5101-207
Flat-Plate
Solar Arrav Project

DOE/JPL-1012-70
Distribution Category UC-63b

Advanced Czochralski Silicon Growth Technology for Photovoltaic Modules

Taher Daud
Akaram H. Kachare

September 15, 1982

Prepared for
U.S. Department of Energy
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The JPL Flat-Plate Solar Array Project is sponsored by the U.S. Department of Energy and is part of the Photovoltaic Energy Systems Program to initiate a major effort toward the development of cost-competitive solar arrays.

This publication reports on work done under NASA Task RD-152, Amendment 66, DOE/NASA IAA No. DE-AI01-76ET20356.

ABSTRACT

Large-area silicon sheet growth is one of the important elements of photovoltaic modules. In order to reduce the cost of these modules, a number of silicon sheet growth approaches encompassing both the ingot and the ribbon technology have been developed. Advancement of the Czochralski (Cz) growth method has been one of these approaches because of its existing broad technical base.

Several economic analyses had indicated that large-diameter, multiple ingot growth using a single crucible with melt replenishment would be required for Cz growth to be economically viable.

Based on the results of these analyses, two liquid and two solid feed melt replenishment approaches were initiated. The sequential solid feed melt replenishment approach, which demonstrated elements of technical feasibility, is described in detail in this paper. Growth results of multiple ingots (10-cm-diameter, totaling 100 kg; and 15-cm-diameter, totaling 150 kg weight per crucible) are presented. Solar cells were fabricated and analyzed to evaluate the effects of structure and chemical purities as a result of multiple growth. The results indicate that, with semiconductor-grade silicon, feedstock impurity build-up does not seem to degrade cell performance. For polycrystalline cells, the average efficiencies are 15 to 25% lower than those of single crystalline cells. Concerns regarding single crystal yields, crucible quality and growth speed are indicated, and present status and future research thrusts are also discussed.

ACKNOWLEDGMENT

The authors wish to thank Dr. R. L. Lane of Kayex Corporation for providing some of the data and for helpful discussions. They also wish to express their gratitude to Drs. K. M. Koliwad, M. H. Leipold, and J. K. Liu for their encouragement, support and suggestions for improvement of the report. They further appreciate the help of Catherine Edwards in editing the manuscript.

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SECTION I

INTRODUCTION

The Flat-Plate Solar Array (FSA) Project was started at the Jet Propulsion Laboratory (JPL) in 1975 as part of the U.S. Department of Energy's Photovoltaic Program. At that time (Reference 1) terrestrial photovoltaic modules using 5.0-cm (2-in.)-diameter single crystal silicon wafers were priced at about \$30 per peak watt (W_p).[1]

The objective of the FSA Project was to develop technology to meet the module price goal of \$0.70/W_p. At that price level photovoltaic technology was perceived to be competitive in the domestic energy market. This cost reduction required innovative approaches for all elements of the module, including that of silicon sheet growth. Silicon sheet is the key element of the module, as well as that of the technology itself.

Among various options of silicon sheet growth, the Czochralski (Cz) technique, primarily used by the semiconductor industry, is capable of producing a superior quality of crystalline silicon ingot. However, it is expensive and yields ingots that must be sliced into wafers for solar cell processing. Cast silicon, comparatively less expensive, also needs slicing. Growth of silicon sheet in the form of ribbon eliminates the cost of slicing and conserves material because of the absence of material loss (kerf) in slicing. However, the resulting sheet may suffer in quality compared with Cz-grown material. Parallel with the development of these new techniques of ribbon and cast silicon sheet growth, the advancement of Cz technique was pursued.

Economic analyses of Cz technique (Reference 2) showed that an increase in throughput was necessary, requiring a larger-diameter ingot growth (10-cm diameter and larger). In addition, major cost drivers were identified as crucible, furnace parts, and other consumable materials. Therefore, an investigation of multiple ingot growth from a single crucible through replenishment of silicon melt during growth was initiated. It was estimated that an increase in throughput from 20 to 100 kg of 10-cm (4-in.)-diameter ingots per crucible would lower the ingot price from \$7/W_p to \$1.60/W_p. This reduction, along with comparable cost reduction in other elements of the module, would result in a module price reduction from \$30/W_p to \$2.80/W_p.

Because of the initial technical success of the Cz program and continued updating of economic analyses, the technology was advanced from a throughput of 100 kg/crucible and 10-cm (4-in.)-diameter ingots to 150 kg/crucible and 15-cm (6-in.)-diameter ingots. However, an increase in throughput rate from 1 to 2.5 kg/h was required to lower the add-on ingot growth price to \$14/kg. This achievement in ingot growth technology, parallel with advancements in low-cost slicing of silicon, would then be commensurate with the project goal of \$0.70/W_p. This is equivalent to a \$27.4/m^2 (\$0.193/W_p) add-on price of ingot growth and slicing, each sharing 50% of that cost. Table 1 lists the technical features required to achieve the price goals of \$2.80/W_p and \$0.70/W_p, respectively.

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[1]All prices are in 1980\$.

Table 1. Advanced Czochralski (Cz) Growth Features for Technical Readiness

Features	Goals	
	$2.80/$W_p$	$0.70/$W_p$
Ingot diameters (cm)	10	15
Output/crucible (kg)	100	150
Growth rate (kg/h)	1.8	4
Throughput rate (kg/h)	1.4	2.5
Ingot yield (single crystal) (%)	95	90
Encapsulated cell efficiency (% AM1)	14.3	14.3
Automation	---	Full

This report describes the technique of Cz ingot growth in Section II. Section III describes the advanced Cz growth approaches as pursued under this program. A sequential solid feed melt replenishment Cz growth is further described in detail in Section IV. A brief economic analysis is presented in Section V, and material characterization with results is given in Section VI. Present Cz technology status and concerns as well as future research activities are summarized in Sections VII and VIII, respectively. Concluding remarks and a summary are presented in Section IX.

SECTION II

CZOCHRALSKI (CZ) INGOT GROWTH TECHNIQUE

The Czochralski (Cz) crystal growth technique, as generally practiced in the semiconductor industry today, consists of dipping a single-crystal, oriented silicon seed in a melt and pulling a cylindrical, single-crystal ingot 15 to 25 kg in weight and 3 to 4 inches in diameter. Normally, the melt is contained in a quartz crucible 10 to 12 inches in diameter, with a graphite susceptor around it. Heating is achieved by electrical resistance elements surrounding the susceptor. The ingot quality is determined by such growth parameters as proper thermal profile, clean ambient of inert gas at reduced pressures, attainment of optimal convective melt flow, and purity of furnace and crucible parts. The important rate-limiting parameter for the growth of ingots is the dissipation rate of the latent heat of fusion (L) of silicon at the solid-liquid interface. This dissipation rate, along with the transfer of heat to the crystal from the melt, occurs through the body of the crystal itself, as illustrated in Figure 1.

