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LSA LARGE AREA SILICON SHEET TASK, CONTINUOUS
LIQUID FEED CZOCHRALSKI GROWTH

Quarterly Report No. 9 for October—December 1979

By
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January 1980

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

Siltec Corporation
Menlo Park, California

MASTER

U.S. Department of Energy

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CONTINUOUS LIQUID FEED CZOCHRALSKI GROWTH

QUARTERLY REPORT #9

October - December 1979

Program Manager: George Fiegl

Prepared by: Dorothea Walters

January 1980

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

Now nearing the end of its second phase, the purpose of this program is the design and development of equipment and processes to demonstrate the continuous growth of crystals, by use of the Czochralski method, suitable for producing monocrystalline silicon for use in solar cells. This involves the growth of at least 150 kg of monocrystalline silicon ingots, 150 mm in diameter, obtained from a single growth container. Our approach to meeting this goal is to develop a furnace with continuous liquid replenishment to the growth crucible.

The major objectives for the new melt transfer system were accomplished this quarter. This included a reduction in the overall dimensions of the transfer tube system in order to accommodate 150-mm diameter ingot growth from 12-inch diameter crucibles, and the design, development and fabrication of a reusable transfer tube heater. Due to a lack of available space for thermal insulation in the vertical section of the transfer tube, primarily a result of the larger rigid graphite heater and the smaller outer shell, we experienced some difficulty in achieving the proper temperature profile over the melt transfer path.

Fabrication of the lump/particle feed mechanism, designed to adapt to the gate valve of the meltdown chamber, was completed and installed during the month of December. This system is capable of supplying polycrystalline particles to the meltdown chamber in a continuous mode in such a manner that the average mass introduced into the meltdown chamber is in balance with the solidification rate in the growth chamber. Melt replenishment with simultaneous crystal growth need not be interrupted while the polysilicon supply is replenished in the storage container.

A preliminary economic analysis, using the IPEG model, has been performed. With unlimited melt replenishment, the most significant accomplishment in reducing crystal add-on costs can be achieved by reducing the number of crystal sections pulled (i.e., pulling 150 kg crystals). In support of this time- and cost-saving effort, various experiments have confirmed that crystal weights as great as 150 kg can be sustained by the "necked down" portion of the crystal seed.

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1.0 INTRODUCTION

The overall purpose of this program, which began in October 1977, is to demonstrate continuous crystal growth via the Czochralski method; "continuous" being the throughput of silicon producing 150 kg of single crystal material, 150 mm in diameter, from one common container.

The program under Phase II includes:

- . Furnace conversion for low pressure operating conditions.
- . Design, development and fabrication of continuous solid feed systems for silicon, in various forms, for the meltdown chamber.
- . Design, development and fabrication of the continuous liquid silicon feed system with a reusable heater and 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 this past quarter, activities were directed primarily toward:

- . The final stages in the design, fabrication and installation of a new melt transfer system.

INTRODUCTION (Cont'd)

- . Installation of the polyrod feed mechanism. This was completed, permitting continuous recharging of the meltdown chamber.
- . Several runs from 5 to 12-kg charges were performed. Some were in order to establish process parameters for conventional CZ growth of 150 mm ingots, while others were to establish the successful repeatability of melt transfer.
- . Improvements were made to the melt level sensor.
- . A new improved automatic diameter control (ADC) system was installed.
- . A silicon crystal neck strength evaluation was performed.
- . Finally, wrapping up this quarter, a demonstration run using the new, redesigned transfer tube system was conducted under reduced pressure.

2.0 RESULTS

2.1 Growth Under Low Pressure

Crystal growth continued this quarter under a low pressure environment. All ingots produced were 150 mm in diameter. Silicon monoxide buildup in the furnace interior has ceased to be a problem following various modifications made last quarter. As a result, yield in monocrystalline ingots has increased significantly.

In order to establish process parameters for conventional CZ growth of 150 mm ingots, several runs from 12-kg charges were performed. Solidification rates of 2.7 to 3.5 kg/hr were achieved. Of the total material grown, 96% was monocrystalline in nature. This series of runs provided a control group for future runs utilizing continuous melt replenishment.

