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LSA LARGE AREA SILICON SHEET TASK CONTINUOUS
LIQUID FEED CZOCHRALSKI GROWTH

Quarterly Report, January—March 1979

MASTER

By
Dorothea Walters

April 1979

Work Performed Under Contract No. NAS-7-100-954886

Siltec Corporation
Menlo Park, California

U.S. Department of Energy

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Solar Energy

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QUARTERLY REPORT #6

January - March 1979

Project Manager: George Fiegl

Prepared by: Dorothea Walters

April 1979

SILTEC CORPORATION

Menlo Park, California

The JPL Low-Cost Silicon Solar Array Project is sponsored by the U.S. Department of Energy and forms part of the Solar Photovoltaic Conversion Program to initiate a major effort toward the development of low-cost solar arrays. This work was performed for the Jet Propulsion Laboratory, California Institute of Technology by agreement between NASA and DOE.

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ABSTRACT

Siltec Corporation's contract with JPL is directed towards the design and development of equipment and processes, to demonstrate continuous growth of crystals by the Czochralski method, suitable for producing single silicon crystals for use in solar cells. Continuous growth is defined as the growth of 100 kg of single silicon crystal, 10 cm in diameter, from one container. Siltec's approach to meeting this goal is to develop a furnace with continuous liquid replenishment of the growth crucible, accomplished by a meltdown system and a liquid transfer mechanism, with associated automatic feedback controls.

Several test runs were performed during the final quarter of Phase I with the main aim of achieving 100 kg of ingot growth from one container. The best results accomplished during these demonstration runs were 70 kg of ingot produced in 70 hours.

Three significant limitations were experienced during this period and are briefly outlined below.

The major limiting factor in achieving monocrystalline growth for large portions of processed material was the silicon monoxide build-up on the furnace interior. Oxide particles from deposits throughout the furnace chamber frequently entered the melt surface after only 12 to 15 hours of furnace operation, disturbing the solid/liquid interface. As a result, only 10% of the material produced was monocrystalline.

Batch recharging, a manually-performed function, influenced overall furnace efficiency considerably, and reduced the lifetime of the meltdown crucible drastically. This was due primarily to the required thermal cycling.

ABSTRACT (cont'd)

Transfer tube heater reliability prevented full achievement of 100 kg throughput in one run. The heating element, in close contact with the quartz transfer tube, transformed into SiC over a period of 60 to 70 hours. This ultimately led to the destruction of the heater element.

The experience gained in the last three months has shown clearly that by overcoming these major limitations, continuous CZ can produce structured material as well as meet the economic goals as set forth by JPL.

The main thrust in follow-up work will be devoted toward overcoming these limitations.

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1.0 INTRODUCTION

The overall purpose of this program, which began in October 1977, is the demonstration of continuous crystal growth by the Czochralski method. "Continuous" is defined as a throughput of silicon that produces 100 kg of single crystal material, 10 cm in diameter, from one common container.

The program includes:

- . Design and fabrication of a basic CZ furnace with a second chamber to be used for melting of the polysilicon.
- . Design, development, and fabrication of the continuous liquid silicon feed system with automatic feedback controls.
- . Demonstration of continuous crystal growth, as defined above, with an after growth yield >80%.
- . Definition of process control variables for continuous Czochralski growth.
- . Theoretical analysis of silicon sheet growth to be considered during design and development of the furnace.

During the last quarter, the main effort was primarily directed towards achieving 100 kg throughput runs, and further development of the following areas:

- . Reduction of SiO₂ deposition in the growth furnace
- . Recharging of the meltdown crucible
- . Liquid transfer tube system

2.0 RESULTS

2.1 Transfer Tube

The transfer tube displayed a tendency toward melt solidification at both ends. To remedy the situation, each end of the transfer tube verticle shell was thermally isolated through physical separation from the rest of the tube. This step has proven quite successful in reducing this tendency. In addition, the cross section of the heating element was reduced to increase the temperature in that area.