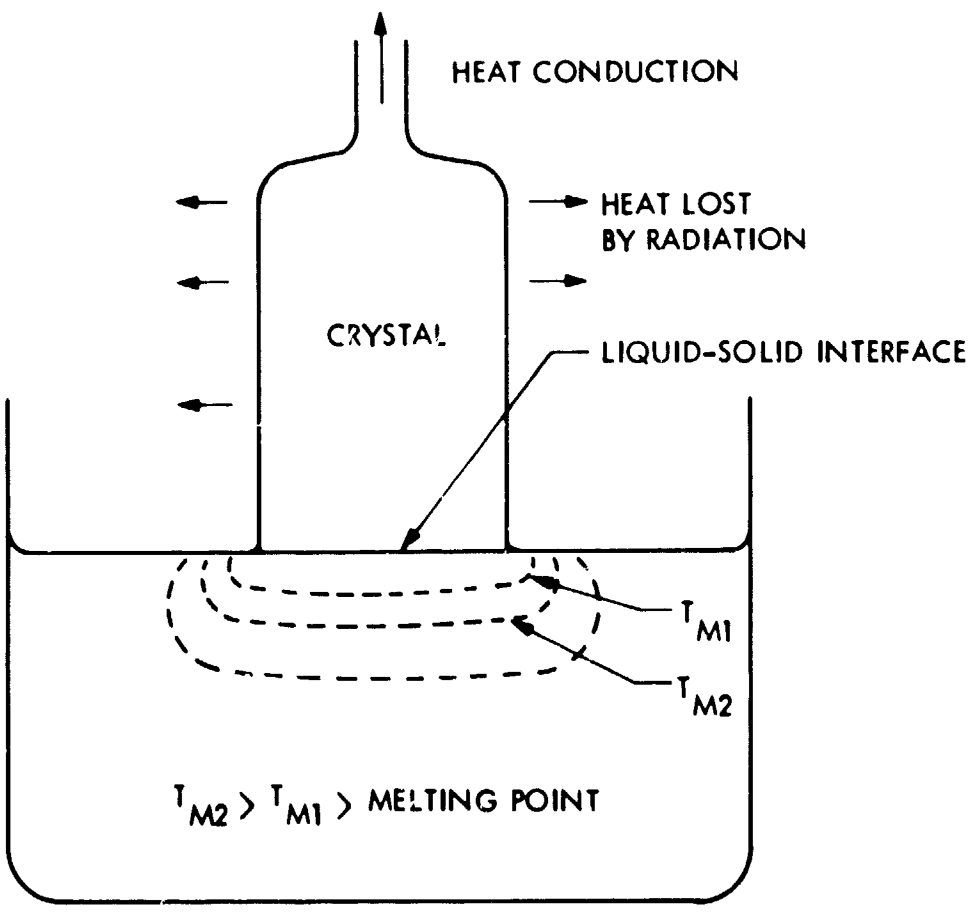

Figure 1. Heat Transfer during Czochralski (Cz) Growth

Thus, the heat transfer relationship for the growth can be written: heat of freezing + heat transferred from the melt = heat received by the crystal.

This equation is expressed as:

$$L\, V_p\, \delta A_s + k_\ell \frac{dT}{dx_\ell} A_s = k_s \frac{dT}{dx_s} A_s \qquad (1)$$

This equation neglects the slight drop in the melt level in the crucible during growth, where V_p is the growth velocity, δ the crystal density, A the area of cross-section, k thermal conductivity, dT/dx the temperature gradient: and, the subscripts ℓ and s refer to liquid and solid, respectively.

For maximum pull rate (V_{pmax}),

$$\frac{dT}{dx_\ell} = 0$$

and

$$V_{pmax} = \frac{k_s}{L\delta} \frac{dT}{dx_\ell} \qquad (2)$$

This relationship with simplifying assumptions gives V_{pmax} as inversely proportional to the square root of the ingot diameter. Based on these assumptions, various authors have obtained a range of V_{pmax} values. Figure 2 compares their models (References 3 through 7), showing wide variations depending upon the assumptions made.

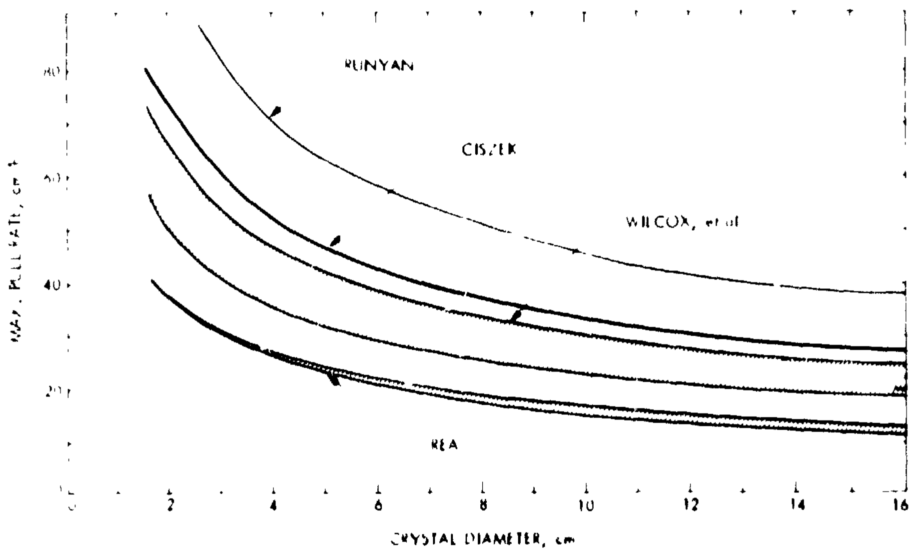

Figure 2. Maximum Pull Rates versus Crystal Diameter

Maintenance of constant melt level in the crucible during growth, as against decreasing level, may increase the pull rate for a 12-cm-diameter crystal from about 13.8 cm/h to 17.7 cm/h (Reference 2). Similarly, higher V_{pmax} is possible with lower ambient gas temperature, with helium giving the higher velocity, compared to argon and vacuum, in that order. Increasing crucible diameter from 20 cm (8 in.) to 40 cm (16 in.) may increase pull rate from 17 cm/h to 18.4 cm/h. Further improvements in pull rate are expected by reduction of linear emittance or faster heat removal, such as might occur with the use of cooling coils around the growing ingot.

The use of an automatic diameter control (ADC) maintains a stable thermal profile and aids in stabilizing growth, which is achieved by controlling the input power to the heating elements with a feedback loop through a liquid-solid interface temperature probe and pull rate monitor.

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SECTION III

ADVANCED CZOCHRALSKI (CZ) GROWTH APPROACHES

Originally, four approaches toward multiple ingot growth and melt replenishment were considered. Two of these approaches involved continuous silicon feed during growth and two were based on batch recharging after the growth of each ingot without a cool-down of the crucible.

A. CONTINUOUS FEED SILICON

1. Continuous Liquid Feed (CLF) Furnace (Reference 8)

This approach by Siltec Corporation consisted of melting silicon in a separate chamber and transferring it to the growth chamber. Figure 3 is a schematic of this CLF furnace showing an inverted U-shaped transfer tube with heating elements to transport the silicon melt continuously during growth from the melt crucible to the growth crucible. This new approach of continuously transferring the melt during growth led to several design changes of the transfer tube. The main difficulty was freezing of the melt in the tube. After a number of iterations of the heating element design, as much as 55 kg of melt transfer was demonstrated. The largest ingot grown with melt transfer weighed 65 kg and was 15 cm in diameter.

After completion of the contract, the growth furnace was delivered to JPL for installation and use in various in-house growth experiments.