2.2 Transfer Tube System

A crucible 12 inches in diameter was chosen primarily to minimize the power requirements for the hot zone. This crucible diameter has also proven to exhibit excellent thermal stability while growing ingots 150 mm in diameter. In order to accommodate ingots of this size diameter, the overall diameter in the vertical section of the transfer tube system was reduced from 2.5 to 2.0 inches (Figure 1).

The design of the graphite heating element must take into consideration the mechanical strength requirements for both the assembly of the transfer system as well as the thermal stresses the element is subjected to during the cooldown process following the completion of a run. In addition, the applied voltage to the heater must be kept at a low level to avoid arcing, which adds considerably to the required cross section of the heating element.

Figure 1. Detail drawing of newly redesigned transfer tube shell with silicon carbide coating; vertical leg.

The available space left between the heater and the 1.75-inch I.D. of the outer shell is minimal, barely accommodating the least amount of thermal insulation material required. For this reason, it was necessary to increase the heater temperature via additional energy dissipation by modifying the shape of the heater -- specifically, the critical area between the melt surface in the crucible and the tip of the heating element where melt solidification problems have been experienced. Figures 2 and 3 depict two designs, both accomplishing the required temperature increase; however, the design shown in Figure 3 is less fragile.

Thermal profiles were made with the use of platinum and rhodium thermocouples, moving the junctions in small increments of 0.25 and 0.5-inch through the tube. This method, however, could only be applied when temperatures were kept below 1400°C -- otherwise, moving the temperature probe would be difficult if not impossible due to fusion of the quartz tube and the aluminum insulator. Fixed thermocouple junctions in critical areas of the tube (at the end and near the electrodes) would permit measurement of actual operating temperatures which are above 1400°C. The profiles obtained are shown in Figure 4.

Figure 2. Initial modification on rigid graphite heater for the transfer tube. This design proved too fragile.

Figure 3. Second, and preferred, design of rigid graphite heater.

Figure 4. Illustrated are the results of four separate thermal profiles recently performed on the transfer tube system. Readings A, B and C were registered by pulling a thermocouple through to the exterior of the transfer tube, thus plotting the path and recording data every quarter or half inch along the length of the tube; Reading D was performed by installing a multijunction array within a thin-wall tube, taking the readings at the corresponding points identified in this drawing by the dots within the tube. Method D consists of several different readings over an extended period of time, displaying small ranges of variance. This drawing shows the high and low figure recorded for each point.

Reading A used the rigid graphite heater as originally designed, while Reading B was recorded following the first modification to the heater ends. Readings C and D were performed with the most recent modification, when the previous design proved too fragile.

Transfer Tube System (Cont'd)

A rapid drop-off in temperature near the end of the tube can be seen in this diagram. With the main heater at operating temperature and the tube end submerged in molten silicon, the actual temperature gradient in this region is less severe. The critical area, consisting of approximately 0.375 to 0.5-inch of tube length (unheated) between the melt surface and the heater tip, must be above 1420°C in order to avoid melt solidification.

The area around the electrodes shows a major increase in temperature resulting from increased contact resistance. Generally speaking, the horizontal section of the tube temperature is controlled by the transfer tube heater while temperatures in the vertical sections are determined to a large extent by the hot zone environment of the growth and meltdown chambers. In the previous transfer tube design, the tube temperature was controlled entirely through the tube heater, possible through the unrestricted use of thermal insulation.

The difficulties encountered with the new design of the transfer tube system were mainly a result of all internal components being confined to an extremely small space.

Figure 5. Rigid graphite heater.

An important consideration of melt transfer technology is precise temperature control of the quartz transfer tube within 5° to 10°C above solidification temperature. This requirement is met through resistance heating by two graphite heating elements connected in series, and with a power input of 2 kw. The above cross section demonstrates one view of how these two heating elements fit into the overall transfer tube system.

2.3 Continuous Solid Replenishment

2.3.1 Polyrod Feed Mechanism

The polyrod feed mechanism is fully installed, and the system is ready to test. The only delay is a result of the problems associated with the transfer tube.

The control system for the polyrod feed turret was installed and demonstrated in November. This new system enables us to rotate, raise and lower the polyrod at adjustable velocities. Manual control switches and adjustable dials were installed in a small subconsole located on the furnace frame within easy reach above the meltdown chamber, and is connected to control units located within the main console.