As a follow-up, several end-to-end thermal profiles of the transfer tube were made under power. The temperature was found to be quite uniform, with two minor cold areas. The ends of the tube were cooler than the rest of the tube. This may be attributed to their not being immersed in molten silicon during the profiling (see Figure 1).

The design of the tube heater, formed of strips of Grafoil, was modified to compensate for the cold areas, and to reduce the number of joints. With regard to heater lifetime, further improvements were made by using high purity silica tape for electrical insulation. The low purity tape used in previous runs became electrically conductive in the temperature range of 1420°C after 20 to 25 hours, so that arcing of the heating element to the electrically conductive graphite felt would occur.

The graphite shells which encapsulate the entire exterior of the transfer tube system, and in particular the end caps near the melt, showed considerable deterioration after 50 to 60 hours of exposure to operating conditions (see Figure 2).

Deterioration of the quartz transfer tube ends was visible after 70 hours of melt contact (see Figure 3). The dissolution rate

Transfer Tube (cont'd)

of quartz is highly sensitive to temperatures above 1450°C as well as the relative velocity between the quartz and the melt.

Due to the low velocity of the melt through the transfer tube, only a small diameter increase, after 70 hours continuous operation, was measured.

2.2 Recharging

As originally envisioned, the CLF Furnace was to use batch recharging of the meltdown crucible for all growth demonstrations under Phase I. This was to be done as a time overlap operation during crystal withdrawal. This operation was successfully performed but is severely affecting the efficiency of the CLF Furnace. Some of the constraints with this method are:

- . The transfer tube needs to be withdrawn from the melt during a batch recharge to avoid uncontrolled melt transport into the growth crucible and requires repriming afterwards.
- . The recharge and meltdown cycle combined are longer than the crystal cooldown, therefore up to 2 hours of the total cycle time are added. Multiple meltdown cycles lead to an early breakdown of the quartz liner causing melt contamination.

Recognizing these limitations, continuous recharging was explored and was successfully demonstrated by dropping wafer fragments into the meltdown crucible through a tubular chute. No obvious functional problems were encountered. The wafer fragments formed a floating island in the middle of the crucible and did not interfere with the transfer operation, nor with the growth of the crystal.

Recharging (cont'd)

On this initial attempt, the rate of melting was not in complete balance with the rate of solidification of the growing crystal. A more sophisticated heat input control system will eliminate this difficulty. However, it should also be noted that wafers are not the ideal shape in that the area of heat loss is effectively equal to the area of heat gain.

We conducted additional recharging trials over the remainder of the quarter. Generally speaking, these trials were limited to the techniques used in conventional CZ growth (e.g., tongs).

2.3 Crystal Lift Mechanism

Early this quarter, a new lift mechanism for the S.S. bellows was designed, fabricated and installed, forming the cooldown chamber for the ingot. A cleaning rack and the associated procedure for the bellows was devised.

2.4 Melt Level Sensing System

The reflected laser melt level sensing system became operational early this quarter. The system output is currently to a chart recorder, but eventually it will be integrated into a closed-loop crucible lift control.

2.5 Argon Consumption/SiO Deposition

New argon purge inlets, with divergent nozzles, were installed in the viewports. The high argon usage of the CLF Furnace was an available method of holding the SiO deposition to a reasonable level, significantly reducing the SiO accumulation on the viewport windows.

3.0 CONCLUSIONS

3.1 Transfer Tube

The cost of materials and time of construction of the present transfer system design are all too high to warrant suitability for production. The present design has been developed to a fairly high degree of perfection, but it still has too short a lifetime. Taking this into consideration, it is obvious that an alternate approach is needed.

The goal of 100 kg material throughput in one run is achievable and was only limited by the lifetime of the transfer tube heating element. A reusable heater of rigid construction will be needed to achieve reliability and cost effectiveness.

3.2 Recharging

In all tests we have run to date, a set of tongs were used to lower the silicon lumps into the meltdown crucible. This is by far the simplest form of solid recharging. A continuous solid recharge mechanism is needed to fully utilize the advantages of the CLF Furnace and in order to avoid quartz breakdown of the meltdown crucible due to thermal cycling.

