at 605 cm^{-1} and 1107 cm^{-1} is proportional to the carbon and oxygen concentrations respectively. Carbon and oxygen concentrations for recently grown ingots are listed in Table 5. Normal carbon and oxygen concentrations found in Czochralski grown material are in the range of 2.5 to $5 \times 10^{17} \text{ atoms/cm}^3$ for carbon and 5 to $150 \times 10^{16} \text{ atoms/cm}^3$ for oxygen.

3.2 Evaluation of Experimental Silicon Materials

Several complementary capabilities to assess the impact of impurities in silicon feedstock on subsequently fabricated solar cells have been developed as part of this program. Among these are the ability to project via mathematical models the expected cell performance from the raw material analysis. With slight modification, the models also permit sensitivity analyses to be carried out so that for a given impurity-bearing feedstock we can identify which impurity has the greatest impact on performance and how much further refining is necessary (and cost effective) to reduce the impurity to tolerable levels. This approach,

TABLE 3. BEST ESTIMATE OF IMPURITY CONCENTRATIONS

<u>Ingot Identification</u>	<u>Best Estimate of Impurity Conc. (X 10¹⁵ ATOMS/CM³)</u>
W-198-00-000	NA*
W-199-00-000	NA
W-200-V-004-Poly	0.38
W-201-Mo-007-Poly	0.003
W-202-Ti-013-Poly	0.018
W-203-V-005-Poly	0.05
W-204-Cr-008-Poly	0.82
W-205-Fe-009-Poly	0.61
W-206-V-006	0.026
W-207-Mo-008	0.002
W-208-Cr-009	0.19
W-209-Ti-014	0.02
W-210-Ti-015	0.10
W-211-Cu-007	1.8
W-212-Cu-008	12.5
W-213-Pb-001	ND+
W-214-V-007-Poly	0.4
W-215-Mo-009-Poly	0.002
W-216-Cr-010-Poly	1.0
W-217-Ta-005	0.0003
W-218-Ta-006	0.0001
W-219-V-008	0.009
W-220-W-005	0.0007
W-221-Ni-005	8.2

TABLE 3. BEST ESTIMATE OF IMPURITY CONCENTRATIONS (Continued)

<u>Ingot Identification</u>	<u>Best Estimate of Impurity Conc. (X 10¹⁵ ATOMS/CM³)</u>
W-222-Ag-002	4.6
W-223-Ni-006	1.1
W-224-HSC/DCS057	++
W-225-Mn-009	1.5
W-226-Mn-010	***
W-227-Cr-011-Poly	Processing
W-228-Gd-001	Processing

- * Not applicable
- + ND - Non detectable
- ++ No intentional impurity
- *** Single growth prohibited due to excessive impurity doping for permanence studis

TABLE 4. INGOT ELECTRICAL AND DEFECT CHARACTERISTICS

<u>Ingot Identification</u>	<u>TGT Resistivity (ohm-cm)</u>	<u>Actual Resistivity (ohm-cm)</u>	<u>Etch Pit Density (/cm²)</u>
W-198-00-000	4.0	4.1-3.9	0-3K
W-199-00-000	4.0	3.7-3.5	1-5K
W-200-V-004/Poly	4.0	3.6-2.3	NA
W-201-Mo-007/Poly	4.0	3.8-2.3	NA
W-202-Ti-013/Poly	4.0	5.3-3.9	NA
W-203-V-005/Poly	4.0	4.4-3.8	NA
W-204-Cr-008/Poly	4.0	4.7-4.3	NA
W-205-Fe-009/Poly	4.0	4.0-3.2	NA
W-206-V-006	4.0	3.7-3.6	0-5K
W-207-Mo-008	4.0	3.8-3.5	0-15K
W-208-Cr-009	4.0	3.7-3.5	0-15K
W-209-Ti-014	4.0	4.0-3.3	0-10K
W-210-Ti-015	4.0	4.0-3.5	0-5K
W-211-Cu-007	4.0	4.0-3.1	0-5K
W-212-Cu-008	4.0	3.9-3.3	5-20K
W-213-Pb-001	4.0	3.3-2.7	10-20K
W-214-V-007-Poly	4.0	3.8-3.1	NA
W-215-Mo-009-Poly	4.0	3.8-1.7	NA
W-216-Cr-010-Poly	4.0	7.6-2.9	NA
W-217-Ta-005	4.0	3.5-3.0	0-10K
W-218-Ta-006	4.0	3.7-3.2	0-5K