We are fully prepared to demonstrate continuous solid replenishment of molten silicon following the test runs with the new transfer tube system.

2.3.2 Lump/Particle Feed Mechanism

In addition to the already existing method of polyrod feed to the meltdown chamber, introduction of lumps, some up to 50 grams in weight, is also under investigation. The design and fabrication of this lump feed system were completed during this quarter (see Figures 6 and 7). This particular method has the advantage of feeding polysilicon in processed pieces direct from the supplier as well as being able to feed polycrystalline material obtained from large polyrods containing substantial fractures (in contrast to the direct use of whole polyrods, where it is essential these rods be crack-free).

As with the polyrod feed mechanism, the lump feed system is designed so that melt replenishment occurs concurrently with meltdown and crystal growth. The length and diameter of the

Figure 6. Photograph of installed continuous lump/particle feed mechanism positioned over the meltdown chamber of the CLF-CZ furnace.

Lump/Particle Feed Mechanism (Cont'd)

tube connecting the two crucibles is dimensioned so that a critically dampened system is obtained whereby lump introduction into the meltdown crucible will not cause any detrimental changes in the melt level of the growth crucibles; thus, crystal growth will be unaffected. It has been calculated through simulated tests that monocrystalline growth will not be impaired by a rise of the melt level in the growth chamber as will occur during the admittance of a 50-gram polysilicon lump into the meltdown chamber.

Figure 7. Illustration of continuous lump/particle feed process within CLF furnace system.

Lump/Particle Feed Mechanism (Cont'd)

The crystal growth process will not be interrupted by a reloading of the storage hopper in that the meltdown crucible will contain sufficient molten silicon to continue the replenishment during the reloading.

Lump/particle feeding by this method has the disadvantage of bringing polysilicon in contact with metals, thereby potentially increasing the impurity level.

2.4 Melt Level Sensor

Improvements were made to the circuitry in the melt level sensor in order to eliminate any electrical noise problems as well as to achieve a more effective melt level control system. The furnace is currently using this improved circuit in breadboard form while boards and packaging are on order for a final unit.

2.5 Automatic Diameter Control (ADC)

A new 4-mode automatic diameter control system has been installed. This replaces the L&N standard 3-mode control unit which served as a substitute until the new 4-mode unit was put into operation. The new control system features adjustable time constants, permitting better control of large diameter growth. All basic modifications have been completed, and trim adjustments are in progress.

2.6 Crystal Neck Strength Evaluation

An experiment was designed to determine the tensile strength of a silicon crystal neck. As we will soon be growing crystals of substantial weight, we wanted assurance in the knowledge that the "necked down" portion of the seed would support weights in excess of 100 kg.

Crystal Neck Strength Evaluation (Cont'd)

With this thought in mind, a seed crystal was suspended from the seed lift mechanism into the furnace chamber. Weights were slowly added to a pail (suspended from a hook emanating from a steel plate epoxied to the base of the ingot) until a total weight of 229 pounds was amassed. Neither the crystal neck nor the seed chuck failed (Figure 2).

Figure 8. Illustration of crystal neck strength evaluation process.

Various published data on similar experiments indicate that the typical silicon crystal neck should be capable of supporting at least 1,000 kg in steady weight.

Although we did not carry this experiment through to the point of actual separation, we did conclude that the silicon neck can in fact bear 100-kg ingots without tearing the thinned down portion of the seed apart.

2.7 Demonstration of Melt Transfer

The most significant event occurring this quarter was the demonstration of melt transfer utilizing the redesigned transfer tube and rigid graphite heater.

Although the amount of time involved in actual melt transfer activity during this run was relatively short, a great deal of useful information was gleaned; the most important being the occurrence of melt solidification at the tube ends indicating the need for an increase in temperature at these points. We have made some minor modifications to the rigid graphite heater (in-house) with the intent of distributing more heat to the tube ends (refer to Figures 2 and 3), and will follow this adjustment with additional thermal profiles, thus recording the effects of this step in comparison to those data collected from previous thermal profiles conducted to date.

As a result of this action, the next attempt at melt transfer should prove a far smoother experience. (See the CONCLUSIONS portion of this report for further detail.)