3.3 SiO Deposition in Furnace Interior

SiO particles coming from the transfer tube vertical shell in the growth chamber and from the furnace areas above the growth crucible were the major cause of preventing monocrystalline growth for a large portion (90%) of the grown material. To achieve high quantities of structured material, control over SiO deposits in the furnace interior must be obtained. Operation under low pressure will be required to eliminate SiO interference with monocrystalline growth.

3.4 Equipment Control

The CLF Furnace, as presently configured, is almost entirely manually controlled. While this has been suitable for demonstrating technical features of the system, many of the functions of the furnace must be handled automatically before technical readiness can be achieved.

4.0 PROJECTION OF ACTIVITIES

In the final quarter of this contract, we are pleased to report that the majority of the goals set forth in Phase I were accomplished. Several runs, with good results, were conducted, and three major areas for further development were identified.

Plans are to renew this contract in April 1979 and initiate Phase II of the program. Projections are currently being compiled toward this new goal.

APPENDIX I

Figures

1 2 3 4

Figure 1: Thermal Profile of Transfer Tube.

Figure 2: Pictured above is a graphite end cap, which fits over the end of the transfer tube, viewed after 60 hours exposure to normal operating conditions. Note the build-up of silicon carbide and resultant deterioration of the cap.

Figure 3: An example of deterioration of the quartz transfer tube ends is shown above. This erosion occurred after 70 hours of melt contact. Silicon melt solidification is also visible in the tube.

Figure 4: Cross-Section of Transfer Tube

Figure 4: This figure depicts a mid cross-section of the standard quartz transfer tube.

This area of the tube was not submerged in the silicon melt, and as a result did not experience the eroding effect normally seen on the end which is constantly immersed in the melt.

Reasons accountable for the reduced amount of erosion of the inner diameter of the tube are primarily that melt was at a lower velocity in this portion of the transfer tube. The rate of dissolution of the quartz tube is highly dependent on the relative velocity between melt and quartz as well as the temperature of the melt. Higher velocities and velocity fluctuations of the melt lead to a boundary layer reduction and breakdown, thereby increasing the dissolution rate.

Note as well that after 70 hours continual use, there was no melt solidification clogging the tube. This was a result of increasing the temperature in the heating element, and therefore lessening the tendency for the melt to freeze.

Figure 5: Cross-Section of Crucible

Figure 5: Pictured in this figure is a cross-section of a quartz crucible (meltdown side) following 70+ hours use in growing silicon crystals.

Note the "banding" on the inside of the crucible (right side), depicting the build-up of silicon monoxide (SiO), and therefore indicating the variations and changes in melt level during the melting/crystal growth process. The darker the band, the denser the presence of SiO . This contaminant build-up is the largest limiting factor in achieving monocrystalline growth.

The "spotting" toward the bottom of the crucible is primarily pitting caused by the molten silicon. High heat intensity as well as the interaction of the chemical properties of the molten silicon itself with the quartz take a definite toll on the crucible. This can also be seen in the difference in thickness; the portion of the crucible actually in continuous contact with the melt shows signs of erosion. The crucible wall is definitely thinner toward the base.

APPENDIX II

New Technology

No new technology was generated during this reporting period.

APPENDIX III

Summary of Characterization, Design, and Economic Analysis Data

APPENDIX III

ECONOMIC ANALYSIS FOR CONTINUOUS CZOCHRALSKI

The SAMICS/IPEC model was applied to the input data, which are outlined in Table I, for the purpose of a cost estimate of the continuous Czochralski process in its present state of development and for the optimized CLF furnace.

