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16. Abstract

The goals of this program, sponsored by NASA-Lewis Research Center under Contract NAS 3-18034, were to determine the feasibility of growing wide, thin silicon dendritic web for solar cell fabrication and to develop conceptual designs for the apparatus required. An analysis of the mechanisms of dendritic web growth indicated that there were no apparent fundamental limitations to the process. Additionally, the analysis yielded quantitative guidelines for the thermal conditions required for this mode of crystal growth. Two classes of crucible designs were then investigated: the usual quartz crucible configurations and configurations in which silicon itself is used for the crucible. The quartz crucible design seems most feasible at this time and has been incorporated into a conceptual design for a laboratory scale crystal growth facility capable of semi-automated, quasi-continuous operation.

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SILICON RIBBON STUDY PROGRAM

R. G. Seidensticker and C. S. Duncan
Contract NAS 3-18034
Final Report

I. SUMMARY

The early commercial practice of growing silicon dendritic web crystals for solar cells fell short of the anticipated growth rates required for advanced solar cell technology. The objective of the present program was to study the feasibility of increasing the yield of the process by growing wider (5 cm), thin (0.015 cm) dendritic web crystals. Further, conceptual designs for appropriate crucible systems and laboratory growth equipment were to be developed if warranted.

The initial evaluation of feasibility was based on a comparison of theoretical models for dendritic web growth with the thermal geometry of earlier pilot line and laboratory growth systems. The results suggested that the circular geometry of the crucible was apparently the limiting factor on the width of web grown at that time. Further, the analysis indicated that the growth of the supporting dendrites and the growth of web itself were to a large extent independent so that different growth parameters, e.g., temperature and pull speed, were dominant in the growth of the two portions of the crystal.

Two main concepts were advanced for crucible systems for growing web: homocrucibles, where solid silicon is used to constrain the liquid silicon, and heterocrucible systems, where a separate crucible material is used. The technical difficulty in achieving the required thermal geometry in the homocrucible designs led to the choice of a quartz crucible system for advanced conceptual design.

The resulting molybdenum susceptor/fused quartz crucible design was incorporated into a conceptual design for a laboratory web growth facility. This system incorporates an automated material feed system to permit quasi-continuous, steady-state growth. Automated control features have also been incorporated in the design. Since the system is intended for laboratory development purposes, special emphasis was placed on versatility to readily accommodate any design modifications suggested by operating experience.

2.0 INTRODUCTION

2.1 General

This report summarizes the analysis and conceptual design efforts for improving the growth of silicon dendritic web. Dendritic web material has a demonstrated capability for high quality solar cell fabrication; however, the material throughput of the prior growth apparatus was generally too small to meet the possible demands of low cost solar arrays. The goal of the present program was to define the requirements for semi-automated, quasi-continuous growth of wide, thin, silicon dendritic web crystals. The initial phase of the study program was to synthesize existing data on web growth, both practical and theoretical, into a set of requirements for the web growth system. On the basis of these requirements, conceptual designs were developed for the crucible/heater/feed system needed for the advanced growth apparatus. Finally, a conceptual design for the complete laboratory system was developed.

2.2 Background

The growth of semiconductor material in ribbon form has always been an attractive technique. When Billig reported the growth of germanium dendrite crystals in 1955,[1] an intensive study program was undertaken at the Westinghouse Research Laboratories to understand and develop this mode of growth. The result was a comprehensive evaluation of the characteristics of what was called "controlled dendritic growth" in semiconductors, reported in a number of papers detailing growth mechanisms, impurity distributions and crystal perfection.[2-9]

During the course of the studies, it was observed that occasionally two coplanar dendrites would grow simultaneously with a film of liquid freezing between them. Thus "dendritic web crystals" were discovered.[10-13] These dendritic web crystals were much wider than the dendrites themselves and were apparently much better suited for semiconductor device fabrication. It was but a short time before not only germanium but also silicon was grown by this technique. Eventually, a pilot production facility was placed in operation and pilot quantities of solar cell arrays were produced. The hoped-for market for these devices did not develop and the development of techniques for growing large diameter Czochralski silicon reduced market potential of web crystals for other silicon devices. With this poor economic prognosis, the silicon web program was terminated.

In the early 1970's the need for new, nonpolluting energy sources became apparent. Direct photovoltaic conversion of solar energy is an attractive technique if it can be made economically viable. One possibility in this direction would be the automated production of silicon solar cells. Silicon web crystals offer possibilities in this direction.

At the conclusion of the previous web crystal programs, pilot production facilities were routinely producing high quality material of 1 cm usable width and 6 to 15 meters in length. Wider material, up to 3 cm, was being grown on a laboratory scale. It was apparent that while there did not seem to be any fundamental limitations to the process, nevertheless, new concepts in apparatus were necessary to grow significantly wider and longer crystals than those being produced.

The opportunity to pursue such studies arrived with a study program under the aegis of the NASA-Lewis Research Center. The goal of this program was to develop the concepts required for apparatus to grow 5 cm wide silicon web crystals in a semi-continuous manner. The specific goal of the program was to develop a conceptual design for a laboratory system capable of growing at least 5 cm wide web and incorporating a silicon feed system to allow continuous replenishment of the melt.

3.0 ANALYSIS OF DENDRITIC WEB GROWTH

3.1 Description and Analysis of Results of Previous Program

The initial task in the program was to review the available data from previous web programs including not only published scientific papers, reports, etc., but also laboratory notebooks and interviews with personnel associated with the projects. The next step was to synthesize the information into a coherent picture of the equipment used for web growth, especially the general thermal geometry of the apparatus used for laboratory studies and for pilot production. Concurrently, a model for the pertinent features of the crystal growth processes of web crystals was generated.

3.2 Mechanics of Web Growth

The actual mechanics of growing a web crystal of silicon were very much the same whether in the laboratory or on the pilot production line. Normal practice was to first lower a small, etched, dendrite seed into the silicon melt and adjust the temperature so that the seed neither melted nor grew. This procedure provided an operational measurement of the controller setting for the melting point of silicon. Although the "Radiomatic" sensor used for temperature control was not very reproducible from run to run, the calibration procedure prevented this drawback from becoming a real problem. Once a setting was determined, however, the equipment would hold the control setting with a high degree of constancy, estimated at $\pm$ 0.2°C. Further, temperature changes could be accurately determined from the relation $\Delta T = (T_m/4E_o)\Delta E$ where T_m is the experimentally determined melting temperature and E_o is the sensor output at that temperature.

After the melting point setting was determined, the temperature was lowered slightly until lateral growth occurred on the melt surface from each side of the seed in the plane of the twin planes. Although the control system was not well calibrated in an absolute sense, changes in temperature could be readily determined to a fraction of a degree, and the usual practice was to lower the set point temperature from 2 to 4°C below the melting temperature in the buttoning stage. After the lateral growth had reached a convenient size, the dendrite seed was slowly pulled from the melt allowing the coplanar dendrites to grow downwards from the "button". A film of molten silicon was held by surface tension between these bounding dendrites and the button to form the web section of the crystal; the solidification front was slightly above the surface of the melt. Figure 1 is a schematic drawing of a growing web crystal shortly after starting growth and shows the initial seed and button.

After the initial reduction in temperature and the start of the pull, both the control point temperature and occasionally the pull speed were slowly changed during the balance of a run. The change in melt temperature was for the primary purpose of maintaining the size of the supporting dendrites and had little effect on the thickness of the web section of the crystal. The increase in pull speed had the primary effect of changing the thickness of the web section. Control changes were typically of the order of 3°C per meter of growth. It should be emphasized that these temperature changes may reflect long term drift in the "Radiomatic" temperature sensor as well as changes in the thermal geometry of the crucible/melt/web heat loss. If no changes were made, then the supporting dendrites would thicken as the melt level dropped and nucleation of spurious dendrites ("Thirds") would occur. With care, web crystals could be grown as thin as 0.005 cm; the best thickness for ease in growth was from 0.01 to 0.02 cm. Thicker webs produced additional problems resulting from silicon droplets freezing in channels on the supporting dendrites.