TABLE 4. INGOT ELECTRICAL AND DEFECT CHARACTERISTICS (Continued)

<u>Ingot Identification</u>	<u>TGT Resistivity (ohm-cm)</u>	<u>Actual Resistivity (ohm-cm)</u>	<u>Etch Pit Density (/cm²)</u>
W-219-V-008	4.0	3.6-3.3	0-5K
W-220-W-005	4.0	3.7-3.2	0.20K
W-221-Ni-005	4.0	3.5-3.1	OK
W-222-Ag-002	4.0	5.8-5.7	0-Gross Lineage
W-223-Ni-006	4.0	3.6-3.1	0-5K
W-224-HSC/DCS057	1.0	1.4-1.2	5-20K
W-225-Mn-009	4.0	5.5-3.5	0-5K
W-226-Mn-010	4.0	***	***
W-227-Cr-011-Poly	4.0	3.9-3.5	NA
W-228-Gd-001	4.0	Processing	Processing

* Not applicable

*** Single growth prohibited due to excessive impurity doping for permanence studies

TABLE 5. INGOT CARBON AND OXYGEN CONCENTRATIONS

<u>Ingot Identification</u>	<u>Carbon Concentration ($\times 10^{16}$ atoms/cm³)</u>	<u>Oxygen Concentration ($\times 10^{16}$ atoms/cm³)</u>
W-201-Mo-007/P	7.0	61
W-203-V-005/P	12	59
W-205-Fe-009/P	8.0	34
W-207-Mo-008	5.4	43
W-209-Ti-014	6.4	61
W-211-Cu-007	6.0	57
W-213-Pb-001	8.0	57
W-215-Mo-009-Poly	10.0	56
W-217-Ta-005	12.0	50
W-219-V-008	25.0	43
W-221-Ni-005	10.0	53
W-223-Ni-006	20.0	77
W-225-Mn-009	4.0	58
W-227-Cr-011-Poly	16.0	82

coupled with deep level transient spectroscopy and other characterization data, provides powerful tools to evaluate experimental silicon materials as they are developed, to identify critical impurities which may enter the process stream, and to suggest possible remedial action to the producer.

The first of these materials to be characterized as part of the Phase IV effort was silicon produced by Hemlock Semiconductor Corporation (HSC) under JPL Contract 955533.⁽⁵⁾ In this process, trichlorosilane is chemically redistributed to form dichlorosilane (DCS) which subsequently is decomposed to silicon by chemical vapor deposition. The projected advantages of the DCS process over conventional methods include higher conversion efficiencies and deposition rates while using less energy and expendable materials. The material is expected to resemble semiconductor grade silicon in form and purity, but at considerably reduced cost.

A bar of the HSC silicon was grown into a Czochralski crystal using the same furnace and growth conditions previously employed throughout this program. The ingot is designated W224-HSC/DSC-057 as noted in Table 1. The melt was doped to produce a nominal ingot resistivity of 1.5 Ωcm , slightly lower than the 4 to 6 Ωcm typical of other ingots we have studied.

Twenty-five wafers from ingot W224 were fabricated into solar cells, along with five 4 Ωcm baseline wafers from ingot W198. The standard process sequence we use includes an 825°C phosphorus diffusion to form an n + p cell. This typically produces uncoated devices with AM1 conversion efficiencies in the 9 to 10% range (12.7 to 14.3% with antireflective coatings) for 4 Ωcm material.⁽³⁾

In this first run cells from ingot W224 exhibited an uncoated efficiency of $9.13 \pm 0.75\%$ ($\sim 12.8\%$ coated) compared to $9.28 \pm 0.25\%$ for the baseline devices (the scatter for all the data in this run is higher than we usually observe and some evidence for impairment of junction quality was noted). The individual uncoated cell efficiencies for ingot W224 ranged from a high of 10.11% (14.2% coated) to a low of 7.9%

(11.1% coated), although the majority of the cell efficiencies clustered around 9%. As expected from the lower resistivity of ingot W224, the solar cells made on the DCS material exhibited higher open circuit voltages (average 0.571 mV) than those made on the baseline silicon (average 0.556 mV).