TABLE I

Parameter	Present State CLF - Furnace	Optimum CLF - Furnace
Furnace run size	105 Kg	102 Kg
Crystal diameter	100 mm	150 mm
Crystal growth velocity	100 mm/h	83.4 mm/h
Crystals per run	5	3
Crystal pull yield	95%	98%
Crystal pull weight	100 Kg	100 Kg
Crystal usable after grinding	90.9 Kg	90.27 Kg
Crystal yield	86.5%	88.5%
Run cycle time	74.3 h	52 h
Pullers per operator	2	3
Direct labor cost	\$5.50/h	\$6.50/h
Operating supplies cost	\$956/run	\$567/run
Equipment cost	\$100,000	\$160,000
Equipment floor space	150 ft. ²	150 ft. ²
Equipment utilization	85%	90%
Power requirements	5646 KWH/run	3952 KWH/run
Silicon/slice conversion 1982	20	20
Silicon/slice conversion 1986	25	25

All dollar related parameters are based on 1978 dollars.
Add on costs will be converted back to 1975 dollars using IPEG inflation factors (see price allocation guidelines LSA Project 5101-68).

Economic Analysis for Continuous Czochralski

For the determination of crystal add on cost per IPEG model the following equation is applied:

$$\text{Cost} = (C_1 * \text{EQPT} + C_2 * \text{SQFT} + C_3 * \text{DLAB} + C_4 * \text{MATS} + C_5 * \text{UTIL}) / \text{QUAN}$$

Numerical values for overhead factors per IPEG (see Appendix C5101-33) are:

$$C_1 = 0.49$$

$$C_2 = 97.0$$

$$C_3 = 2.1$$

$$C_4 = 1.3$$

$$C_5 = 1.3$$

Elements of the direct manufacturing cost were determined for present state and optimum version of CLF furnace.

TABLE II

Elements	Present State CLF - Furnace	Optimum CLF - Furnace
EQPT = equipment cost	\$100,000	\$160,000
SQFT = space requirements	150 ft. ²	150 ft. ²
DLAB = annual direct labor	\$ 20,379	\$ 16,934
MATS = annual materials cost	\$ 95,504	\$ 85,730
UTIL = annual utilities cost	\$ 22,617	\$ 23,966
QUAN = annual output 1982	8,999 m ²	13,230 m ²
QUAN = annual output 1986	11,248 m ²	16,539 m ²

CONTINUOUS CZOCHRALSKI ECONOMIC MODEL
CRYSTAL GROWTH OPERATING SUPPLY COSTS

TABLE III

Item	Cost / Run	
	Present State	Optimum
Quartz crucible (2 each)	\$300.00	\$300.00
Graphite susceptor	16.00	16.00
Pedestal	1.50	1.50
Graphite heater	15.50	13.50
Outer heat shield	3.00	2.25
Bottom heat shield	1.20	1.20
Top heat shield	.80	.80
Electrode sleeve	1.00	1.00
Heater leg	1.50	1.50
Quartz spacer	1.00	1.00
Screw, heater leg	.50	.50
Purge tube	4.50	4.50
Seed chuck	1.50	1.50
Seals, crucible shaft	2.00	4.00
	<hr/> \$350.00	<hr/> \$349.25
Transfer tube	180.00	60.00
Transfer tube heater	5.00	15.00
Transfer tube insulation	20.00	--
Outer graphite shell	12.00	--
Temperature sensor	10.00	1.20
	<hr/> \$227.00	<hr/> \$ 76.20
Vacuum pump oil	2.00	15.00
Seed	5.00	3.00
Argon	372.00	124.00
	<hr/> \$379.00	<hr/> \$142.00
Total	\$956.00/run	\$567.00/run

Economic Analysis for Continuous Czochralski (continued)