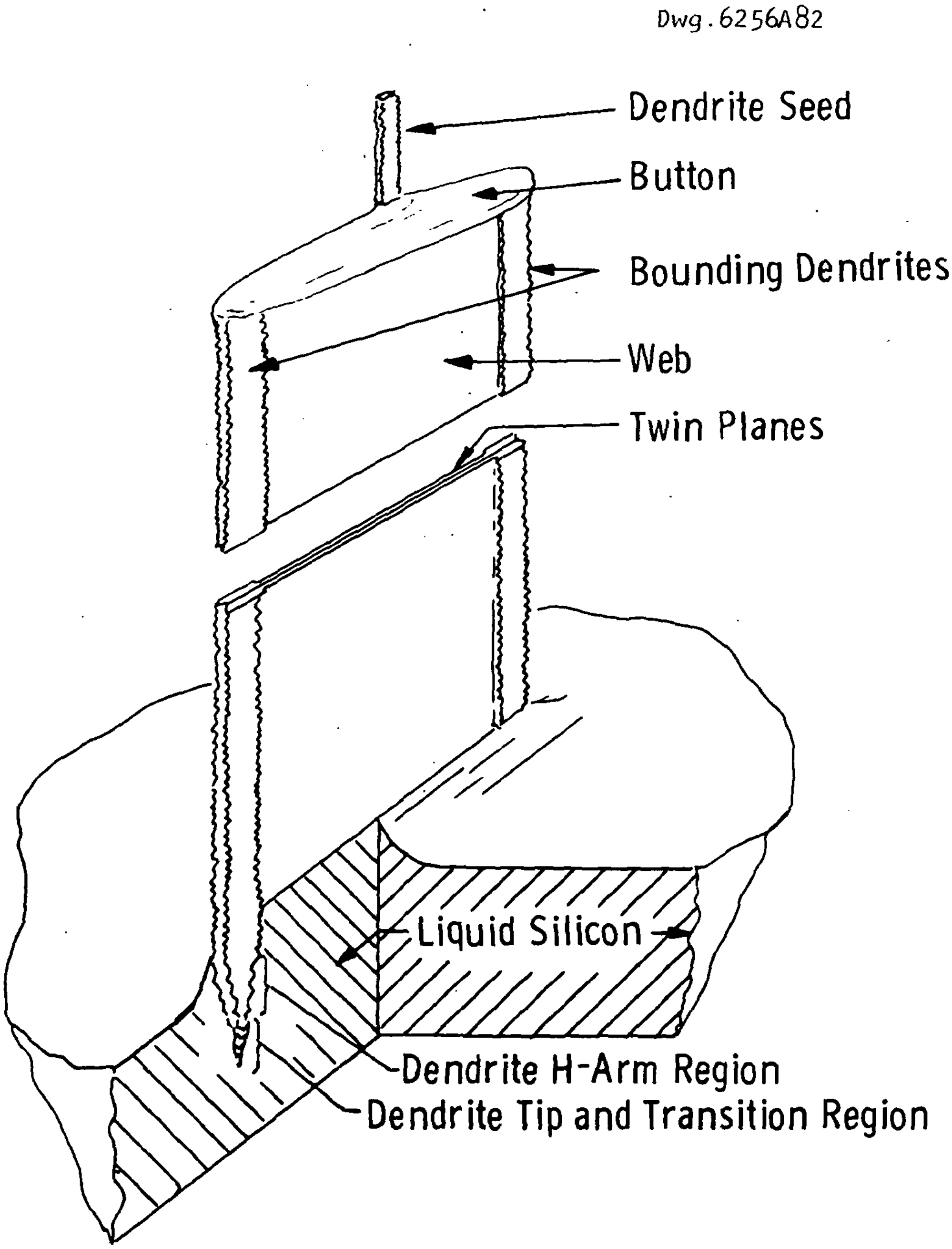

Fig. 1. Schematic sections of web growth.

The width of the initiating button was kept relatively small, of the order of 1 to 1.5 cm. This was necessary because the initial seed was very small in order to achieve good nucleation and was not supported other than being attached to a leader tape on the pulling mechanism. This small seed could not resist much torque and if the button were not grown with equal mass on either side of the seed, it would tilt when the pull started and the crystal would be lost. Efforts were made to use pieces of web for seeds, but without success.

After the initial 30 cm or so of web had been grown, the crystal was supported by guides so that it could widen without danger of tilting. This widening process was an intrinsic feature of the growth system resulting from a slight lateral thermal asymmetry at the dendrite tips caused by the evolution of latent heat from the web section of the interface. The widening continued until a balance was reached between the thermal fields generated by the growing crystals and the thermal fields intrinsic to the crucible. There were apparently no limitations on the width to which web could be grown other than those imposed by the geometry of the growth system being used.

A final feature of the mechanics of web growth which should be mentioned is the adjustment of the induction coil position with respect to the susceptor. Because of the slight resistivity inhomogeneities in the susceptor material or emissivity variations in the radiation shielding, the thermal center of the melt would not be located at the mechanical center. Moving the induction coil with respect to the susceptor would compensate for these problems; once the proper adjustment had been found, it was usually not necessary to make further changes unless the susceptor or shielding was changed.

3.3 Analysis of Crucible Thermal Geometry

In previous programs, a number of crucible/susceptor designs were used in the course of laboratory studies and for pilot production of web crystals.[13] Although these crucible/susceptor designs differed in detail, they all had a number of common features, and web crystals were grown successfully from all. Basically, all designs incorporated a round fused quartz crucible held in a molybdenum susceptor. All designs incorporated a slotted radiation shield or lid on the crucible, and most of the later designs incorporated lateral radiation shields to decrease the heat loss from the web itself.

At various times, attempts were made to measure the temperature distribution in these melts, but the results were far from satisfactory. Therefore, in this program, finite element analysis techniques were used to model the thermal fields in a typical crucible design. The actual configuration analyzed and the thermal model used for the analysis are shown in Fig. 2. The double top lid of the real system was replaced by a single "thick" lid and the multi-section molybdenum susceptor was modeled as a solid body. In order to analyze this model, the areas were broken up into 349 internal nodes, four floating external nodes to permit linearization of the radiation transfer and three fixed nodes at ambient temperature. The nodal geometry used is shown in Fig. 3. Appropriate area factors were included in the node connections so that the solution to the two-dimensional net duplicates the solution for a rotationally symmetric solid. The boundary conditions imposed on the problem were a constant temperature at the peripheral nodes of the susceptor.

The temperature distribution for the complete model is shown in Fig. 4 with isotherms plotted at 15°K intervals. A more detailed picture of the liquid silicon and neighboring regions is shown in Fig. 5 where the isotherms are either at 1°K or 2°K intervals. Near the center of the melt the isotherms are more or less "yo-yo" shaped so that the axial temperatures at the top and bottom of the melt are nearly the same.

The calculated thermal fields are a consequence of the assumed boundary conditions of a constant temperature on the periphery of the

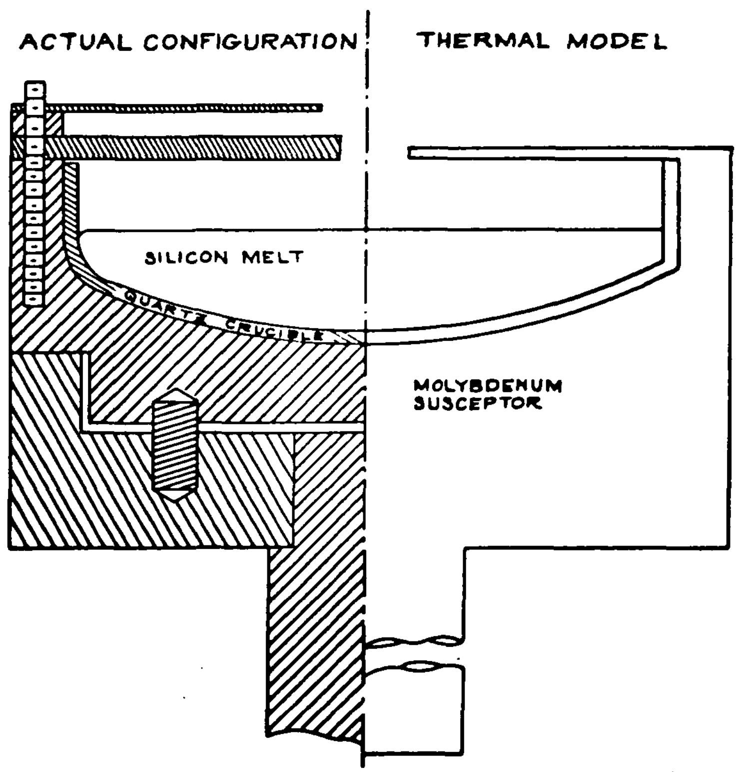

Fig. 2. Geometry used for thermal analysis.

