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

Quarterly Report No. 4, July–September 1978

By
George Fiegl

September 1978

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

Siltec Corporation
Menlo Park, California

U.S. Department of Energy

MASTER

Solar Energy

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CONTINUOUS LIQUID FEED CZOCHRALSKI GROWTH

QUARTERLY REPORT #4

July - September 1978

George Fiegl

September 1978

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 melt-down system and a liquid transfer mechanism, with associated automatic feedback controls.

Fabrication of all furnace parts was completed during the past quarter. Mechanical assembly of the furnace was accomplished and all electrical work completed. The furnace underwent final systems testing and is now ready for test runs. Two crystals from 15 Kg charges were grown by conventional CZ technique.

Simulated transfer of molten silicon was performed using a liquid of approximately the same kinematic viscosity as that of molten silicon. Each end of the transfer tube was submerged about one-half inch below the melt surface and filled with liquid through pressure differential. To create sufficient energy difference between the two containers to overcome the friction losses, and to generate the required replenishment rate of .24 cc/sec, the melt level of the melt-down crucible was raised approximately 0.6 mm above the melt surface of the growth crucible.

A study was made to determine the effects of a stationary 7 mm O.D. quartz tube submerged into the molten silicon in the rotating crucible during the growth of a 2-inch diameter crystal. The presence of the tube had no negative effects on the growth of the crystal, nor was any mechanical disturbance in the melt noticeable.

The design of the transfer tube was completed. It consists of a 1-inch O.D. and 8 mm I.D. quartz tube with grafoil heater sections spiralled around the periphery of the tube. The entire system is insulated by graphite felt and is encapsulated by a graphite tube. The thermocouple and electrode connections are located close to the mounting flange.

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1. 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 directed towards the fabrication and assembly of the CLF furnace. The 12 inch hot zone was evaluated and several 15 Kg crystals were grown.

INTRODUCTION (continued)

A study was made to determine the effects of a stationary quartz tube placed into molten silicon while a crystal was grown.

Simulated transfer of molten silicon was performed and the design of the transfer tube was finalized.

2.0 RESULTS

2.1 FURNACE ASSEMBLY AND TEST RUNS

The fabrication of all furnace parts was finished during the last quarter. Mechanical assembly of the furnace was accomplished and all electrical work was completed. (See Appendix 1 pages 11 through 14, Figures 1, 2, 3, 4.) In spite of considerable delivery delays on the part of the furnace parts, we were able to meet our program plan of demonstrating the growth of crystals by conventional CZ process.

This was made possible by extensive testing of the new 12 inch hot zone in our R & D furnace prior to its use in the CLF furnace. The thermal characteristics of the new hot zone were found to be stable and highly reproducible. (See Appendix 1 Page 15, Figure 5.) Two crystals were grown from 15 Kg charges in the CLF furnace with an after-growth yield of 85%. A typical crystal grown with the new hot zone is shown in Appendix 1 page 16, Figure 6.

2.2 SIMULATED TRANSFER OF MOLTEN SILICON

Simulation of transfer of molten silicon was performed using water at 90°C which has the same kinematic viscosity as molten silicon at 1450°C ($0.3 \times 10^{-6} \text{ m}^2/\text{sec}$). (See graphs, Appendix 1 Page 17, Figures 7 and 8.) The set-up of the test is shown in Figure 9, Appendix 1 page 18. Two 1800 ml beakers were used to simulate the growth and the melt-down crucible. The melt-down crucible was placed on an adjustable height pedestal. Both containers were filled with 1000 ml of wafer at 90°C and heated with infrared lamps to maintain the desired viscosity. The transfer tube was simulated by using polyethylene tubing 5 mm I.D. and 32 inches in length. A third beaker was used to allow simulation

Results

of the solidification rate of a 4-inch diameter crystal with a growth velocity of 4 inches/hour. The simulated solidification rate of 0.24 cc/sec. was achieved by generating a 1/2 inch head differential between the melt surface of the growth crucible and the discharge point of the 12 inch long, 2 mm I.D. tube. The main transfer tube between the two crucibles was initially filled by pressure differential. It was demonstrated that the melt level in the simulated growth crucible stayed constant, while liquid could be drained at a rate of 0.24 cc/sec. This was accomplished by raising the simulated melt-down crucible at a rate directly proportional to the ratio of the solidification rate of the simulated crystal to the area of the melt surface of the melt-down crucible. The required head differential between the two crucibles was 0.6 mm which was sufficient to overcome the hydrodynamic losses. A sudden change in melt level position in the melt-down crucible (1 inch head differential), would lead to a melt level equilibrium after 25 seconds. The damping characteristics of the system are high enough to eliminate the possibility of melt level oscillation.

Using a sensing device to detect the melt level position in the growth chamber, the experienced time constant during the transfer would allow a rapid melt level adjustment in the melt-down system so that the melt level in the growth chamber can be kept constant. This aspect is of importance with regard to feeding raw material into the melt-down crucible while the melt is being transferred and a crystal is being grown. This approach is a very simple way to accomplish melt transfer.

Results

2.3 EFFECTS OF STATIONARY QUARTZ TUBE IN MELT

A study was made to determine the effects of a stationary 7 mm O.D. quartz tube submerged into the molten silicon in the rotating crucible during the growth of a 2 inch diameter crystal. (See Figure 10, Appendix 1 page 19. The tube was approximately 1/4 inch distant from the growing crystal and its end was 1/2 inch below the melt surface. A very light film of SiO₂ deposition was formed on the tube approximately 4 1/2 inches above the melt surface. The tube was inserted through the laser port following melt-down of a 5 Kg charge. The presence of the tube in the melt created no mechanical disturbance of the melt surface nor was any negative effect on the growth of the crystal observed.