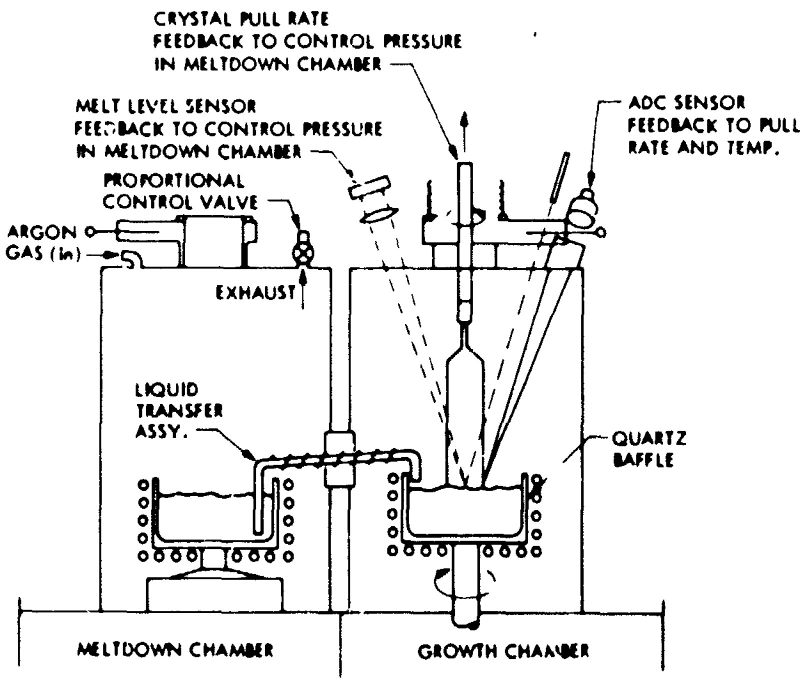

Figure 3. Basic Elements of the CLF Furnace

2. Continuous Nugget Silicon Feed

This approach by Texas Instruments consisted of transporting the nugget silicon into an auxiliary crucible, then melting and transferring the melt into the growth crucible (Reference 9). Figure 4 shows a feed hopper with auger transport system attached to a Cz growth furnace. The silicon nuggets stored in the hopper are transported for melting by the auger mechanism to the auxiliary crucible (premelter) in the growth chamber. The molten silicon is then led continuously to the crucible through a trough during growth. A comparatively satisfactory arrangement of the premelter, however, was a vertical, cylindrical graphite heater containing a small, fused silica test-tube liner. Incoming silicon was melted here and flowed into the main crucible through an opening at the bottom of the premelter.

During the course of this development, difficulties were encountered in the operation of the auger feed mechanism and silicon melting in the auxiliary crucible.

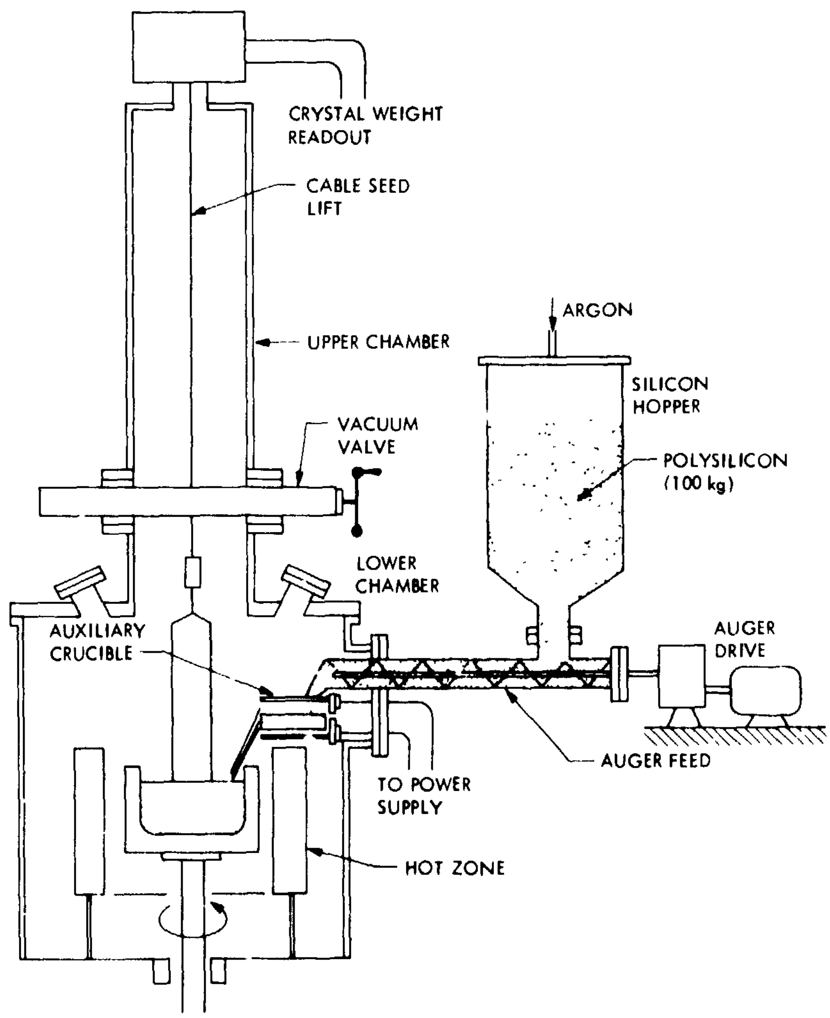

Figure 4. Continuous Nugget Silicon Feed for Czochralski (Cz) Grower

B. BATCH RECHARGING

1. Granular Silicon Recharging

This batch recharging effort by Varian Associates consisted of a modification of their 2850 Cz grower (Reference 10). After ingot growth, granular silicon is transported into the growth crucible using a vibratory feeder arrangement (Figure 5 shows this transfer system schematically). The pellet hopper is attached to a vibratory feeder and an isolation lock. The feed tube from the isolation lock transfers the granular silicon into the growth crucible after each ingot pull. The lock then isolates the feed arrangement from the growth chamber during the melt-down and growth cycle. Using this method, an ingot of approximately 25 kg was grown, remelted in the same crucible without cool-down, and another ingot of nearly the same weight was regrown. This growth run demonstrated that multiple growth and maintenance of the crucible in a high-temperature environment for longer periods were feasible. Because of difficulties in the operation of the vibratory feeder, granular recharging of silicon could not be demonstrated.