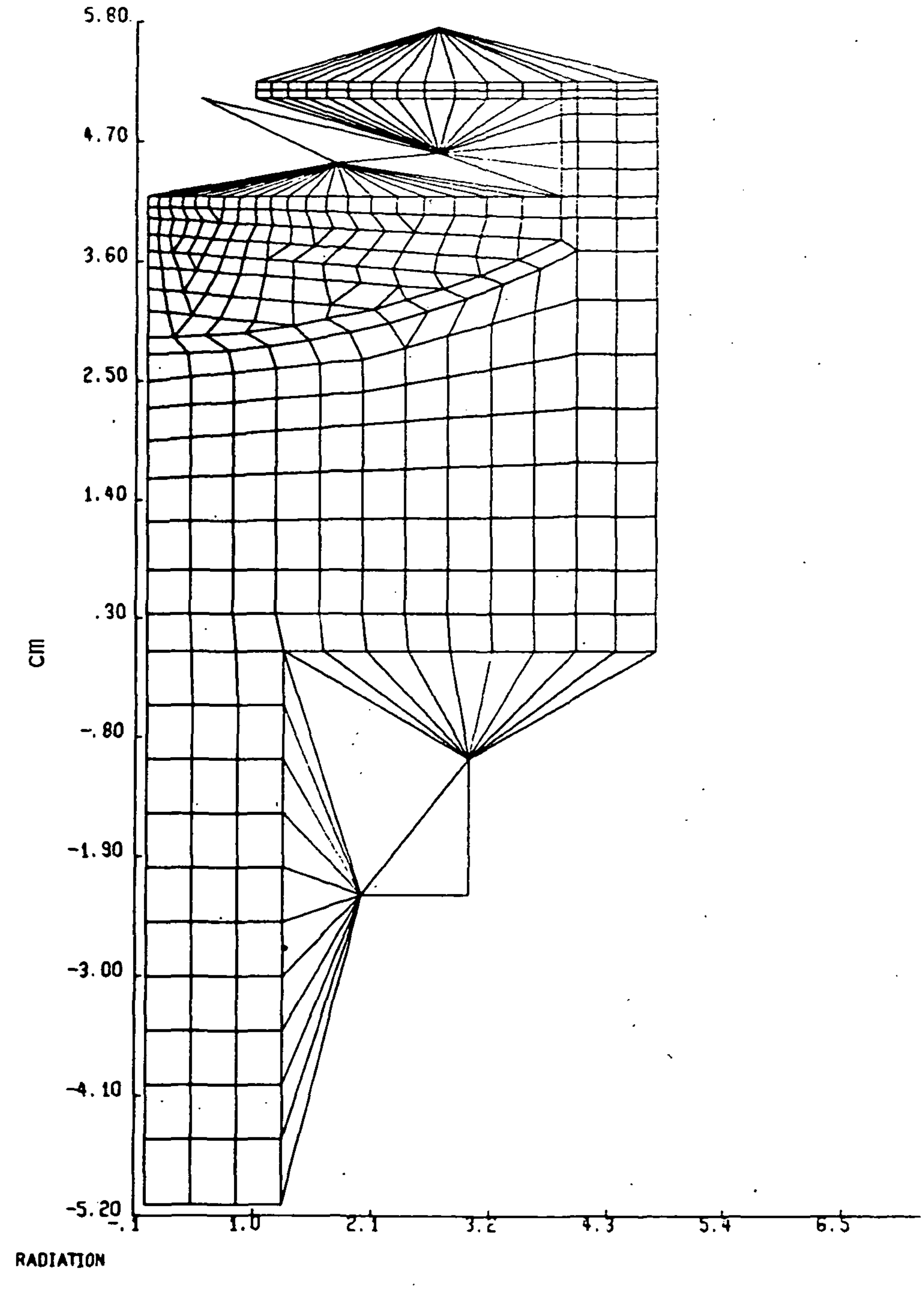

Fig. 3. Nodal connection paths for thermal model.

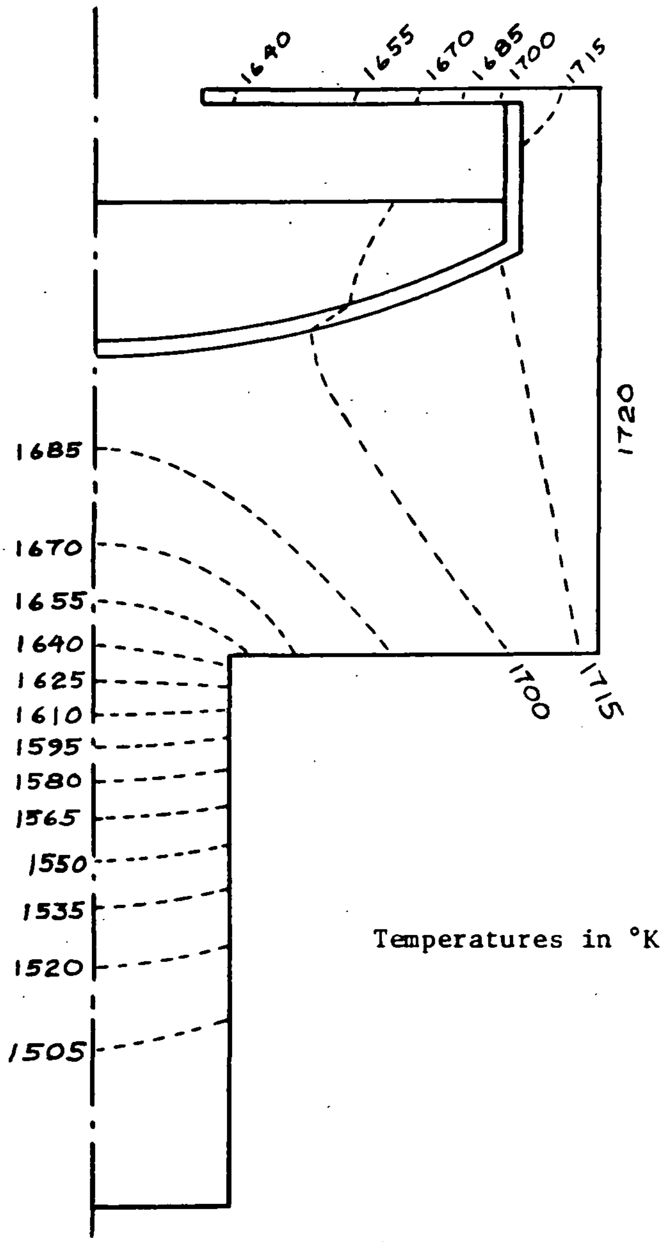

Fig. 4. Isotherms in complete model.