The possibility of the creation of mechanical or thermal disturbances in the rotating crucible by the presence of the quartz tube in the melt was of major concern. The possibility of any solidification of silicon on the outside surface of the tube is reduced by the external heating of the transfer tube.

2.4 HEATING ELEMENTS

Possible alternative heating elements to the 1/2 inch wide, 15 mils thick grafoil heating elements were tested. Molybdenum wire 25 mils in diameter was coiled around the entire length of a 3 feet X 1 inch O.D./8 mm I.D. quartz tube. The heating coil was subdivided into 3 sections so that only portions of the tube could be heated. The entire heated length was embedded in brick mortar 1 inch thick. After the brick mortar had cured

Results

for 36 hours at 300°F, the entire assembly was placed into the 120 mm diameter quartz test tube and purged with argon. The heating element was brought up to an operating temperature of 1450°C. Over a period of 20 hours the molybdenum wire gradually transformed into molybdenum trioxide causing the conductive cross section to be reduced to a value where melting of the wire occurred. The source of oxygen has not been determined. The possibility of oxygen being enclosed in the brick mortar during the molding process is very likely and can be avoided by placing the transfer tube into a vacuum before bringing to operating temperature. The best results to date were achieved using grafoil which, under the same conditions as described above, did not show any change in the electrical characteristics of the heating element.

2.5 TRANSFER TUBE

The preliminary design of the transfer tube was completed. It is a single piece of 1 inch O.D. quartz tube with a 8 mm inside diameter. The entire transfer length is approximately 36 inches (a 12 inch horizontal section and two vertical 12 inch sections, one in the growth chamber, and one in the melt chamber). The heating element is made out of two pieces of grafoil 15 mils thick and 1/2 inch wide, wound in a double helix around the periphery of the tube. Molybdenum metal strips are placed approximately in the center of the heater to form electrode connections so that two parallel heater loops are formed. Thermal insulation is provided through graphite felt which is encapsulated with graphite tubing. Thermocouples are placed at the end of the discharge side of the transfer tube, to assure proper temperature of the

Results

replenished melt. The mounting flange is placed approximately in the center of the horizontal section, permitting the formation of a sufficient seal to achieve the required pressure differential for transfer of molten silicon.

2.6 ELECTROMAGNETIC THROTTLE VALVE

The electromagnetic throttle valve was tested under simulated conditions. Graphite rods were used because the electrical conductivity of molten silicon and graphite are in the same range. Graphite rods, 7 mm diameter and 12 inches long were placed into the quartz tube and vertically suspended by a rubber band. The working coils were placed in close proximity to the tube. The displacement of the graphite rod in the tube measured, and the imposed force derived from a force-displacement chart obtained previously from the suspension system for the rod.

These experiments demonstrated that the electromagnetic throttle valve is functional and effective in maintaining precise replenishment control. However, it has been determined experimentally that the same results can be accomplished more simply by utilizing the principle of inter-connected containers which dispenses with the need for any throttling action on the molten silicon.

3.0 CONCLUSIONS

- 3.1 The new 12-inch hot zone has proven capable of growing crystals 6-inches in diameter and is characterized by high reproducibility of its thermal conditions.
- 3.2 By applying the physics of inter-connected containers, the melt level in the growth chamber can be kept constant. This is accomplished by raising the melt level in the melt-down chamber at a rate equal to the ratio of the solidification rate (by volume), and the area of the melt surface in the melt-down system. This approach simplifies melt transfer considerably.
- 3.3 A stationary quartz tube of 7 mm O.D. submerged one-half inch into molten silicon in a rotating crucible and placed vertically within one-quarter inch of a growing crystal, has no negative effects on the growth process.

4.0 PROJECTION OF ACTIVITIES

Activities during the coming quarter will be directed towards actual melt transfer. Melt-down, transfer of molten silicon, and crystal growth will be demonstrated.

APPENDIX I

Figure 1. Layout drawing for the continuous liquid feed furnace.

Figure 2. The continuous liquid feed furnace.

Figure 3. Dismantled continuous liquid feed furnace.

Figure 4. Siamese chamber lift and tilt machine.

Figure 5, 12" hot zone for CLF furnace.

Figure 6. Typical crystal pulled with 12" hot zone.

Silicon Viscosity (From Baum, Gel'd, and Kocherov, Russian Metallurgy, I, 27, 1967)

Figure 7.

VISCOSITY OF WATER
Hydromechanic, Rodl, 1958, p. 79

Figure 8.

Figure 9. Test set up for simulated transfer of molten silicon with hot water.

Figure 10. 7 mm quartz tube placed in close proximity to growing crystal

TRANSFER TUBE ASSEMBLY

Figure 11.

APPENDIX II

NEW TECHNOLOGY

No new technology was generated during the reporting period.

APPENDIX III

Summary of Characterization, Design, and Economic Analysis Data

There was no new data generated during this quarter to
analyze.

APPENDIX IV

Milestones

Labor Summary

Cost Summary

PROGRAM PLAN

Revised: 6-8-78

Milestone Ledger:

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

Previous Total	1495
May 1978 Total	755
Total to Date	2250

Man-Hours

LSA LARGE AREA SILICON SHEET TASK
CONTINUOUS LIQUID FEED CZOCHRALSKI GROWTH

PLAN VERSUS ACTUAL MAN HOURS

*Per Baseline Cost Estimate, Revision #2 (as of December 14, 1977)

Project Costs

Previous Total	\$ 74768.54
May 1978 Total	• 25557.29
Total to Date	\$100325.83

LSA LARGE AREA SILICON SHEET TASK
CONTINUOUS LIQUID FEED CZOCHRALSKI GROWTH
PLAN VERSUS ACTUAL EXPENDITURES

*Per Baseline Cost Estimate, Revision #2 (as of December 14, 1977)