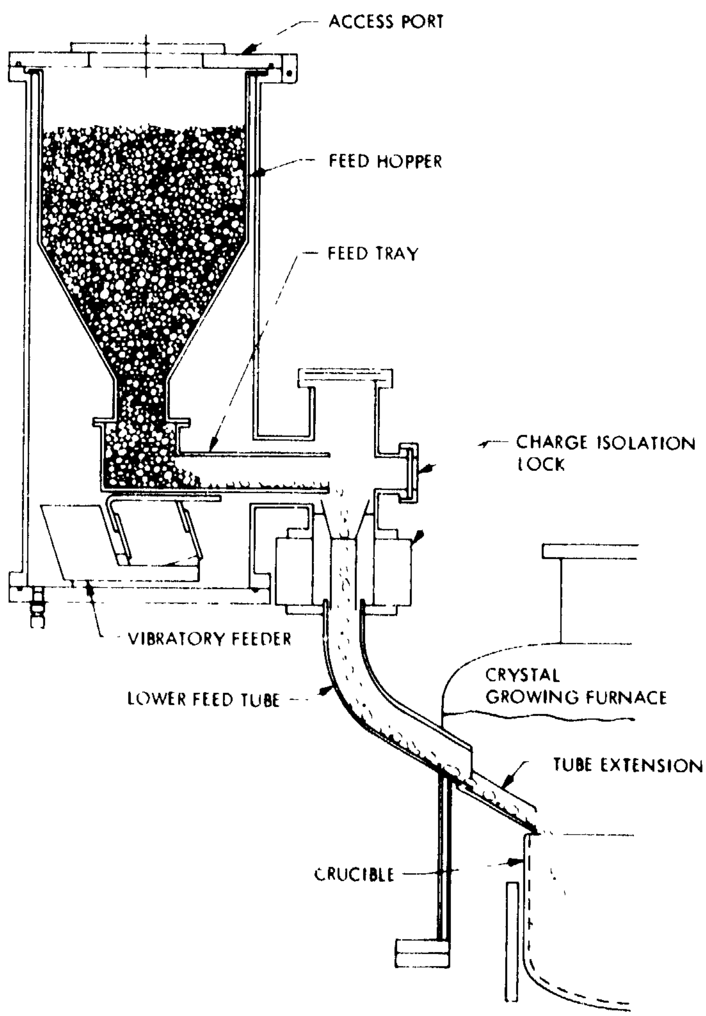

Figure 5. Granular Silicon Recharging

2. Solid Feed Recharging

This effort by the Hamco Division of the Kayex Corporation initially consisted of suitably modifying the Hamco CG2000 grower to meet the following objective (Reference 11):

The growth of 100 kg single crystal silicon ingots of 10 cm or greater in diameter, using one crucible to demonstrate technology for \$2.80/W_p.

This objective was demonstrated, and the program was extended toward the growth of 150-kg single crystal ingots of 15-cm-diameter at 2.5 kg/h throughput rate. Details of this method are given in Section IV.

SECTION IV

ADVANCED CZOCHRALSKI (CZ) GROWER WITH SEQUENTIAL SOLID FEED MELT REPLENISHMENT

In this approach a pull chamber above the growth chamber is used for introduction of polysilicon rod or lumps, lowering it in the growth chamber for melt replenishment in the crucible, and growth and removal of the grown ingot for each sequential growth.

A. DESIGN CRITERIA

The overall equipment design required the following criteria to be met during multiple ingot growth (Reference 12).

(1) The growth chamber must always be under vacuum or low pressure to reduce the silicon oxide build-up.

(2) The crucible must be kept hot during each multiple ingot growth run to prevent breakage.

(3) Polysilicon recharge must be added to the hot crucible without contaminating the silicon melt or the hot-zone system.

(4) Ingots must be removed from the furnace without admitting air in the growth chamber.

The last two criteria require a vacuum-tight isolation valve between the pull and growth chambers.

Figure 6 is a side view of the grower. It consists of a growth chamber (C) containing the crucible (F) and the heating elements (not shown), mounted on the base (P). The pull chamber (B) is isolated from (C) by a "flapper" type vacuum-tight valve (A). This water-cooled valve is the most important element of the modified system, which maintains the crucible under vacuum or at low argon pressure at all times, even when the pull chamber is opened out for removal of the grown ingot (D) or the introduction of the polysilicon rod or the lump charge hopper (E).

After completion of an ingot growth, the ingot is pulled out by the mechanism (O) fully into (B) and the isolation valve is closed. After the pulled crystal cools, air is admitted, and the pull chamber is opened for retrieval of the ingot, followed by seed mounting on its holder (K) for the next ingot growth from the same crucible. The polysilicon recharging system (G), mounted at the top of the pull chamber, controls the movement of the feedstock during the charging period. The feedstock (either in rod form or lumps contained in a hopper, as shown in Figure 7) is held by a fixture (I) and cable (J). During the recharge cycle, the seed crystal with its holder (K) is stationed above the recharge mechanism and out of the way of the feedstock. A weight-measuring device, called a torque transducer (H), in the recharge mechanism allows the correct amount of material to be melted. The remaining feedstock, suspended by the cable, is moved at the rear of the pull chamber so that it does not interfere with the crystal during the growth.

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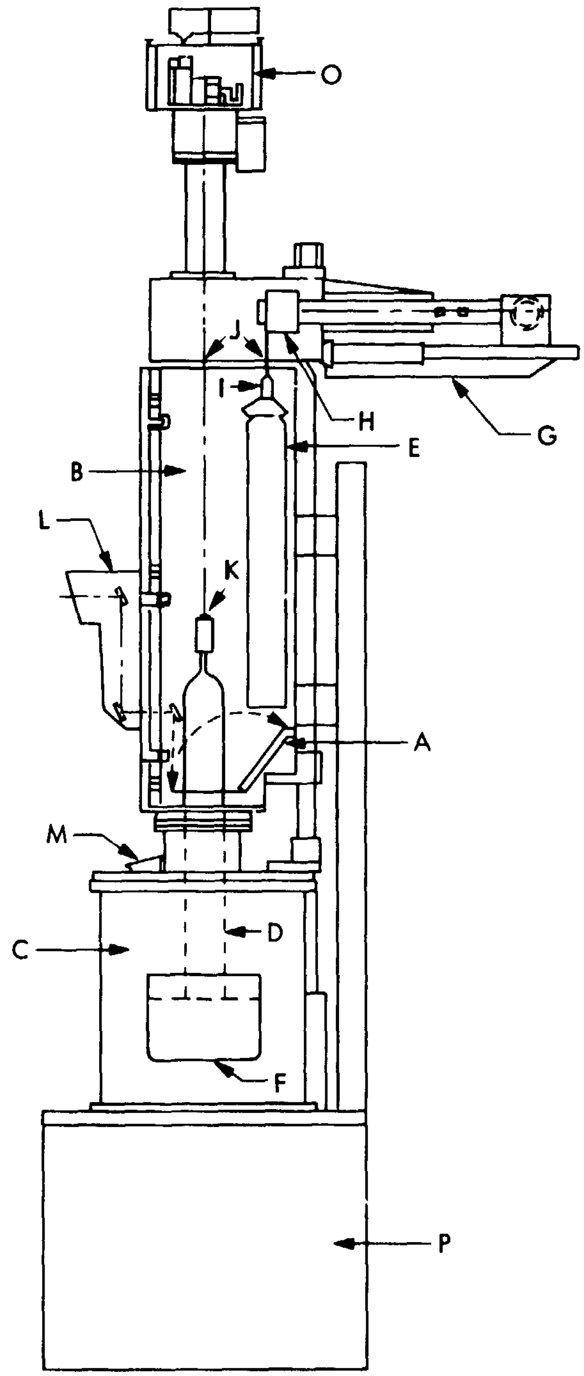

Figure 6. Side View of the Melt Rerlenisnment Czochralski (Cz) Grower

(A) Isolation Valve, (B) Pull Chamber, (C) Growth Chamber, (D) Crystal, (E) Recharge Rod or Lump Charge Hopper, (F) Quartz Crucible, (G) Recharge Mechanism and Weight Measuring System, (H) Torque Transducer, (I) Recharge Holder, (J) Cable, (K) Seed Holder, (L) ADC Optical System, (M) View Port, (O) Pull Mechanism, and (P) Base.

Other features of the grower include a doping fixture (not shown in Figure 6), an ingot weight readout to control the melt position in the furnace, an automatic diameter control optical assembly (L), and a view port (M) to observe the melt, the growing crystal, or the liquid-solid interface.