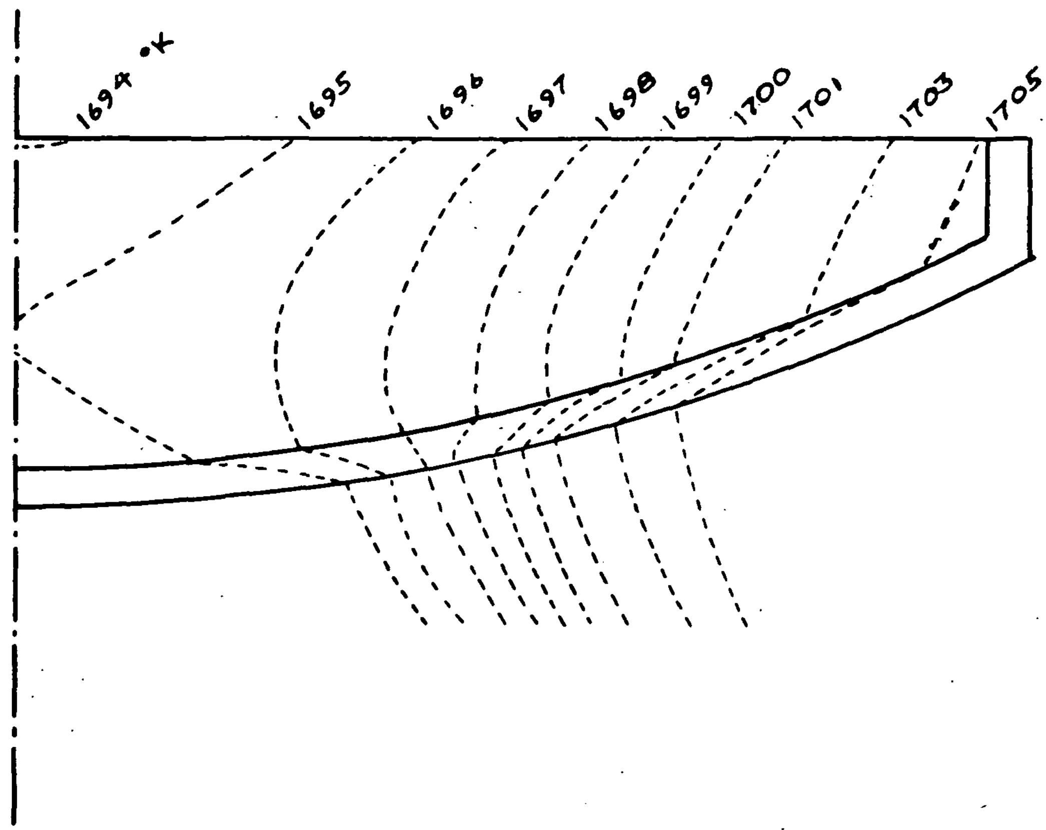

Fig. 5. Isotherms in crucible region.

crucible and radiation losses elsewhere. In fact, the induction field partially coupled to the lids, to the susceptor bottom and even to the support rod. This coupling would have the effect of reducing the surface losses and hence diminishing the vertical components of the temperature gradients. The effect would be to make the isothermal surfaces more dish-shaped although the bottom of the crucible would still differ by only a few degrees from the surface. Since a supercooled melt is necessary for the growth of the bounding dendrites, the shallow vertical gradient implies that slightly supercooled liquid silicon could be in contact with the quartz crucible. It has been commonly believed that this would cause nucleation of solid silicon and freeze out of the entire melt. That this did not occur is one of the more interesting results of the comparison of observed reality with the analytical predictions.

3.4 Model for Web Crystal Growth

The growth processes of dendritic web crystals can be separated for analysis into two separate parts: the growth of the web section of the crystal and the growth of the supporting dendrites. Further, both modes of growth can be analyzed both in terms of transport mechanisms (heat flow, solute redistribution, etc.) and in terms of crystal kinetic mechanisms (atomic attachment kinetics, etc.). For a complete model of the growth process, all the various approaches must be integrated; however, for ease in constructing the model, they may be considered separately. As a rather overly broad generalization, one might say that the transport phenomena dominate the macroscopic aspects of the growth processes, while the crystallographic phenomena determine the microscopic details of the processes. Heat and mass flow are the over-riding factors in determining the growth velocity of both the web and the dendrites, while crystallographic factors determine the detailed morphology of the crystals and, in the case of dendritic growth, provide a needed constraint for the solution of the heat flow problem.

3.4.1 Web Portion of Crystal

The physical basis for the analysis of the effect of thermal conditions on the web dimensions can be outlined very simply. Consider a ribbon crystal being pulled from the melt. The temperature distribution in this crystal can be calculated from the heat flow equation with boundary conditions which only involve the heat losses from the ribbon and the temperatures at the growing interface and far from the growing interface. The resulting temperature distribution is a function of the ribbon thickness, the heat transfer coefficients, the growth rate and the thermal environment of the crystal. Once the temperature distribution has been found, then obviously the temperature gradients in the solid can also be calculated.

The temperature gradients in the crystal are, of course, related to the heat flow; at the growth interface the heat flux in the crystal must equal the latent heat of fusion plus any heat flux from or to the melt. When this requirement is combined with the temperature gradient calculated from the temperature distribution in the crystal, a relationship can be established between the crystal dimensions, the growth velocity (i.e., the latent heat) and the heat flux from the melt. To a first approximation, the heat flux to or from the melt is proportional to the difference between the melting temperature and the crucible temperature. Thus, maintaining a constant crucible temperature will result in a steady state growth morphology if none of the other system parameters change.

The actual calculations for the ribbon problem would proceed as follows. Consider the geometry shown in Fig. 6, which shows a web of thickness a and width b growing from a melt. The primed coordinate system is fixed in the web and thus moving at a velocity v with respect to the unprimed coordinates which are fixed with respect to the growth interface. Since the web is very thin, the problem may be considered as one dimensional, i.e., the temperature only varies in the z direction, however, the heat loss from the web surface must be explicitly included.

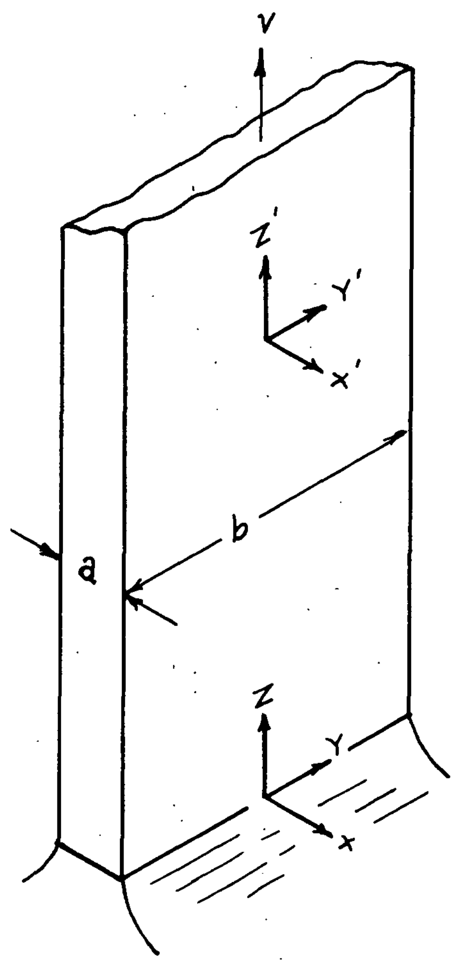

Fig. 6. Coordinates for web growth analysis.

In calculating the temperature distribution in a web crystal growing in a real system, different heat loss conditions must be used for different parts of the system. The thermal environment of the section of web growing in the crucible differs from the environment in the "after shield" region which further differs from the environment of the web above the after shields. Although the detail afforded by this procedure would warrant the added complexity in the analysis of a specific design, it is not justified in the present treatment. The purpose here is to illustrate the technique and arrive at a semi-quantitative description. A single linear heat transfer coefficient and a constant ambient temperature will be used for the calculations.

The one dimensional heat flow equation in the primed frame (fixed w.r.t. the web) is (see list of symbols)

$$ab\kappa \frac{\partial^2 T}{\partial z'^2} = 2bh(T-T_\infty) = abC\rho_s \frac{\partial T}{\partial t} . \tag{1}$$

Transforming to a coordinate frame fixed at the interface and dividing by b, this equation becomes

$$a\kappa \frac{\partial^2 T}{\partial z^2} - 2h(T-T_\infty) = aC\rho_s \left[\frac{\partial T}{\partial t} + v \frac{\partial T}{\partial z}\right] \tag{2}$$

In steady state, $\partial T/\partial t = 0$ giving the final equation

$$\frac{\partial^2 T}{\partial z^2} - \frac{v}{\alpha}\frac{\partial T}{\partial z} - \frac{2h}{a\kappa}(T-T_\infty) = 0 \tag{3}$$

where $\alpha = \kappa/C\rho_s$ = thermal diffusivity, and the partial differential equation can now be treated as an ordinary differential equation. Equation 3 has the solution

$$T - T_o = A \exp(m_+ z) + B \exp(m_- z) \tag{4}$$

where

$$m_{\pm} = \frac{v}{2\alpha} \pm \sqrt{\left(\frac{v}{2\alpha}\right)^2 + \frac{2h}{a\kappa}} \tag{5}$$

For boundary conditions we now take $T = T_m$ at $z = 0$ and $T = T_\infty$ at $z = \infty$. The constants A and B are found to be $A = 0$ and $B = T_m - T_\infty$ so that

$$T - T_\infty = (T_m - T_\infty) \exp(m_- z) \quad . \tag{6}$$

The temperature gradient at the freezing interface $z = 0$ is readily found to be

$$\left(\frac{dT}{dz}\right)_o = (T_m - T_\infty)\left[\frac{v}{2\alpha} - \sqrt{\left(\frac{v}{2\alpha}\right)^2 + \frac{2h}{a\kappa}}\right] \quad . \tag{7}$$