Gold-Ti-Si Schottky barrier diodes were fabricated on representative wafers from ingot W224 to facilitate deep level transient spectroscopy measurement.⁽³⁾ No deep levels were found in the DCS material, in keeping with its expected high purity and the fact that any metals present in the starting material would be segregated during crystal growth. The sensitivity of the DLTS method is about 10^{12} trapping centers per cubic centimeter at 1.5 Ω cm resistivity.

In general, our preliminary findings are that the silicon produced by the Hemlock Semiconductor dichlorosilane process is comparable in behavior to our standard Czochralski material made by trichlorosilane decomposition. We plan further analysis to verify these findings.

3.3 Evaluation of Impurity Behavior in Polycrystalline and Single Crystal Silicon by Spectral Response Measurements

In this quarter, we focussed on spectral response measurements to gain better insight into the effects of impurities and grain boundaries on solar cells. A detailed description of our spectral response measurement apparatus was given in the 17th quarterly report.⁽³⁾ The apparatus automatically compares the photocurrent from the cell to that from a standard detector of calibrated spectral response and plots the ratio of the two currents as a function of wavelength.

The spectral response from a large number of single crystal and polycrystalline cells was measured. Figure 1 shows the effects of 10^{15} cm^{-3} Cr, $4 \times 10^{12} \text{ cm}^{-3}$ Mo and $2 \times 10^{14} \text{ cm}^{-3}$ Ti on single crystal solar cells. These concentrations also typify the upper limit of the respective impurity concentrations which can be incorporated during Czochralski growth of single crystal silicon. It is quite clear that

Figure 1 The Effect of Cr, Mo, and Ti, on the Spectral Response of Single Crystal Silicon Solar Cells.

the presence of impurities degrades the red response of the solar cell. Since poor red response is a good indication of low bulk lifetime, the spectral response data are consistent with our I-V data, OCD lifetime measurement, and the model assumption.⁽³⁾ $2 \times 10^{14} \text{ cm}^{-3}$ Ti had the greatest impact while $1 \times 10^{15} \text{ cm}^{-3}$ Cr had the least effect on the spectral response, in excellent agreement with measured cell efficiencies, Table 6, (7.8% for Cr-04 and 4.2% for Ti-137). This supports our previous findings that impurity-induced degradation in the cell performance is due to reduction in bulk lifetime.

Figure 2 shows a comparison of the spectral response of uncontaminated single crystal solar cells with the cells containing $1.8 \times 10^{13} \text{ cm}^{-3}$ and $2 \times 10^{14} \text{ cm}^{-3}$ Ti. The data indicate that the red response, or the carrier lifetime, is gradually impaired with the addition of more Ti.

Figures 3 and 4 show the spectral response of single crystal and polycrystalline solar cells, with and without Ti-impurity. The data in the figures indicate that the presence of grain boundaries alone can degrade the spectral response, which is also seen as a loss in the cell efficiency (Table 6). Figure 3 shows that there is very little difference in the spectral response of single and polycrystalline cells containing $2 \times 10^{14} \text{ cm}^{-3}$ Ti. However, Figure 4 shows that this difference becomes appreciable when the Ti concentration is reduced by about an order of magnitude. This is because, at high Ti concentrations, the bulk lifetime is limited by Ti-induced deep levels and grain boundary influence becomes negligible ($\frac{1}{\tau} = \frac{1}{\tau_{\text{Ti}}} + \frac{1}{\tau_{\text{gb}}}$). At lower Ti concentrations, the impact of grain boundaries on the cell efficiency can be seen more readily. Table 6 shows that the difference between the efficiencies of Ti-contaminated single crystal (5.75%) and poly crystal (5.4%) also becomes more apparent at smaller Ti concentration, inspite of the somewhat higher Ti concentration in the single crystal. DLTS data in Table 7 (section 3.4) show that, within the experimental error of about a factor of 2, the electrical activity of Ti (~40%) is not appreciably altered by the presence of grain boundaries.