The grower is designed to accommodate up to a 40-cm (16-in.)-diameter crucible. A microprocessor control is designed and built to grow the crystals in an automatic mode to reduce labor cost and operator error for improving yield. In addition, the microprocessor enables better control of growth parameters and acts as a diagnostic tool.

B. CRYSTAL GROWTH PROCEDURE

For a 100-kg growth run per crucible (4 ingots, 25 kg each), approximately 18 kg of lump silicon and the required amount of dopant are placed in a 30-cm-diameter x 22-cm-high quartz crucible of 30-kg capacity, and the grower is evacuated to about 20-Torr pressure. At this point, the argon flow is adjusted to maintain a furnace pressure of about 20 to 25 Torr, and the furnace is allowed to heat up and stabilize at approximately 1550°C. (The melting point of silicon is 1410°C.)

To grow the first 25-kg ingot, a total of approximately 30-kg charge is needed. After the initial charge of 18 kg has melted, making space available for the balance, silicon feedstock is added into the crucible. In earlier experiments, a polysilicon rod was used for this purpose. However, the rod cracked in several places because of uneven thermal conduction in polysilicon, which made it difficult to control the charge in the crucible. To overcome this difficulty, silicon lump charging with a hopper and cone arrangement was used in place of a poly feed rod. The arrangement, shown in Figure 7, consists of a cylindrical hopper with a cone at the bottom acting as a feed distributor. The hopper, filled with lump silicon attached to the cable J (Figure 6), is lowered into the growth chamber over the crucible during charging. Lowering of the cone from the hopper bottom releases the lumps into the crucible. When the desired amount of silicon has been added, as indicated by the weight-measuring device, the charging is stopped by retracting the cone in the hopper. The hopper is then raised and moved to the rear into the pull chamber. To start the crystal growth, the furnace temperature is brought to slightly above 1410°C, and the seed is lowered to establish stable contact with the melt. The seed lift is started, and small adjustments in furnace temperature are made to form a neck and "crown." When the crown reaches very nearly its desired diameter of 10 cm, the pull speed is increased to approximately 0.02 cm/s until the crystal rounds over and begins to grow straight. The pull speed is then adjusted to the required growth rate, and the diameter controller is placed in automatic mode.

The control system consists of three closed-loop process controllers: a diameter controller, a growth speed controller, and a temperature controller that controls the furnace temperature within 0.5°C. At the end of the growth, the crystal is tapered at the bottom to minimize the thermal shock that occurs when the crystal is withdrawn from the melt. The grown ingot is raised into the pull chamber, and the isolation valve is closed. The ingot is cooled down to approximately 200°C by purging the chamber with argon. The ingot is then removed and seed is replaced if required.

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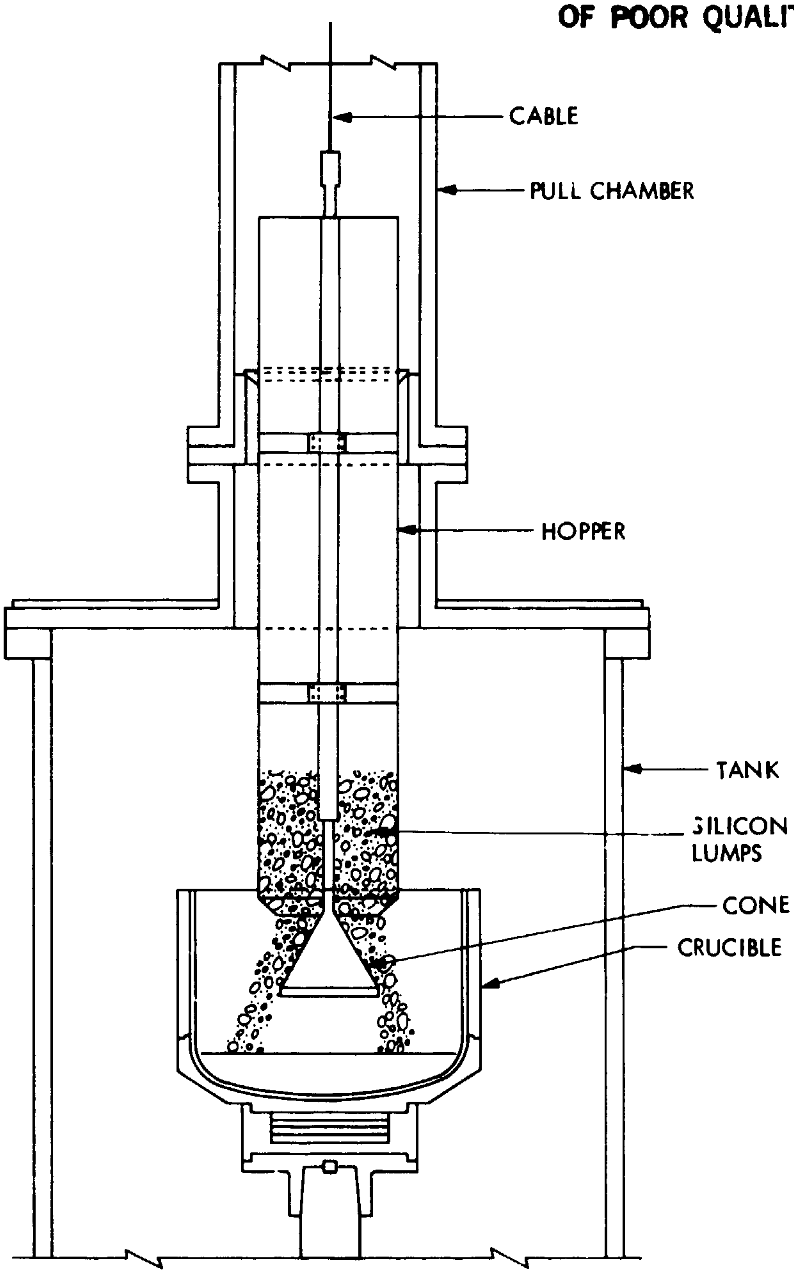

Figure 7. Lump Recharging by Self-Dumping Hopper

For recharge cycle, the hopper is filled with silicon lumps, the chamber is closed and evacuated, the isolation valve is opened and the replenishment is started after moving the hopper over and lowering it very near to the crucible.

The entire process is repeated by melting enough polysilicon to replace that which is consumed by the grown crystal. When the crucible is replenished, the hopper is raised and moved to the rear of the pull chamber, and the appropriate amount of dopant is added through the dopant fixture. At this point all conditions are identical to that established at the beginning of the first crystal growth cycle. Thus, from this point on, the multiple Czochralski process involves alternate growth and recharge cycles as previously described.

In these runs the crucible contained molten silicon for a period of over 90 hours, indicating that it is capable of sustaining growth for such long periods. For a 150-kg growth run (5 ingots, each 30 kg in weight and 15 cm in diameter), a 40-cm-diameter crucible was used. Several runs, each of 100 kg and 150 kg silicon per crucible with multiple ingots, have been successfully made. These runs produced single and polycrystalline material in various amounts.

The quality of the crystal is affected, among other factors, by the chemical impurities present in the melt and in the solid. These impurities are segregated from the solidifying mass into the melt. As the rejection of impurities continues from ingot to the melt, the impurity content of the melt constantly rises. The effect is compounded when multiple ingots are grown from the same crucible with polysilicon recharging. Consequently, the impurity levels in successive ingots grown from one crucible also rise. Furthermore, higher levels of impurity degrade the structural quality of the crystals leading, finally, to polycrystallinity.