The conservation of heat at the freezing interface generates the additional condition that

$$-\kappa_s \frac{\partial T_s}{\partial t} = Q + vL\rho_s \tag{8}$$

where Q is the heat flux to the interface from the melt either by conduction or convection. The gradients from Eqs. (7) and (8) must be the same yielding the relationship

$$-\kappa_s (T_m - T_\infty)\left[\frac{v}{2\alpha} - \sqrt{\left(\frac{v}{2\alpha}\right)^2 + \frac{2h}{a\kappa}}\right] = Q + vL\rho_s \quad . \tag{9}$$

Equation (9) can be solved for the web thickness a in terms of the other parameters:

$$a = \frac{\kappa_s^{\,2}(T_m - T_\infty)/2h}{\left(Q + vL\rho_s\right)\left(\frac{v}{\alpha} + \frac{Q + Lv\rho_s}{\kappa(T_m - T_\infty)}\right)} \tag{10}$$

The results of Eq. (10) should be used more as a qualitative guide than a quantitative measure because of the simplifying assumptions used in the derivation. The result does show that the web thickness is inversely dependent on both the pull velocity and on Q, the heat flux from the melt to the growing interface. Which variable exerts the greatest influence depends on the relative magnitude of the two terms; it has been observed that the velocity dependence was much larger than the temperature dependence.

LIST OF SYMBOLS AND APPROXIMATE VALUES

Symbol	Definition	Approximate Value and Units
a	Thickness of web	0.01 cm
A	Constant of integration	
b	Width of web	5 cm
B	Constant of integration	
C	Heat Capacity of Solid Silicon	1.04 J/gm-K
h	Heat transfer coefficient	W/cm^2-K
L	Heat of fusion of Si	1804 J/gm
Q	Heat flux from melt to interface	W/cm^2
r	Dendrite tip radius (mean: $\frac{1}{r} = \frac{1}{R_1} + \frac{1}{R_2}$)	10^{-2} to 10^{-4} cm
T	Temperature	K
T_m	Melting Temperature	1695 K
v	Crystal pull rate (1 in/min)	0.042 cm/sec
α_ℓ	Thermal diffusivity ($\kappa/C\rho$) liquid Si	0.23 cm^2/sec
α_s	Thermal diffusivity ($\kappa/C\rho$) solid Si	0.095 cm^2/sec
γ	Eulers No. (= 1.78107...)	
ε_ℓ	Emissive power of liquid Si	$\sim$ 0.22
ε_s	Emissive power of solid Si	$\sim$ .3
σ	Stephan-Boltzmann Constant	5.669(-12) W/cm^2-K^4
κ	Thermal conductivity (@ 1685 K) Solid	0.216 W/cm-K
	Thermal conductivity (@ 1685 K) Liquid	0.6 W/cm-K
ρ	Density-Solid	2.29 gm/cm^3
	Density-Liquid	2.49 gm/cm^3

The details of the heat flow in the melt near the web are difficult to calculate because of uncertainties in the exact geometry; however, since the melt near the web is supercooled, the heat flux, Q, must be negative. Thus, a faster pull velocity can be used than if Q were positive, i.e., heat flow from the melt to the ribbon as in the EFG process. An approximate thermal analysis shows that because of the small thickness of the web, the temperature gradients in the liquid at the interface should be relatively large compared with the intrinsic temperature gradients in the melt. This is in agreement with the fact that long lengths of relatively flat web were grown from melts which must have had at least approximately circular symmetry.

Even though the intrinsic thermal gradients in the melt were small compared with those generated by the growing web, they nevertheless did influence growth to some extent. Measurements of web flatness indicated that the web thickened at the center to conform with the crucible geometry. Further, this "bulging" was more pronounced with thicker web[13] which would generate smaller gradients in the melt and thus be more influenced by the crucible geometry. The model thus offers support to the intuitive conclusion that web should be grown from a crucible system having an intrinsic thermal geometry which is similar to the thermal fields generated by the growing web. The obvious advantage would be that web should maintain its flatness during long growth times (quasi continuous growth) since there would be no driving force to cause it to bulge.

The existence of a supercooled melt near the web raises an interesting question: why doesn't the growing interface break down with uncontrolled dendritic growth? In practice, such phenomena were observed and the web was said to have grown a "third"; nonetheless, this occurrence was not common and did not constitute a serious problem during the pilot line production of literally miles of web.

The mechanism by which the interface might break down is conceptually simple although a detailed mathematical analysis can be formidable. Imagine a flat, smooth interface growing into a supercooled melt. The growth rate is limited by the rate at which latent heat can be rejected. If now a small perturbation occurs on the interface, this perturbation can lose heat more readily than the surrounding material and thus grow more rapidly. The process is accelerated and soon the originally smooth interface has degenerated to a forest of dendrites.

The salient question for the present is the reason for demonstrated stability of a growing silicon web interface. What deus ex machina must be invoked to maintain an apparently smooth growth front under conditions which should cause it to become unstable? Although little direct evidence can as yet be marshalled on this problem, the answer probably lies in the energy barrier to nucleation presented by crystal facets of the {111} family. Such facets may require appreciable undercooling for the nucleation of new layers.[8] This point is considered by O'Hara and Bennett[11] in their description of web growth and they point out that nucleation should not be a barrier to the normal growth of web material. The intersection of the supporting dendrites with the web provides a ready source of nucleation sites from which layers can spread across the growth front normal to the macroscopic pull direction. Such layer sources cannot, however, provide the necessary growth conditions for a perturbation to protrude from the interface. Thus, crystallographic factors could provide the necessary stabilization, although the argument so far is suggestive rather than rigorous.

Similar crystallographic factors are undoubtedly operative on the flat surfaces on the web as well[11] and when the thermal geometry is favorable should result in extremely flat facets covering the majority of the web surface. It is important to realize, however, that crystallographic kinetic factors provide only a "fine tuning" when the other growth conditions have already brought the crystal close to the desired geometry. In a practical sense, this means that large {111} facets will develop on the crystal only when the surface is already nearly a {111} plane. In the web growth process this is automatically accomplished by the crystallography of the supporting dendrites; in other processes such as die-grown ribbons (the Stepanov technique[14]) or meniscus-fed Czochralski ribbons (the Tyco EFG process), very accurate seed orientation must be achieved a priori and maintained during growth. Thus, due to crystallographic faceting, the web growth technique is to a large extent self-stabilizing as far as thickness and surface penetration are concerned.

Additional stabilization of the interface may result from heat flow patterns in the melt. There was some indication that the freezing interface lies above the curved portion of the meniscus and the resulting heat fluxes could differ appreciably from the classical model of a supercooled interface. A full understanding of the situation requires a much more detailed evaluation than has been possible so far. Not only must the heat flow in the web be evaluated for more realistic conditions, but also the interaction of the web with the thermal fields in the crucible must be considered. These calculations will yield a more detailed picture of the growth process, but should not alter the general features demonstrated by the present analysis.

3.4.2. Dendrite Growth

So far, this discussion has been concerned only with the growth processes pertinent to the web region of the crystal. A bounding dendrite on either edge of the web is essential to the development of the web morphology and the dendritic growth processes are quite different from the mechanisms discussed so far. Unlike the branching morphology usually associated with dendritic growth in metals, the dendrites associated with web crystals grow as fine, wire-like projections 3 to 5 mm in front of the balance of the growth front. In common with other dendritic growth the silicon dendrites require a supercooled liquid to sustain their growth.

The growth mechanisms of semiconductor dendrites have been extensively studied, especially for the case of germanium. Since the crystallography of silicon is very similar to germanium, most of the results of those studies should be applicable to the present case with appropriate allowance for the material parameters. Much of the relevant information has been discussed in the literature, but will be summarized here for completeness.

Dendritic growth requires a supercooled melt so that the heat of fusion can be readily dissipated into the liquid phase. Various authors, such as Ivantsov,[15] Temkin,[16] and Horvay and Cahn[17] have treated the problem theoretically and although their results differ somewhat in detail according to the assumptions made in the analysis, all results find that the quantity (vr) is a function of the melt supercooling where v is the propagation velocity and r is the (effective) radius of curvature of the dendrite tip.