TABLE 6

Lighted I-V data from the solar cells used for Spectral
Response Measurements

Ingot ID	I _{SC} (mA)	V _{OC} (Volts)	FF	η (%)	τ_{OCD} (μ secs)
002-Baseline	22.4	.55	.76	10.03	4.5
076-Poly Baseline	19.23	.505	.66	6.90	1.06
209-Ti	16.01	.495	.67	5.75	.32
202-Ti-Poly	15.39	.486	.69	5.40	.48
137-Ti	12.63	.463	.68	4.2	.85
102-Ti-Poly	13.64	.449	.61	4.0	.64
139-Mo	18.43	.507	.68	6.82	.63
215-Mo-Poly	17.04	.493	.69	6.10	.45
004-Cr	18.61	.525	.755	7.80	1.05
216-Cr-Poly	18.61	.491	.571	5.5	-

Curve 725239-A

Figure 2 Change in the Spectral Response of Single Crystal Silicon Solar Cells with Ti Impurity Additions.

Figure 3 The Effect of $1.2 \times 10^{14} \text{ cm}^{-3} \text{ Ti}$ on the Spectral Response of Silicon Solar Cells.

Curve 725237-A

Figure 4 Effect of $2 \times 10^{13} \text{ cm}^{-3}$ Titanium in the Spectral Response of Single and Polycrystalline Solar Cells.

Figure 5 depicts the effect of Mo on the spectral response of single crystal and polycrystalline solar cells. The data clearly show that the polycrystalline cell containing $\sim 2 \times 10^{12} \text{ cm}^{-3}$ Mo exhibits degradation from grain boundaries as well as from the impurity. From the cell efficiency, Table 6, of uncontaminated polycrystalline cells (6.9%) and Mo-contaminated single crystal (6.8%), one would expect to see the influence of both Mo and grain boundaries because neither dominates the lifetime. Within the experimental error, we can say, from Table 7, that the electrical activity of Mo (100%) at these concentrations is not affected by the grain boundaries.

In Figure 6 we show the spectral response of uncontaminated, as well as Cr-doped, single crystal and polycrystalline cells. Again we see the effect of both the grain boundaries and impurity on the spectral response. It is important to note that there was a large scatter in the efficiency (2 to 6%) of Cr-doped polycrystalline cells due to the presence of inclusions so it is difficult to draw any conclusions. Schottky barrier diodes fabricated for DLTS measurements were also very leaky. Very limited data on only two diodes showed an active Cr concentration of only $7.5 \times 10^{12} \text{ cm}^{-3}$. Active Cr concentration in a single crystal containing 10^{15} cm^{-3} Cr was $\sim 10^{14} \text{ cm}^{-3}$. We require better cells and more DLTS data to confirm these observations. A preliminary examination of this ingot revealed partial crystal breakdown and inclusion formation. Based on these very limited observations one can speculate that Cr, being a fast diffuser, may form precipitates where the local Cr concentration exceeds the solubility limit. If these precipitates make some of the Cr electrically inactive, then we will see a smaller electrically active concentration in DLTS measurements. Finally, these precipitates may get into the depletion region to produce excess leakage, or even short the junction, which may account for the poor cells and large scatter observed. More work is in progress to test this hypothesis. In order to gain a better understanding of Cr in polysilicon, a new ingot (227) with much lower Cr concentration has been grown.

Figure 5 The Effect of Molybdenum on the Spectral Response of Single Crystal and Polycrystalline Silicon Solar Cells.

Figure 6 The effect of Cr on the Spectral Response of Single and Polycrystalline Silicon Solar Cells.

3.4 Silicon Material Evaluation by DLTS

We have continued to monitor the electrically active impurity concentration of representative wafers (as grown) and solar cells to correlate device and materials effects due to impurities. Table 7 is a summary of current information collected by deep level transient spectroscopy for single and polycrystalline ingots. As before, we have found no levels for Cu, and the Ta concentrations are too low to be detected in the as grown ingot. This compilation will be updated as the study continues.

3.5 The Permanence of Impurity Effects in Silicon Solar Cells

3.5.1 Temperature Effects

Experimental evidence ^(1,2) has shown that solar cell performance is dependent upon both material quality and device process history. Since the operating life of a solar cell array in the field is expected to exceed twenty years, it is important to understand what effects specific impurities will have on long term efficiencies.

The time period of interest is extremely long compared to practical test times, so extrapolations based upon accelerated aging tests must be used to estimate long term solar cell performance. On the basis of earlier results and fundamental considerations, temperature is believed to be the dominant parameter affecting gradual cell degradation. Thus a series of experiments is underway to determine the magnitude of the temperature-dependent degradation mechanisms for individual impurities.