These effects raise related issues of performance degradation of solar cells with various species and concentration levels of impurities as well as structural imperfections. Material characterization, therefore, formed an essential part of this development activity, as described in Section VI.

SECTION V

ECONOMIC ANALYSIS

During the development phase of the Czochralski (Cz) growth process, economic analyses were performed and up-dated on a regular basis (Reference 11). Interim Price Estimation Guidelines (IPEG) methodology (Reference 13) was used to estimate the add-on price ($/kg) for growing Cz ingots from polycrystalline material. This calculation takes into account the direct as well as indirect costs (capital equipment, operation and maintenance). The details of this formulation are described in Reference 12. Table 2 gives a summary of parameters and estimated add-on price ($/kg) for 100 and 150 kg throughput per crucible.

Table 2. Economic Analysis for 100 kg and 150 kg Growth

Parameters	100 kg/crucible	150 kg/crucible
Crystal diameter (cm)	10	15
Growth rate (cm/h)	10	8.56
Total crystal pulled (kg)	100	150
Usable ingot yield (%)	72	81
No. of crystals/crucible	5	3
Total cycle time (h)	75	60
Throughput rate (kg/h)	1.3	2.5
Capital equipment ($)	175,000	267,000
Equipment life (yr)	7	7
Floor space (ft^2)	100	100
Direct labor ($/yr)	18,722	14,790
Materials ($/yr)	60,000	113,526
Utilities ($/yr)	15,390	30,109
Total quantity (kg/yr)	8,671	19,044
Add-on price ($/kg)	27.73	19.24

An add-on price of \$95/kg of silicon growth is equivalent to a \$2.80/W_p PV module price (interim goal). The calculated add-on price for 100 kg/crucible growth is \$27.73/kg as shown in Table 2. This clearly demonstrates the economic feasibility of this process.

For the \$0.70/W_p price goal to reach technical readiness by 1982, the add-on ingot growth price is \$14/kg. The calculations show that the 150 kg/crucible with 8.56 cm/h growth rate gives an add-on price of \$19.24/kg. Thus, additional performance improvements are required; e.g., it may be necessary to increase the growth rate from 8.56 to 12 cm/h.

It should be noted that these figures are estimated for a multi-million watt per year capacity. The data used here are based on limited number of actual growth runs.

SECTION VI

MATERIAL CHARACTERIZATION

In order to evaluate the quality of the material, structural, chemical and electrical characterizations were performed. Single crystal silicon samples from various ingots were Sirtl etched, and a dislocation density count was made. The measurement showed that the dislocation density was between $500/cm^2$ and $10^5/cm^2$.

A. IMPURITY ANALYSIS

The distribution of the impurities in the ingot and in the melt is generally determined by the segregation coefficient (k_o). It is the ratio of the impurity concentration in the ingot to that in the melt. As crystal growth proceeds, the impurities are rejected into the melt because the segregation coefficients are less than one. In addition, impurity buildup in the melt also increases with time because of the crucible dissolution. Increase of impurity concentration in the melt also causes it to increase in the growing ingot. When the impurities build up to a critical level in the melt, structural breakdown in the growing crystal occurs because of constitutional super-cooling. However, in the growth runs made with semiconductor grade polysilicon, the impurity buildup was not significant enough to cause constitutional super cooling. A study of the impurity buildup in the melt during continuous, as well as sequential, melt replenishment has been presented by Hopkins, et al (Reference 14). By using the Pfann relation (Reference 15):

$$C_L = C_L^i (1 - g)^{k-1}$$

for normal freeze in the usual Czochralski melt, Hopkins and his associates have derived impurity build up in the melt as:

$$C_L^f(n) \simeq C_o[1 + ng/(1 - g)]$$

after the n^{th} sequential melt replishment and

$$C_L \simeq C_o[1 + (1 - k) V_c/V_o]$$

for the continuous melt replenishment process

where

C_L = impurity concentration in the melt

C_L^i = initial impurity concentration in the melt

g = fraction of the melt solidified

k = effective distribution coefficient (related to k_o, crystal growth rate, impurity diffusion coefficient in the liquid, and the width of the stagnant layer at the solid-liquid interface)

$C_L^f(n)$ = final impurity concentration after n^{th} sequential melt replenishment

C_o = impurity concentration in the feedstock

V_o = volume of the melt

V_c = volume of the grown ingot

This study concludes that the impurity content in the ingots grown by continuous melt replenishment will be lower than those grown by sequential melt replenishment approach.

Davis, et al (Reference 16) have developed a model for the effect of the impurities on silicon solar cell performance. The model assumes that the impurity species in silicon act as carrier recombination centers additively and independently of each other. Experimentally, this group has shown that individual impurities such as vanadium, molybdenum, and titanium of the order of 10^{13} atoms/cm^3 deteriorate the solar cell efficiencies by approximately 30%, whereas nickel or copper is tolerated up to 10^{15} atoms/cm^3 with no appreciable reduction in the cell efficiencies.

In another study (Reference 12), multiple ingots were grown by using sequential melt replenishment approach. Impurity content in the ingots was calculated by using mass spectroscopy analysis of the feedstock, segregation coefficients, and the results of an earlier study (Reference 14). Solar cells were made from these ingots, and their performance was analyzed by using the Davis model (Reference 15). The experimental results were in fair agreement with the model calculations.

B. SOLAR CELL ANALYSIS

A number of 2 x 2 cm solar cells were fabricated on single as well as polycrystalline wafers cut from various sections of an ingot and from all the ingots of a growth run. These cells were fabricated using a standard baseline process simply to evaluate the material rather than to optimize the cell performance. This process consisted of a junction of nominal 0.35 μm depth without back-surface field (BSF) or multiple layer antireflection coating. For contact metallization, titanium-palladium-silver was used. Minority carrier diffusion length measurements were made on several of these cells using an approximately 1.5 MeV electron beam. These results showed, on an average, diffusion lengths of 100 μm. The solar cell data are presented in Table 3, indicating the run number, ingot number in that run, and Air Mass One (AM1) cell efficiencies for various sections of each ingot. Four to ten cells were fabricated from each section of the ingot. The averages of these efficiencies are also given in Table 3.