The problem of the dendrite whose shape is an elliptical paraboloid has been treated by Horvay and Cahn[17] and their results were found to agree very well with the observed behavior of germanium dendrites.[8] The applicable equation is

$$v(r_{||}\, r_{\perp})^{1/2}\ \ell n\left[\frac{\gamma}{4}\frac{v}{2\alpha}(r_{\perp}{}^{1/2} + r_{||}{}^{1/2})^2\right] = \frac{2\alpha C}{L}\,\Delta T \qquad (11)$$

where $r_{||}$ and $r_{\perp}$ are the two principal radii of curvature at the dendrite tip. An initial evaluation of the coefficient of ΔT using commonly published values for the thermal parameters gave almost identical values for germanium and silicon. This would suggest that silicon dendrites would follow the same velocity/supercooling curve as germanium dendrites with the same tip curvature (and probably twin spacing). More recent values for the heat of fusion of silicon[18] indicate that the silicon dendrites would grow about 30% slower than the corresponding germanium crystals.

Since the tip curvature and velocity always occur as a product, an additional constraint is required to determine the propagation velocity as a function of undercooling; in the case of germanium dendrites and thus probably for silicon this constraint is the strong dependence of tip curvature on twin plane spacing.[7] Apparently, the wider spaced pair of twin planes controls the tip radius and the third twin plays some auxiliary role at low tip velocities. In the case of germanium (and probably for silicon) three twin dendrites having one twin spacing in common and one differing all had similar velocity versus supercooling characteristics, and in fact behaved the same as a two twin dendrite having the same common spacing. The difference between the two and the three twin cases was that the two twin dendrites would not grow with less than some minimum melt supercooling whereas the three twin dendrites did not seem to have such a cut-off characteristic.[7] Since web growth velocities are slower than single dendrite velocities, this implies that the effective undercooling at the dendrite tips is small between 0.5 and 1.0°C. This must be reconciled with the previous observation that the melt undercooling is of the order of several degrees.

In studies of the controlled propagation of single dendrites, it is found that self-stabilization of the growth occurs as a result of the interface morphology.[8] The tip region of the crystal approximates a paraboloid of revolution; however, farther back on the interface, the thickening process stops while lateral growth continues giving rise to the so-called "H-arm" structure. The temperature gradients at the growing tip are very large, 10^3 to 10^4 K/cm, so that the undercooling sensed by the tip is the melt temperature only 10 to 100 μm ahead of the tip. The H-arm region is much blunter and generates temperature fields which can in fact extend ahead of the tip. As the dendrite tip grows deeper into the melt, the H-arm region grows wider and blunter and the resulting thermal fields steadily decrease the effective supercooling available to the dendrite tip. At some point, a steady state is established and the result is apparent as a range of dendrite pull velocities for a fixed melt supercooling. In the case of the dendrites bounding a web, the thermal fields from the growing web interface would interact with the thermal fields from the H-arm region to stabilize the dendrite tip position in the melt. Further stabilization of the tip position would occur via any intrinsic temperature gradients in the melt, but such gradients are not required to achieve steady state growth. Any melt gradients, however, must be such that some intrinsic supercooling is available to the dendrite tip several millimeters below the surface (observed dendrite tips grow 0.5 cm or so ahead of the web interface).

3.5 Synthesis of Data

The empirical observations and operating data from previous web studies, the analysis of crucible temperature distributions, and the model for web crystal growth can be used to synthesize a number of general requirements for new web growth systems. A number of these requirements have been intuitively obvious for some time; the present study has yielded some quantitative limits.

First, the thermal geometry of any crucible/heater configuration must have liquid supercooled by 1 to 2°K about 0.5 cm below the melt surface in the plane of the web crystal. This supercooling is necessary for the propagation of the supporting dendrites. Further, in order to avoid generation of spurious dendrites on the growth interface, the liquid at the surface must not be too cold which requires that the vertical temperature gradient must be relatively shallow, e.g. 1 or 2 K/cm. Conversely, the liquid in contact with the containing solid should be hotter than the melting temperature to avoid heterogeneous nucleation and freeze out of the entire melt. Then conditions dictate an adequately shielded covered melt to reduce surface losses and a minimum melt depth of 1.0 to 1.5 cm.

Second, some provision must be made to replenish the melt as the web crystal is being grown. The most reasonable technique would seem to be the addition of material symmetrically on either side in the plane of the web. The hotter liquid needed to melt the feed material will also serve as a thermal barrier to balance the inherent tendency of the web to widen. Further, the addition of feed material to the melt will maintain a constant melt geometry which means that any control system can be dedicated to maintaining the status quo rather than compensating for an extended transient as in previous systems.

Control of the process also is simplified by the fact that the growth of the supporting dendrites and the growth of the web crystal are effectively decoupled in their dependence on growth parameters. The dendrites are apparently strongly influenced by melt temperature but only moderately by the pull velocity; the web is strongly dependent on pull velocity but only slightly on melt temperature. This situation lends itself perfectly to a double loop control where web thickness is maintained by pull speed control and melt temperature is used to maintain the appropriate supporting dendrite size.

Finally, the thermal geometry of the system should reflect the geometry of the crystal. This conclusion is far from novel, and the only contribution of the present work has been to give information on the characteristic size of the "trough" required to meet the thermal requirements.

4.0 CONCEPTUAL DESIGNS

4.1 Preliminary Crucible/Susceptor Designs

The basic thermal requirements for web crystal growth has been given in preceding sections. The mechanical arrangements for several growth system configurations, based on these thermal requirements, are described in this section.

Basically, all the growth system configurations can be grouped into two categories: those designs where the molten silicon is bounded by solid silicon (so-called homocrucible designs) and those designs where the liquid silicon is held in a container or some substance other than silicon (so-called heterocrucible designs). A special category of the heterocrucible design is the cold hearth technique, where the liquid silicon is contained in a cooled metallic crucible. Each of these configurations has its particular advantages and disadvantages; however, the cold crucible technique is an area that has to the present time not been adapted to single crystal pulling and the technical difficulties are such that this technique is relegated to a more experimental approach. The heterocrucible approach is favored from the point of view that prior experience in web growth, and indeed in normal solidification by the Czochralski technique, is available. This approach is recommended as the most practicable and is described first.

4.1.1 Heterocrucible Designs

The use of a separate crucible for containing liquid silicon has a number of advantages insofar as simplicity of design and reliability of operation are concerned. The crucible bounds the melt within a specific shape and volume whereby external heaters and heat sinks can be used to develop the desired undercooled region and thermal gradient in the melt.

One conceptual heterocrucible configuration is shown in Fig. 7. In this design, resistance heaters such as "Globars" pass through a heat leveler which holds the crucible. Presumably, the heaters could be independently controlled to balance the temperature distribution in the melt. Shields on either side of the web limit the heat loss from the growing crystal which in turn interact with the web thickness, pull velocity, and melt undercooling. Finally, the silicon melt is kept at a constant level by adding material from feed rods at either end of the crucible. This feed system would probably require auxiliary heaters which are not shown in the figure.

The reactivity of liquid silicon, however, makes contamination of the melt a major drawback in any design where the liquid contacts a foreign material. The unknown factor is the seriousness of the problem, especially with regard to solar cell fabrication. In a practical sense, however, silicon crystals, including dendritic web crystals, have long been grown from fused quartz crucibles and excellent quality solar cells were subsequently fabricated from the material. Thus, for short periods of the order of twelve hours, there is certainly no difficulty. Studies reported in Section 3.4.2 of this report indicate that fused quartz will, in fact, contain liquid silicon for periods of 120 hours even though the material is devitrified and severely corroded. Therefore, for the present purposes, we will assume that a crucible material other than silicon can be used for web growth systems.