3.5.1.1 Experimental Method.

Prediffused silicon wafers containing single impurities are soaked at elevated temperatures for various lengths of time. These wafers are then further processed into test cells and their characteristics are measured. The rate of degradation of efficiency is determined at each temperature, assuming that, at least in the initial stages, cell efficiency decreases linearly with time. A linear least squares fit to

TABLE 7

DLTS RESULTS ON IMPURITY-DOPED INGOTS

Ingot ID	Best Estimate of Metallurgical Impurity Concentration (N_M)	Active Impurity Concentration (cm^{-3})		$\frac{N_{TW}}{N_M}$
		In the Wafer N_{TW}	In the Cell (near junction) N_{TC}	
V-200-Poly	4×10^{14}	1.3×10^{14}	no data	0.32
Ti-202-Poly	2×10^{13}	1.12×10^{13}	1.15×10^{12}	0.56
V-203-Poly	4×10^{13}	1.7×10^{13}	undetectable	0.34
206-V	2.6×10^{13}	6.43×10^{12}	undetectable	0.25
207-Mo	2.0×10^{13}	2.2×10^{12}	9×10^{11}	1.1
208-Cr	1.9×10^{14}	3.91×10^{13}	undetectable	0.21
209-Ti	2.0×10^{13}	8.12×10^{12}	1.15×10^{12}	0.40
210-Ti	1.0×10^{14}	2.91×10^{13}	3.6×10^{12}	0.30
211-Cu	1.8×10^{15}	undetectable	undetectable	-
212-Cu	1×10^{16}	undetectable	undetectable	-
214-V-Poly	2×10^{14}	6×10^{13}	no data	0.30
215-Mo-Poly	2.5×10^{12}	4.5×10^{12}	no data	1.8
216-Cr-Poly	8×10^{14}	7.5×10^{12}	no data	.009
217-Ta	1.5×10^{11}	undetectable	no data	-
218-Ta	6.5×10^{10}	undetectable	no data	-
V-200-Poly	4×10^{14}	1.3×10^{14}	no data	0.32
Ti-202-Poly	2×10^{13}	1.12×10^{13}	1.15×10^{12}	0.56
V-203-Poly	4×10^{13}	1.7×10^{13}	undetectable	0.34

the efficiency-time data gives estimates of the η_0 (the initial efficiency) and $(1/\eta_0) d\eta/dt$ (the normalized rate of change of efficiency). From these parameters, determined for different temperatures, an Arrhenius plot is developed to express the temperature-time dependence of efficiency degradation for a particular impurity.

3.5.1.2 Experimental Results

The behavior of the ingots listed in Table 8 has been previously reported.^(3,7)

TABLE 8
INGOTS USED IN PERMANENCE STUDIES

Ingot Identification	Impurity Concentration (cm^{-3})
W09700000	Baseline
W077Mo001	4.2×10^{12} Mo
W123Ti008	1×10^{14} Ti
W072Cr005	4×10^{14} Cr
W192Ag001	2.2×10^{15} Ag
W183Nb002	$< 9 \times 10^{12}$ Nb
W135Fe005	7.8×10^{14} Fe
W166Fe007	1.06×10^{15} Fe
W167Nb001	$< 4.4 \times 10^{13}$ Nb

During this reporting period, a similar investigation of a copper-doped ingot was performed. Wafers from ingot 211Cu007, containing 1.8×10^{15} copper atoms per cubic centimeter, were temperature soaked at 400, 600, and 800°C for varying lengths of time, up to 200 hours. The efficiencies of cells made from this material were measured and tabulated.

Copper, a fast-diffusing impurity in silicon, was found to behave much like iron,⁽⁷⁾ another fast-diffusing species. That is, no consistent degradation of material properties due to copper is evident. This absence of cell efficiency degradation prevents an extrapolation

of expected behavior at room temperature. The experimental results are represented in Figure 7.

This type of behavior for copper was, in part, expected. Copper (and iron) impurity in silicon does not behave like most heavy metals which cause electronic levels within the forbidden gap. Such levels act as carrier recombination centers, decreasing carrier lifetime and thus reducing cell efficiency. Individual copper atoms in silicon apparently do not cause recombination centers to be formed. Instead, the primary effect of copper impurity is to increase leakage current across the p-n junction, perhaps by forming high resistance conducting paths. The impact on cell performance is thus statistical in nature.