3.0 CONCLUSIONS

3.1 Demonstration of Melt Transfer

Considering that the main objectives for the new transfer tube system are a reduction in the overall dimensions in order to permit the growth of 150 mm ingots from a 12-inch diameter crucible, and the use of a rigid heater construction suitable for reuse in several transfer runs, the melt transfer demonstration was successful in that both these goals were met.

Difficulty arose, however, in maintaining uniform temperature distribution along the path of melt transfer, manifesting itself in the form of melt solidification near the tube ends. This change in the temperature profile was primarily caused by the sacrifice of thermal insulation, necessitated by an increase in the heater dimension and a reduction in the outside diameter of the transfer tube system.

A modification to the critical areas on each end of the heater, increasing the energy dissipation as well as creating a more efficient thermal insulator to this area, is expected to bring the temperature up to the required range, thus achieving unimpeded continuous melt flow.

4.0 PROJECTION OF ACTIVITIES

4.1 Solid Recharging

During the latter portion of the quarter, both the polyrod and lump/particle feed mechanisms were installed on the CLF furnace. It is our intent to demonstrate both methods of solid recharging in order to achieve continuous melt replenishment to the growth crucible early next quarter.

4.2 Transfer Runs

With the advent of our first demonstration run using the redesigned transfer tube system, we are confident of displaying several additional runs following the planned alteration to the rigid graphite heater ends.

4.3 Program Plan

In nearly all respects, activities continued in the pattern suggested in the Program Plan. It is our intent to maintain this schedule of activities.

APPENDIX I

New Technology

No new technology was generated during this reporting period.

APPENDIX II

Summary of Characterization, Design, and Economic Analysis Data

No characterization or design data were generated this quarter.
The summary of economic analysis data follows.

ECONOMIC ANALYSIS

With a continuous melt replenishment supply, the only major constraint in achieving minimum crystal add-on costs is the number of crystal sections necessary to process 150 kg of ingot. The crystal add-on cost is directly proportional to the number of ingots pulled. Crystal sections, when separated from the melt (this is particularly true with 150 mm ingots), require a round-off. This process serves to avoid substantial material losses resulting from thermal stresses which manifest themselves during the slicing operation in the form of fractures. These round-off procedures are extremely time-consuming and require a fair amount of skill. Therefore, in order to optimize the CLF-CZ process in economic terms, the ultimate goal must be the production of one crystal. A single crystal of the size suggested here should present no difficulty as tensile strength tests on the "necked down" portion of the seed under simulated operating conditions showed that weights of 100 kg, and most likely 150 kg, can be sustained.

ECONOMIC ANALYSIS FOR CONTINUOUS CZOCHRALSKI GROWTH

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.

Table I

Parameter	
Furnace run size	160 kg
Crystal diameter	150 mm
Crystal growth velocity	100 mm/hr
Crystals per run	3
Crystal pull yield	98%
Crystal pull weight	156.8 kg
Crystal usable after grinding	137.6 kg
Crystal yield	86%
Run cycle time	56.85 hrs
Pullers per operator	4
Direct labor cost	\$7.00/hr
Operating supplies cost	\$403.05/run
Equipment cost	\$160,000
Equipment floor space	150 ft ²
Equipment utilization	90%
Energy requirements	5200 kwhr/run
Silicon/slice conversion 1982	20 slices/cm
Silicon/slice conversion 1986	25 slices/cm

All dollar figures are in 1980 dollars

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 new IPEG model are:

$C_1 = 0.52$	10 year life for the equipment
$C_2 = 109$	Equipment plus operators
$C_3 = 2.8$	Direct labor without fringe benefits
$C_4 = 1.2$	Material
$C_5 = 1.2$	Utilities

Elements of the direct manufacturing cost were determined as follows:

Table II

Elements		
EQPT = equipment cost	=	\$160,000
SQFT = space requirements	=	150 ft. ²
DLAB = annual direct labor	=	\$ 13,797
MATS = annual materials cost	=	\$ 55,895
UTIL = annual utilities cost	=	\$ 31,536
QUAN = annual output	=	19,082.4 Kg