In Figs. 8 and 9 we show a heater/crucible/feed system design which is based both or prior web growth system experience and consistant with the thermal analysis shown here. The crucible, ordinarily of fused quartz, is heated by an induction coupled susceptor, ordinarily constructed of molybdenum. The crucible and susceptor are shaped to give the elongated melt and thermal gradients corresponding to the web solidification shape. This shape removes the rotational thermal symmetry limitations found in the circular melts used in prior web growth programs. Susceptor cross-sectional thicknesses, melt width

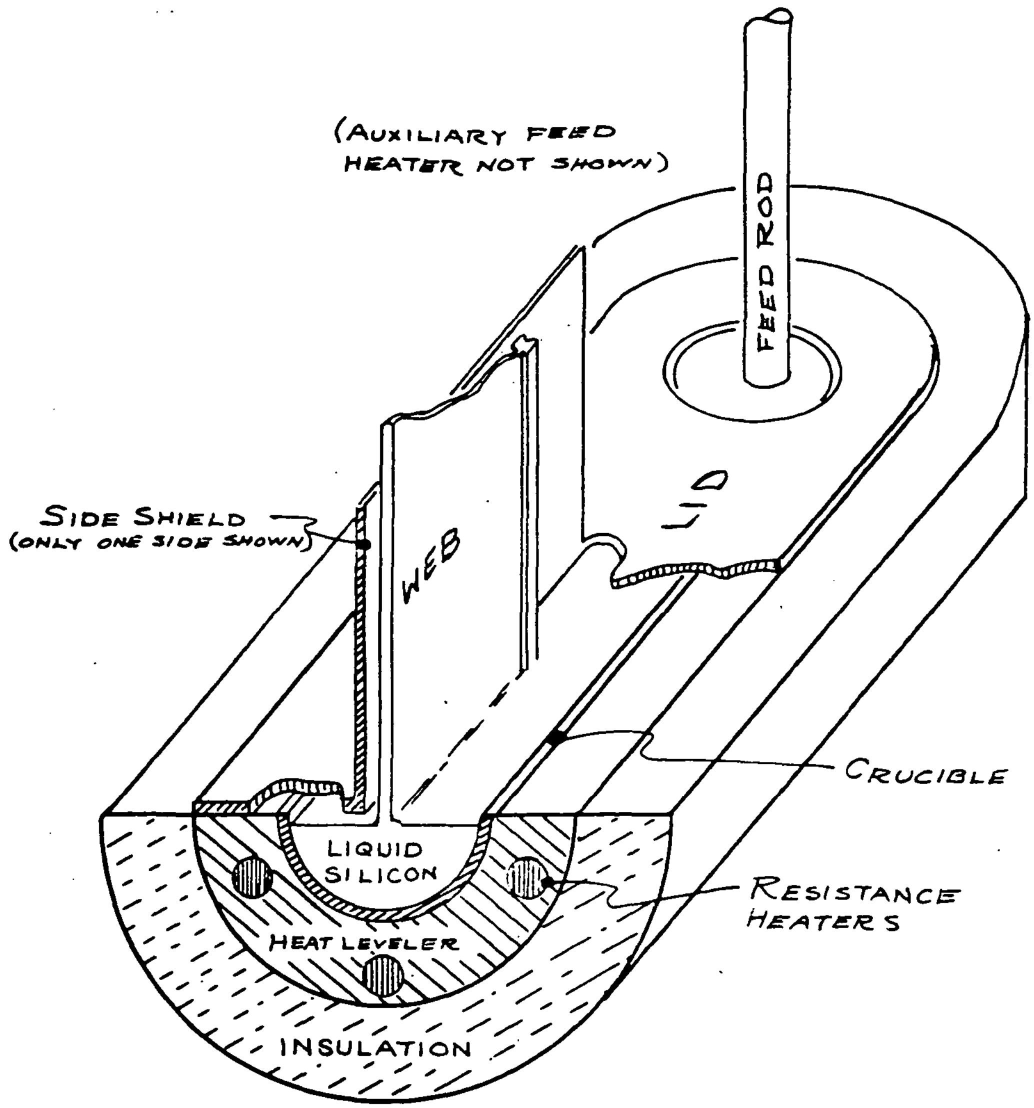

Fig. 7. Schematic concept for resistance heated heterocrucible.

Dwg. 6244A40

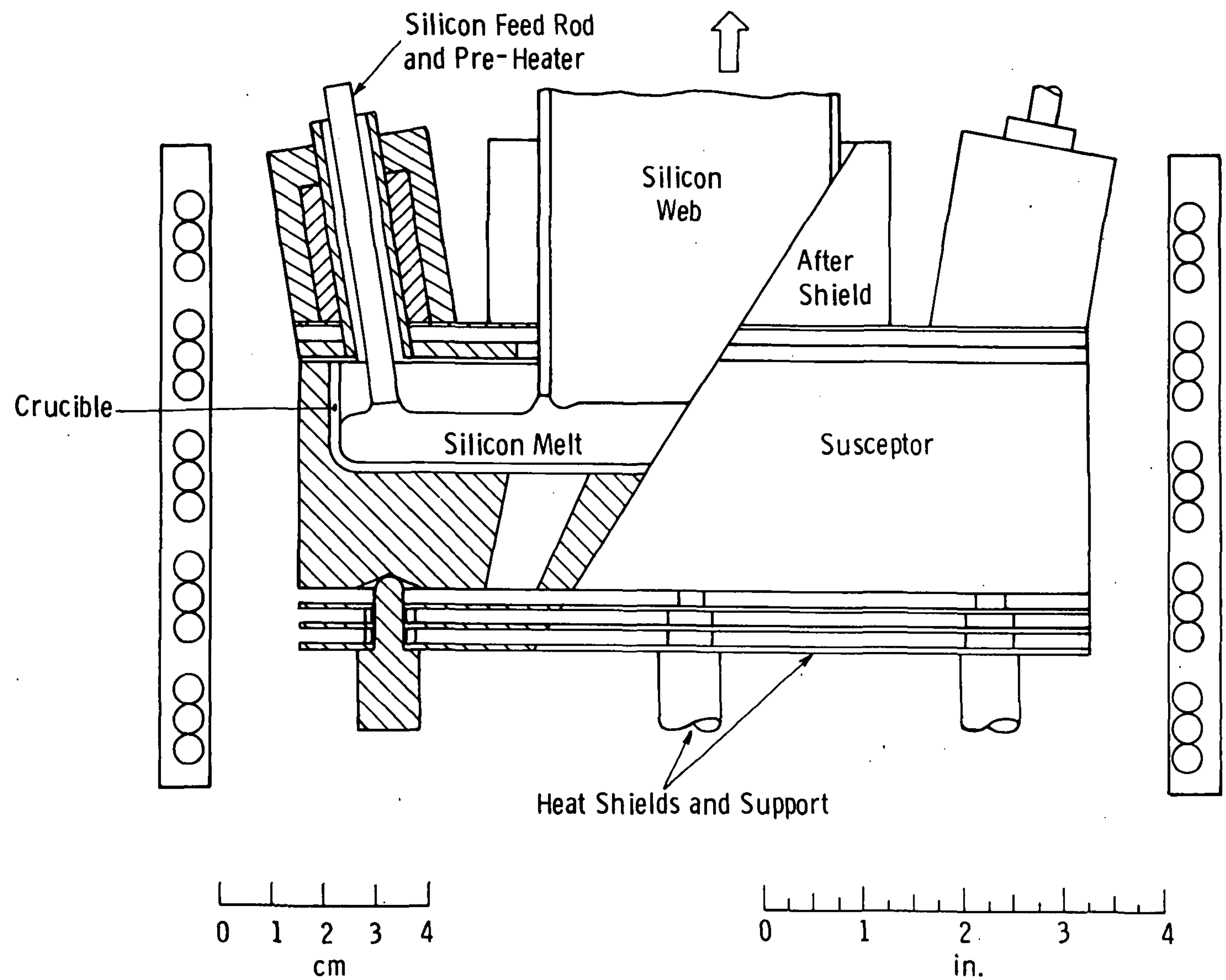

Fig. 8. Side view of induction heated heterocrucible concept.