The copper-contaminated cell data do not fit a simple reliability model. The absence of cell efficiency degradation at multiple elevated temperatures strongly suggests that no degradation would occur at normal operating temperatures.

3.5.2 Electrical Bias Effects

The purpose of this experiment is to determine whether electrical bias coupled with moderate temperatures in operating solar cells may, over long periods of time, affect cell efficiency. The possible interactions between electric field and impurities in the solar cell junction region are not well understood and their effects must be determined empirically.

In these experiments, base line cells and cells containing one of several representative impurities are operated at a current density of 30 mA/cm^2 ; current is externally supplied by a constant current supply. In order to enhance and accelerate any electric field/impurity interactions which may occur, the cells are operated at elevated temperature. Silicon materials incorporated in this study are listed in Table 9.

Figure 7 Relationship of efficiency to temperature and time for a copper-doped ingot.

Experimental cells must be metallized for this experiment. This limits the temperature at which tests can be made since it is recognized that cell operating parameters can be affected by electrode effects at fairly low temperatures.

TABLE 9

SILICON MATERIALS UNDERGOING ELECTRICAL BIAS TESTING

<u>Ingot No.</u>	<u>Impurity</u>
W-198-00-000 Baseline	None
W-166-Fe-007	Fe 1.06×10^{15}
W-167-Nb-001	Nb $<0.044 \times 10^{15}$
W-192-Ag-001	Ag 2.20×10^{15}
W-181-Cr-006	Cr 1.04×10^{15}
W-016-Fe-001	Fe 0.4×10^{15}
W-056-Cu-005	Cu 65×10^{15}
W-183-Nb-002	Nb $<0.009 \times 10^{15}$
W-123-Ti-008	Ti 0.105×10^{15}

Solar cell operation for periods of 100 hours have now been carried out at five different temperatures. The experiment plan calls for further tests at increasingly higher temperatures until definite evidence of cell efficiency degradation is observed.

Average cell efficiency of each cell group after each temperature-bias exposure is plotted in Figures 8 to 16. In order to eliminate any effects which are not due to specific impurities, these data have been normalized to the average performance of the base line cell group, and these data are shown in Figures 17 to 24.

Figure 8 Effect of electrical bias on relative efficiency as a function of stress temperature--base line cells.

Figure 9 Effect of electrical bias on relative efficiency as a function of stress temperature—Fe-doped ($1.06 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 10 Effect of electrical bias on relative efficiency as a function of stress temperature--Nb-doped ($< 0.44 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 11 Effect of electrical bias on relative efficiency as a function of stress temperature--Ag-doped ($2.20 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 12 Effect of electrical bias on relative efficiency as a function of stress temperature--Cr-doped ($1.04 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 13 Effect of electrical bias on relative efficiency as a function of stress temperature--Fe-doped ($0.4 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 14 Effect of electrical bias on relative efficiency as a function of stress temperature--Cu-doped ($65 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 15 Effect of electrical bias on relative efficiency as a function of stress temperature--Nb-doped ($< 0.009 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 16 Effect of electrical bias on relative efficiency as a function of stress temperature--Ti-doped ($0.105 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 17 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Fe-doped ($1.06 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 18 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Nb-doped ($< 6.044 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 19 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Ag-doped ($2.20 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 20 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Cr-doped ($1.04 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 21 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Fe-doped ($0.4 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 22 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Cu-doped ($65 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 23 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature-Nb-doped ($< 0.009 \times 10^{15} \text{ cm}^{-3}$) cells.

Figure 24 Effect of electrical bias on relative efficiency, normalized to base line cell behavior, as a function of stress temperature--Ti-doped ($0.105 \times 10^{15} \text{ cm}^{-3}$) cells.

Clearly, the degradation at temperatures up to 165°C is small, usually less than 10%. No systematic effect is observable in these data, so tests are continuing at higher temperatures.