The top sections of all the ingots are single crystal. The cell efficiencies for these sections do not show significant decrease from the first ingot to the last for the same growth run. For example, for Run No. 30

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Table 3. Solar Cell Data

Run No.	Ingot No.	Section	AM1 7%	Comments
9	2	Top	12.1	
9	2	Middle	11.4	
9	2	Bottom	11.9	
9	2	Bottom	11.9	Dislocated
9	3	Top	10.9	
9	3	Top	10.9	Dislocated
9	3	Bottom	11.3	Poly
11	1	Top	12.5	
11	2	Top	11.6	
11	2	Bottom	12.1	
11	3	Top	12.2	
11	4	Top	11.2	
11	4	Bottom	11.2	
11	4	Bottom	11.5	Dislocated
Controls for Runs 9 and 11			12.6	
30	1	Top	13.7	
30	1	Middle	13.8	
30	2	Middle	9.4	Poly
30	2	Bottom	10.4	Poly
30	3	Top	13.3	
30	3	Middle	9.3	Poly
30	3	Bottom	9.8	Poly
30	4	Top	13.6	
30	4	Middle	10.4	Poly
30	4	Bottom	10.3	Poly
30	5	Top	13.3	
30	5	Middle	9.7	Poly
30	5	Bottom	9.4	Poly
30	6	Top	12.7	
30	6	Middle	8.9	Poly
30	6	Bottom	10.8	Poly
Controls for Run 30			12.6	

Table 3. (Cont'd)

Run No.	Ingot No.	Section	AM1 η%	Comments
62	1	Top	13.3	
62	1	Bottom	13.0	
62	2	Top	13.4	
62	2	Bottom	12.8	
62	3	Top	14.3	
62	3	Bottom	11.6	
62	4	Top	13.0	
62	4	Bottom	13.6	
62	5	Top	13.2	
62	5	Bottom	13.2	
62	7	Top	13.3	
62	7	Bottom	10.9	Poly
62	8	Top	12.7	
62	8	Middle	14.0	
62	8	Bottom	11.3	Poly
62	9	Top	13.9	
62	9	Middle	13.2	
62	9	Bottom	11.5	Poly
Controls for Run 62			13.1	

the efficiency in the top sections of Ingots 1 through 6 ranges from 12.7 to 13.7%. The middle and bottom sections of some of the ingots are single crystal, such as the middle section of Ingot No. 2 of Run No. 9 and the bottom section of Ingot No. 4 of Run No. 11. For other ingots, such as Ingot Nos. 3, 4, and 5 of Run No. 30, the middle and bottom sections are polycrystalline. For polycrystalline material the average cell efficiencies are 15 to 25% lower than those of single-crystal material. Several cells, such as those from the top sections of Ingot Nos. 1, 2, 3, 5, 7, and 9 of Run No. 62, show equal or higher efficiencies compared to the control cells that were fabricated on high quality, single-crystal ingots. Some ingots in a run show a decrease in efficiency from the top to the bottom, whereas others show an increase.

Figure 8 shows AM1 efficiency data for successive ingots/amount of silicon material pulled from a single crucible for Run No. 62. At present, it is difficult to provide any correlation between solar cell efficiency and the position of the wafer with respect to the top, middle and bottom of the ingots. Wide variation in the performance of solar cells fabricated from polycrystalline portions of ingots may be attributed to the variations in grain size and grain boundary properties. This variation in efficiences (8.5 to 12.8%) is shown by the bars in Figure 8. Some of the portions of ingots that were studied were single crystal but had dislocations in the range of approximately 10^5 cm^2. The cells made from these ingots have comparable efficiencies to those obtained from the dislocation of free, single-crystal

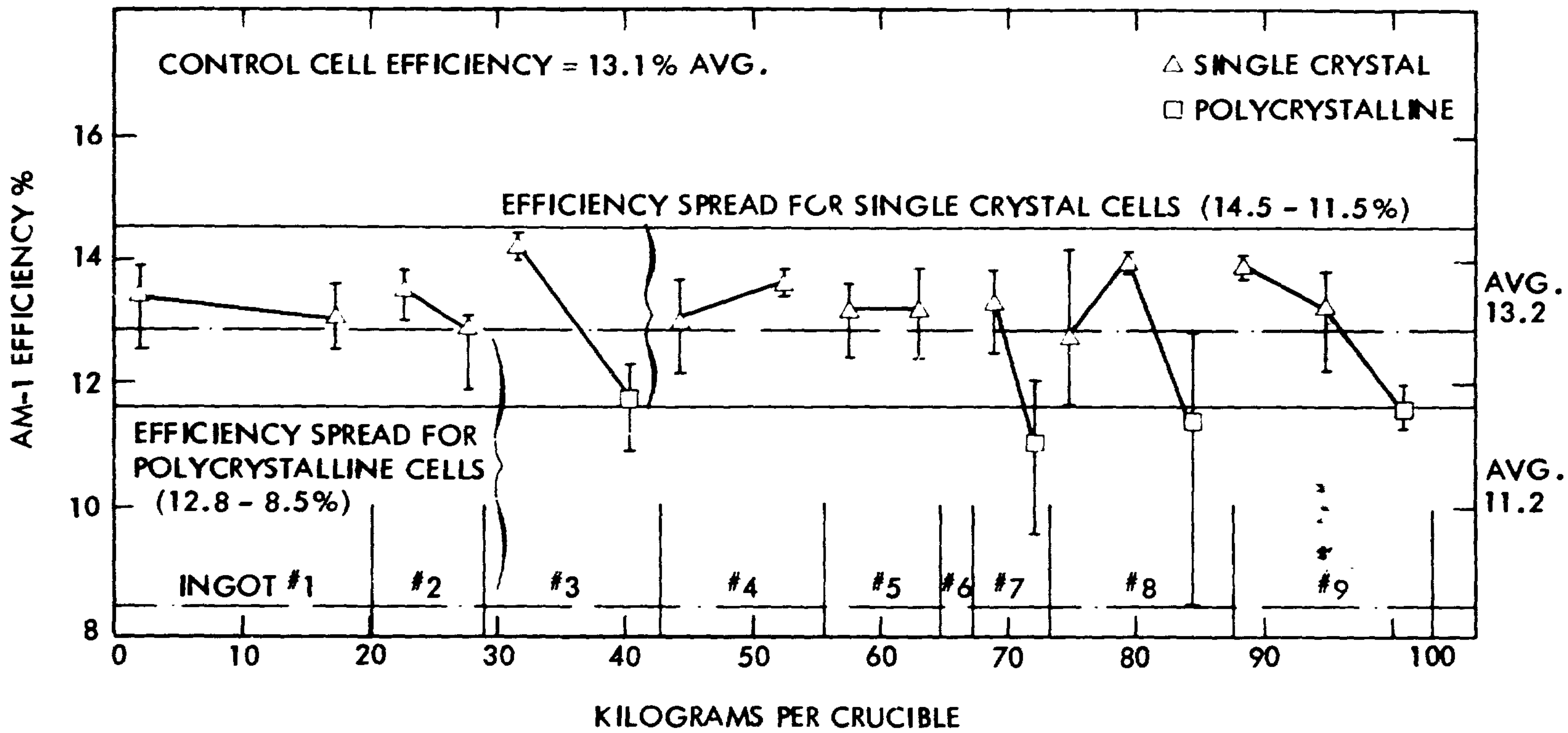

Figure 8. Solar Cell Efficiency versus Kilograms Grown

ingots. Sometimes loss of structure occurred because of silicon monoxide build-up and crucible devitrification for longer growth runs; consequently, a slight decrease in average efficiency toward the end of the run was observed.

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SECTION VII

PRESENT TECHNOLOGY STATUS AND CONCERNS

Several runs of multiple silicon ingots having a total weight of 100 kg have been successfully made by sequentially replenishing the crucible melt with polysilicon feedstock (Reference 10). Figure 9 shows a number of ingots grown in a run from a single crucible. Two runs each of multiple silicon ingots with a total weight of 150 kg have been made. Because of the successful multi-ingot growth runs, it is believed that the technology for the growth of 150 kg per crucible is now in place. Based on the success of the sequential melt replenishment approach, the photovoltaic industry has now begun using Czochralski (Cz) multiple ingot growth machines in its production line. In order to further increase throughput and growth yield and to reduce the operator error, a modified Cz grower CG2000 with microprocessor control (shown in Figure 10), has been built and is being tested at Kayex Corporation. The microprocessor control is now being used to grow the neck, crown and body of the ingot (Reference 16). In a run with a microprocessor, the seed is dipped in the melt; and neck, crown and shoulder growth are controlled according to previously determined and stored process specifications. The transition from neck to crown is initiated by operator response after visual check for proper neck formation. The operator receives a prompting request for this input. This decision is performed by the operator and allows him to determine if the neck growth has acceptable structural quality. Further work on improving automation is in progress.