PROCESS CYCLE TIME

TABLE IV

Operation	Present CLF (Min.)	Optimum CLF (Min.)
<u>Melt-Down/Preparation</u>		
Load silicon	20	20
Close furnace	15	15
Pump down and back fill	10	10
Transfer system heat-up/melt-down	<u>100</u>	<u>100</u>
	145	145
<u>Prime Transfer Tube</u>		
Stabilize melt and lower seed	15	15
Set crucible positions, prime tube	<u>5</u>	<u>5</u>
	20	20
<u>Crystal Growth</u>		
Neck	20	20
Shoulder	60	120
Body	586	525
Taper	<u>60</u>	<u>120</u>
5 cycles Optimum 3 cycles	726	785
<u>Recharge</u>		
Add poly and melt-down	90	--
Cool crystal	60*	120
Remove crystal	15*	30
Prime transfer tube	<u>20</u>	<u>--</u>
4 cycles Optimum 2 cycles	110	150
*Over lap		
<u>Shut-down/Cleaning/Set-up</u>		
Cool-down	60	120
Remove crystal	15	30
Clean furnace	120	120
Set-up furnace	<u>30</u>	<u>30</u>
	225	300
Total Cycle Time	74.3 h	52.0 h

Economic Analysis of Continuous Czochralski (continued)

1. ANNUAL FURNACE RUNS

Parameter	Present	Optimum
Run cycle time	74.3 hrs.	52 hrs.
Equipment utilization	85%	90%
Annual runs	99.9	151.2

2. ANNUAL DIRECT LABOR

Parameter	Present	Optimum
Furnaces/operator	2	3
Hourly rate	\$5.50	\$6.50
Labor \$/run	\$204.00	\$112.00
Annual direct labor	\$20,379.00	\$16,934.00

3. ANNUAL OPERATING MATERIALS

Parameter	Present	Optimum
Materials/run	\$956.00	\$567.00
Annual materials	\$95,504.00	\$85,730.00

4. ANNUAL UTILITIES COST

Parameter	Present	Optimum
Energy cost	\$.040/KWH	\$.040/KWH
Energy/run	5646 KWH	3953 KWH
Annual utilities	\$22,617.00	\$23,961.00

Economic Analysis of Continuous Czochralski (continued)

5. ANNUAL CRYSTAL OUTPUT

Parameter	Present	Optimum
Crystal produced/run (100% yield)	573.5 cm	247.6 cm
Slices/run (20 slices/cm)	11,470	4,952
Slices/run (25 slices/cm)	14,337	6,190
Output m ² /yr. 1982	8,999.5 m ²	13,230.6 m ²
Output m ² /yr 1986	11,248.9 m ²	16,539 m ²

Economic Analysis for Continuous Czochralski (continued)

PRESENT STATE OF DEVELOPMENT CLF

ADD ON COST CRYSTAL 1982

$$\text{COST} = (C_1 \cdot \text{EQPT} + C_2 \cdot \text{SQFT} + C_3 \cdot \text{DLAB} + C_4 \cdot \text{MATS} + C_5 \cdot \text{UTIL}) / \text{QUAN}$$

$$= (0.49 \cdot 10^5 + 97 \cdot 150 + 2.1 \cdot 20,379 + 1.3 \cdot 95,504 + 1.3 \cdot 22,617) / 8,999$$

$$= 28.88; 100\% \text{ crystal yield } \$28.88/\text{m}^2 \quad 1978 \text{ \$}$$

86.5% crystal yield <u><u>\$26.60/m²</u></u>	1975 \$
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PRESENT STATE OF DEVELOPMENT

ADD ON COST CRYSTAL 1986

86.5% crystal yield <u><u>\$21.30/m²</u></u>	1975 \$
---	---------

OPTIMUM CLF 1982

$$\text{COST} = (0.49 \cdot 1.6 \cdot 10^5 + 97 \cdot 150 + 2.1 \cdot 16,934 + 1.3 \cdot 85,730 + 1.3 \cdot 23,961) / 13,236$$

$$= 14.6 \text{ 100\% crystal yield } \$14.6/\text{m}^2 \quad 1978 \text{ \$}$$

88.5% crystal yield <u><u>\$13.18/m²</u></u>	1975 \$
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OPTIMUM CLF 1986

88.5% crystal yield <u><u>\$10.47/m²</u></u>	1975 \$
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APPENDIX IV

Milestones

PROGRAM PLAN PHASE I

Revised: 6-8-78

Milestone Ledger:

⊙ = Due Date
▼ = Start/Finish
→ = Program Revision

*Plan for Phase III