CONTINUOUS CZOCHRALSKI ECONOMIC MODEL
CRYSTAL GROWTH OPERATING SUPPLY COSTS

Table III

Item	Cost/Run
Quartz crucible (2 each)	\$ 260.00
Graphite susceptor	9.00
Pedestal	1.50
Graphite heater	10.50
Outer heat shield	2.25
Bottom heat shield	1.20
Top heat shield	0.80
Electrode sleeve	1.00
Heater leg	1.50
Quartz spacer	1.00
Screw, heater leg	0.50
Purge tube	2.00
Seed chuck	1.00
Seals, crucible shaft	1.00
	<u>\$ 293.25</u>
Transfer tube	40.00
Transfer tube heater	4.00
Transfer tube insulation	0.50
Outer graphite shell	1.50
Temperature sensor	0.80
	<u>\$ 46.80</u>
Seed	3.00
Argon	60.00
	<u>\$ 63.00</u>
TOTAL COST PER RUN	<u><u>\$ 403.05</u></u>

Economic Analysis for Continuous Czochralski (continued)

PROCESS CYCLE TIME

Table IV

Operation	
<u>Melt-down/Preparation</u>	
Load silicon	40
Close furnace	10
Transfer system heat-up/melt down	<u>130</u>
	180 180
<u>Prime Transfer Tube</u>	
Stabilize melt and lower seed	15
Set crucible positions, prime tube	<u>15</u>
	30 30
<u>Crystal Growth (3 crystals)</u>	
Neck	15 x 3 = 45
Shoulder	90 x 3 = 270
Body	657 x 3 = 1971
Taper	120 x 3 = <u>360</u>
	2646
<u>Recharge</u>	
Cool crystal	120 x 2 = 240
Remove ingot	30 x 2 = <u>60</u>
	300
<u>Shut-down/Cleaning/Set-up</u>	
Cool-down	120
Remove ingot	30
Clean furnace	120
Set-up furnace	<u>30</u>
	300 <u>300</u>
	3411 min
TOTAL Cycle Time	56.85 hrs

ADD ON COST FOR CONTINUOUS CZOCHRALSKI

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

$$= (0.52 \times 1.6 \times 10^5 + 109 \times 150 + 2.8 \times 13,797 + 1.2 \times 55,895 \\ + 1.2 \times 31,536)/19082.4$$

$$= 12.74/\text{Kg for 86\% crystal yield}$$

Crystal add-on cost: \$12.74/Kg for 86% crystal yield

for 20 slices/cm - 1982 goal

Crystal add-on cost: \$14.85/m² in 1980 \$

for 25 slices/cm - 1986 goal

Crystal add-on cost: \$11.88/m² in 1980 \$

APPENDIX III

Milestones

TASK DESCRIPTION	PERFORMANCE SCHEDULE									
	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	JAN	
1. Modify the liquid feed (CLF) furnace										
A. Overall design	▲	▲	▲							
B. Reduced pressure operation 10 to 30 torr	▲	▲	▲							
C. Polycryst feed mechanism with feedback control system	▲	▲	▲							
D. Extend crystal lift mechanism to 60 inches above gate valve	▲	▲	▲	▲	▲	▲	▲			
2. Transfer tube system										
A. Design and development	▲	▲	▲	▲	▲					
B. Fabrication		▲	▲	▲	▲	▲				
3. Develop particle feed system										
A. Overall design (mechanical and electronic)	▲	▲	▲	▲	▲	▲				
B. Fabrication				▲	▲	▲	▲	▲	▲	
4. Growth demonstrations										
A. Growth of one crystal 25 kg from one set-up with melt transfer					▲					
B. Growth of 4 crystals 25 kg mass each from one set-up with melt transfer						▲				
C. Growth of one crystal 50 kg mass from one set-up with melt transfer							▲			
D. Growth of 3 crystals 50 kg mass each from one set-up with melt transfer									▲	
5. Economic analysis							▲		▲	
6. Provide representative samples						▲	▲	▲	▲	
7. Support design and performance reviews			▲	▲						
8. Provide support personnel										
A. Project integration meetings, DOE-sponsored meetings & annual workshop				▲				▲		
B. Performance review meetings			▲			▲			▲	
9. Provide documentation	▲	▲	▲	▲	▲	▲	▲	▲	▲	
10. Procure parts, material and services (as required)										

Milestone Legend: ▲ = Due Date; ▼ ▲ = Start-Finish