As discussed earlier, the material obtained from either the 100 kg or the 150 kg ingot growth runs, however, was not all single crystal. The percentage of single-crystal material varied from run to run, but to date, not more than 60% single-crystal material was obtained from any run. This is one of the major areas of concern. The second area of concern is the growth rate. Economic analyses indicate that the growth rate of approximately 10 cm/h for a 15-cm-diameter ingot is required (References 10 and 17). Theoretically, this growth rate should be achievable with even the most conservative of assumptions made in Figure 2. However, to date, the highest average growth rate achieved is approximately 8 cm/h.

The last major area of concern is the crucible quality. Some of the crucible failure modes that result in crystalline structure breakdown are non-uniform devitrification, excessive flow stress, reaction with (and consequent impurity addition to) the melt, and air bubble bursting at crucible surface. The SiO_x particles produced by bubble bursting disturb the growth interface and result in inclusions in the grown material.

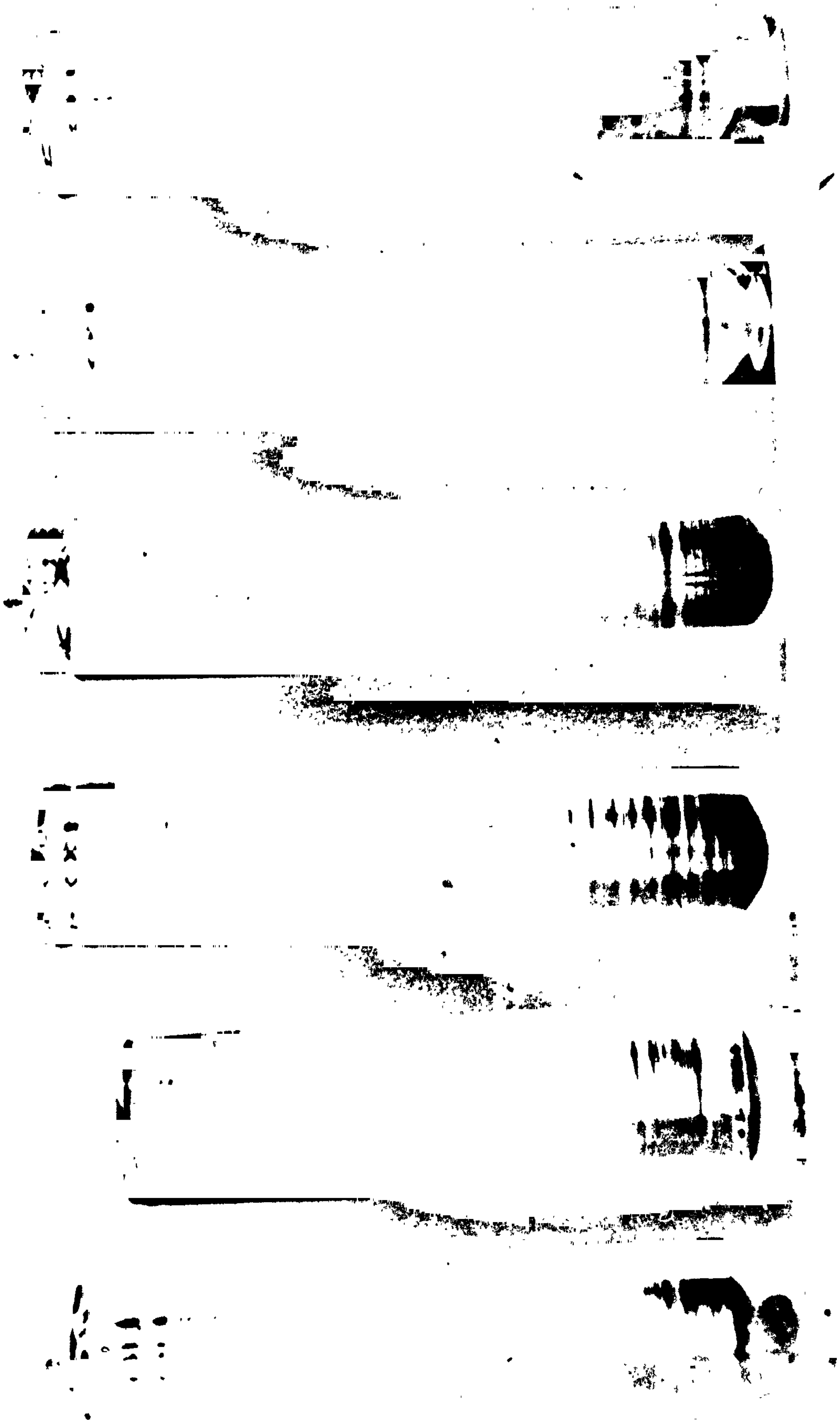

Figure 9. 15-cm-diameter Ingots from 150 kg Run No. 70

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Figure 10. Microprocessor-Controlled Advanced Czochralski (Cz) Grower

SECTION VIII

FUTURE RESEARCH

Although significant advances have been made in Czochralski (Cz) growth and material characterization, further research in the stated areas of concern should be continued.

To improve single-crystalline growth yield and to understand better the effect of impurities (such as oxygen, carbon and other Cz growth-related impurities) on solar cell performance, a systematic evaluation of the role of these impurities is required. Because the quality of the crucible influences the crystal structure, understanding of crucible-melt interaction for longer growth runs needs to be pursued. In order to achieve the required growth rate, understanding of efficient heat removal from the growth interface is needed. Approaches, such as the use of effective radiation shields, cooling coils and directed gas flow could be explored. Higher throughput rate could be obtained by increasing the ingot diameter. However, growth of ingots with larger diameters must be balanced against the increased difficulty of cost-effective wafering to provide an overall improvement in the cost of manufacturing the silicon sheets. An option of sectioning the larger-diameter ingots before slicing has also to be evaluated.

SECTION IX

SUMMARY AND CONCLUSIONS

The growth of silicon ingot by the Czochralski (Cz) technique is briefly described. Since quartz crucible and furnace parts form a large part of the growth cost, advanced Cz technique (for multiple ingot growth with melt replenishment using the same crucible) to lower the cost of photovoltaic modules is described. Four approaches of advanced Cz technique are discussed of which two used solid feed and two used liquid feed melt replenishment systems. Sequential solid feed approach is described in greater detail along with growth results for 100-kg and 150-kg runs. A brief economic analysis is also presented.

An advanced Cz grower with sequential solid feed melt replenishment system and microprocessor control has been successfully operated. The technology has been demonstrated to be in place and its use by the photovoltaic industry is underway.

Material characterization and solar cell evaluation indicates that impurity build-up is of no consequence for semiconductor grade starting feedstock. However, structural imperfections, causing lower single crystal yield and crucible quality, remain a concern requiring further research. Further improvement of growth rate by efficient heat removal and growth of a larger diameter ingot is perceived as a possible future thrust.

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