3.6 Impurity Effects in High Efficiency Solar Cells

Throughout the investigation of solar cell impurity effects, we have relied on a conservative cell design and a minimally complex fabrication technology to reduce the hazard of process-dependent effects masking the impurity-dependent behavior. These devices, when made from baseline ingots with no added impurities, have an AM1 efficiency of $\sim 14.5\%$ with AR coatings. The experimental data agree well with an analytic model based on the impurity concentration dependence of the effective minority carrier diffusion length in the base. The impurity-dependent parameter used to characterize the impact of each impurity is its degradation threshold concentration (N_{ox}), above which cell performance is significantly reduced. The normalized short-circuit current (I_n) is then related to the concentration of impurity X as follows:

$$(I_{n\infty}/I_n - 1)^2 = C_1 (1 + N_x/N_{ox}) \quad (1)$$

where $C_1 = 0.0121$ and $I_{n\infty} = 1.11$ are model constants independent of impurity effects. The efficiency is related to the short-circuit current by:

$$\eta = \eta_0 (0.872 I_n^{1.128} + 0.128 I_n^{12}) \quad (2)$$

The degradation threshold is given by:

$$N_{ox} = \frac{D_{nb}}{L_{no}^2 v_{th} \sigma_x a_x} \quad (3)$$

Where D_{nb} is the electron diffusion constant in the p-base, L_{no} is the electron diffusion length, σ_x is the effective recombination cross-section for impurity X, v_{th} is the thermal velocity, and a_x is the ratio of

electrically active impurity centers to those metallurgically present. Of these, only σ_{xx} and a_x depend on the impurity, the rest are design-dependent. The effective diffusion length in the baseline devices, L_{no} , depends on the total recombination associated with the base region including its back surface. Thus, even if the intrinsic diffusion length of the base material were infinite, the effective diffusion length would remain finite and be controlled by recombination occurring at the back surface. In an ohmic-back device, as are our standard devices, the recombination at the back is very large and limits the effective diffusion length to a value somewhat less than the base thickness. Experimentally, we observe values of L_{no} from $\sim 160 \mu m$ to $\sim 250 \mu m$ for base thicknesses from $\sim 260 \mu m$ to $\sim 300 \mu m$.

High efficiency cells, above 16%, require much greater effective diffusion lengths, $L_{no} > 400 \mu m$. This necessitates careful processing and reduction of the surface recombination to maintain a large intrinsic base diffusion length. The impact of surface recombination can be reduced by means of back surface fields or by making the cell very thick. The latter approach is hardly a cost-effective method, but is of interest experimentally because a wide base device should conform accurately to the established impurity model analysis. However, the increased effective diffusion length will result in lowered degradation thresholds; that is, increased sensitivity to impurities, as indicated by Equation 3. Calculated performance for wide-base devices which are currently being fabricated is shown in Table 10 for four different intrinsic diffusion length cases.

The second high efficiency design uses back surface fields and possibly oxide surface passivation. It remains necessary to conduct the processing so as to preserve a large intrinsic diffusion length, and in accord with Equation 3, this again implies a lower degradation threshold. The effective diffusion lengths still control the device performance but the functional relationship will be different than that indicated by Equations 1 and 2. In order to examine the impurity relationships in these devices,

we have constructed a new model which characterizes the recombination properties within the device in terms of internal carrier velocities. These velocities depend only on the local electronic properties of the silicon, the surface recombination velocities at the physical surfaces of the cell, and the velocity at which light-generated carriers enter the depletion region.

The model, which includes heavy-doping effects, band-narrowing, Fermi-Dirac statistics, and impurity/mobility effects, is described in Reference (8) and in a related but differently applied model in Reference (9). The calculated internal recombination velocities for the ohmic-back devices of Table 10 are shown in Figure 25. The plotted values are obtained by transforming the velocities at the physical surfaces, across each local interior region successively, ending at the depletion layer boundaries. The model analysis shows that the open-circuit voltage is given by:

$$V_{oc} = V_T \ln \left(\frac{J_{sc}}{J_{on} + J_{op} + J_{dep}} \right) \quad (4)$$

where

V_T = the thermal voltage

$$J_{on} = q n_i^2 S_{cjp} / N_{AB}$$

$$J_{op} = q n_i^2 S_{cjp} / N_D (x_{tn})$$

$$J_{dep} = q n_i W_D / \tau_D$$

J_{sc} = the light-generated current

It is clear that the design objective is to obtain a minimum velocity at the depletion boundaries. In the ohmic-back devices, this occurs when a long diffusion length is obtained and the base-width is large.

TABLE 10

CALCULATED PERFORMANCE FOR OHMIC BACK WIDE-BASE SOLAR CELLS

4 ohm-cm, p-base width 150 μ m

Base Width (μ m)	Intrinsic Base Diffusion Length (μ m)	Effective Base Diffusion Length (μ m)	V _{oc} (mV)	V _{sc} (mA)	η (%)
280	209	182	554	35	15.0
"	467	250	563	36	15.8
"	660	264	564	37	16.1
"	1480	276	566	37	16.3
750	209	208	559	36	15.7
"	467	431	577	37	16.8
"	660	537	583	38	17.3
"	1480	692	590	38	17.5

Figure 25 Internal recombination velocity plots for ohmic back solar cells.

Table 11 and Figure 26 illustrate results obtained for narrow-base back-field structures. The performance of these devices exceeds that obtainable with the wide-base cells provided the large effective diffusion lengths can be preserved. This implies an even greater sensitivity to lifetime-killing impurities. Also shown are the results of using oxide surface passivation to lower the recombination velocity at the physical surfaces. Heavy-doping effects are seen as the rising velocity curves in the P+ and N+ regions for the case of typical diffusion profiles ($N_s > 10^{20}/\text{cm}^3$). The most efficient device shown was obtained by limiting the back-field concentration to $10^{19}/\text{cm}^3$.

Wide-base devices are currently being processed and processing experiments are in progress for the narrow-base back-field devices. The model analysis is in good agreement with experiment for presently available data and is expected to provide a sound basis for characterizing the impurity behavior in high-efficiency cells.

4. CONCLUSIONS

Experimental silicon made by the Hemlock Semiconductor dichlorosilane process (DCS) has been grown into a Czochralski crystal, wafered and fabricated into solar cells for evaluation as part of this program. The DCS solar cells produced efficiencies comparable to cells made on our standard 4 Ωcm baseline silicon, i.e. 12 to 14%, AM1. No deep levels were detected in the material by DLTS, a method capable of sensing 10^{12} traps cm^{-3} at 1.5 Ωcm resistivity.

Spectral response data indicate that both impurities and grain boundaries in polycrystalline cells degrade the long wavelength response of the devices. For high impurity concentrations the impurity effect dominates. Preliminary data suggest Cr-grain boundary interactions may occur.

Stepwise heat treating (up to 165°C) of impurity doped solar cells operating under a 30 ma/cm^2 bias have not yet shown any degradation effects which can be attributed to the impurities themselves. In a

TABLE 11

CALCULATED PERFORMANCE FOR NARROW BASE BACK-FIELD SOLAR CELLS

4 ohm-cm, p-base width 150 μ m

Intrinsic Base Diffusion Length (μ m)	Effective Base Diffusion Length (μ m)	V _{oc} (mV)	J _{sc} (mA)	η (%)	Notes
209	324	567	34	15.4	ohmic surface
467	630	587	37	16.9	ohmic surface
209	330	571	34	15.5	passive surface
467	1090	599	37	17.4	passive surface
209	336	573	35	15.6	passive surface
467	1470	605	37	17.8	reduced doping

Figure 26 Internal recombination velocity plots for back field solar cells.

parallel study accelerated aging of Cu-doped cells indicated only small and scattered degradation effects at temperatures to 800°C.

A mathematical model describing impurity effects in high efficiency cells (wide base ohmic back contact and narrow base back field types) indicates that the impurity concentration will be low in these devices if high performance is to be achieved.

5. PROGRAM STATUS

The current milestone chart for the Phase IV program is illustrated in Figure 27.

5.1 Present Status

During this quarter we

- Evaluated the first experimental silicon material made by the dichlorosilane process.
- Examined the effects of impurities in single and polycrystalline solar cells by spectral response and DLTS measurements.
- Completed accelerated aging on Cu doped silicon solar cells.
- Completed low temperature bias tests of silicon solar cells doped with 7 different impurities.
- Developed a mathematical model of impurity effects in high efficiency devices.

5.2 Future Activity

During the next quarter work will continue on (1) polycrystalline cell behavior (2) thermochemical interaction of impurities in silicon (3) permanence studies and (4) impurities in high efficiency devices.

PHASE IV PROGRAM PLAN (SCHEDULE)

57

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