Dwg. 6244A37

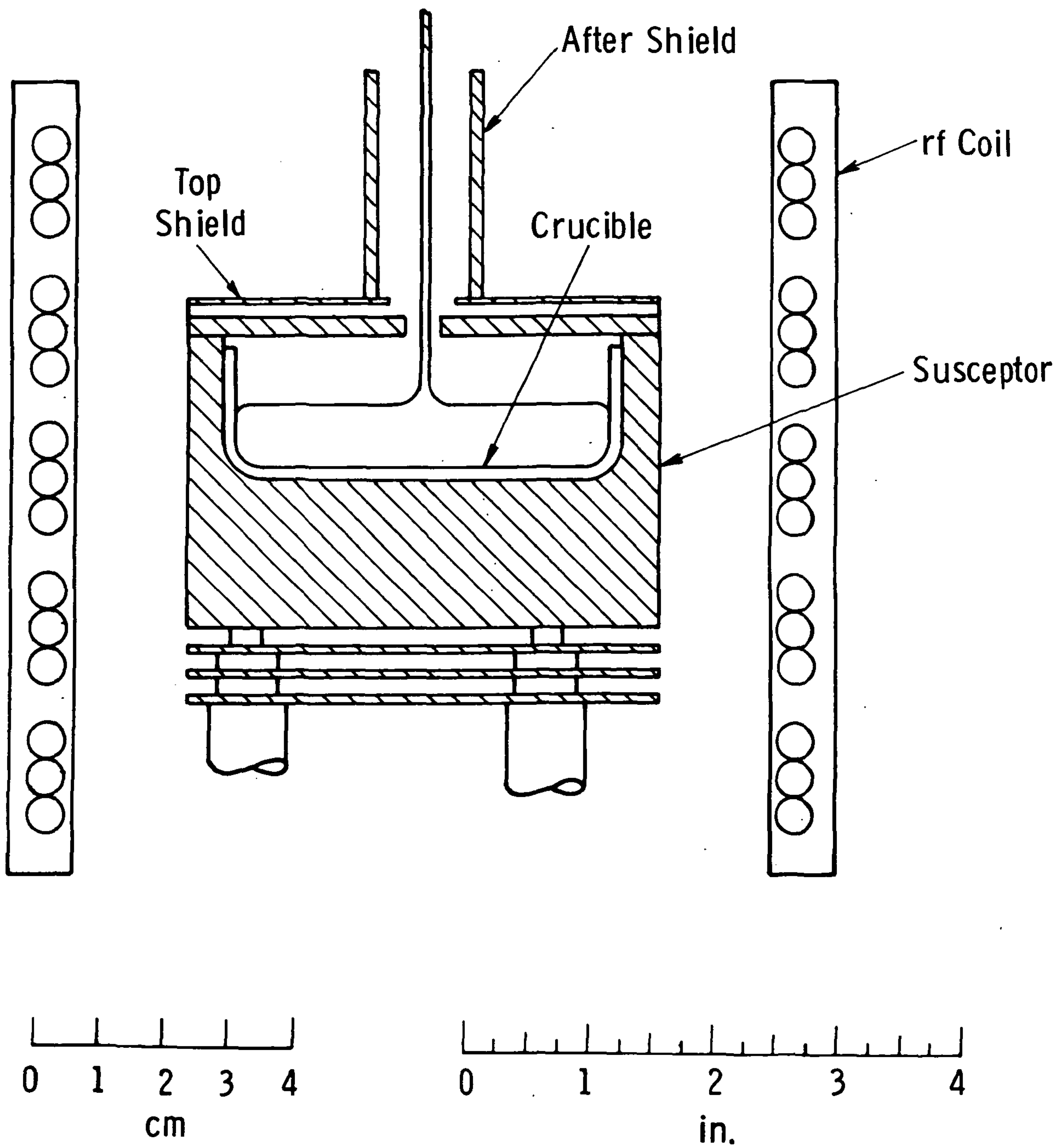

Fig. 9. End view of induction heated heterocrucible concept.

and height, and thermal shielding closely follow the most successfully used designs in previous web growth programs. Silicon feed rods are shown schematically on each end of the elongated crucible, and these represent new technology.

Figures 10 and 11 represent a variation in the growth system, in that the crucible is resistance heated rather than induction heated. The heaters, embedded in a heat leveler, are schematically shown as are the insulation and heat shields. The advantage of this design is that the thermal gradients can be varied by changing the current in the resistance heaters and the thermal shielding about the crucible to optimize the thermal flow into the melt. The feed rod tubes, shown schematically, are resistance heated to obtain a proper temperature at the feed rod-melt interface.

4.1.2 Homocrucible Configurations

In homocrucible configurations, a liquid silicon pool is created in a block of solid silicon by some surface heating technique, such as electron beam heating, laser heating or high intensity lamp. The obvious advantage to such a method is the compatibility of the melt with the container; contamination problems arising from chemical reactions between silicon and the crucible are minimized. As with any system, however, there are a number of problems, especially in the heat source and thermal geometry.

A schematic sketch of a possible homocrucible configuration is shown in Fig. 12. Supplementary heat sources, heat shields, and thermal insulation are not shown for sake of clarity. Feed material would be added at each end of the block and would require separate heat sources for preheating and melting. Several schematic isotherms are sketched on the cross-section.

Although a quantitative thermal analysis has not been performed on this model, there are several problems which have been noted. First, the heat flux from the liquid to the solid silicon must

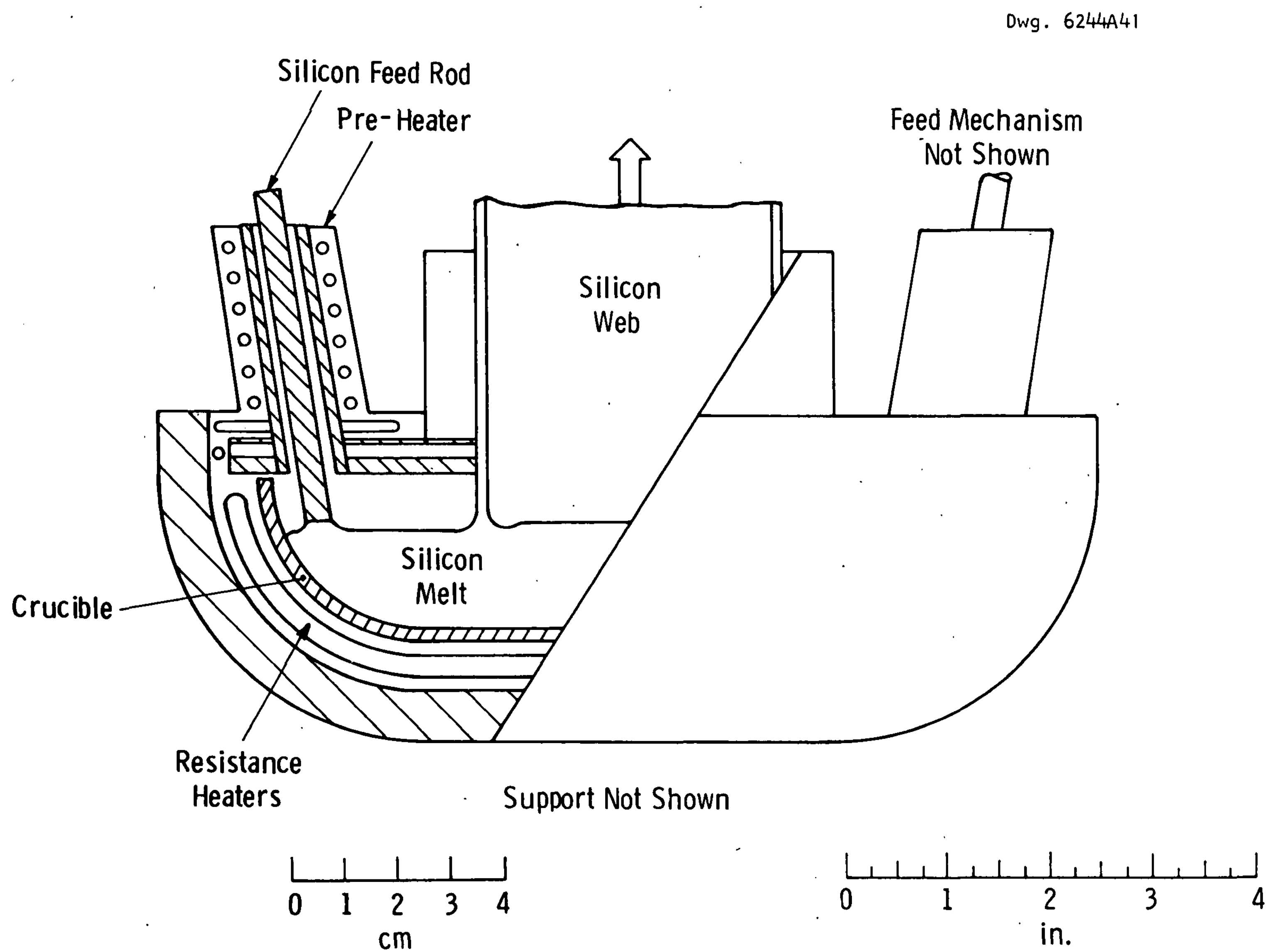

Fig. 10. Side view of resistance heated heterocrucible concept.

Dwg. 6244A38

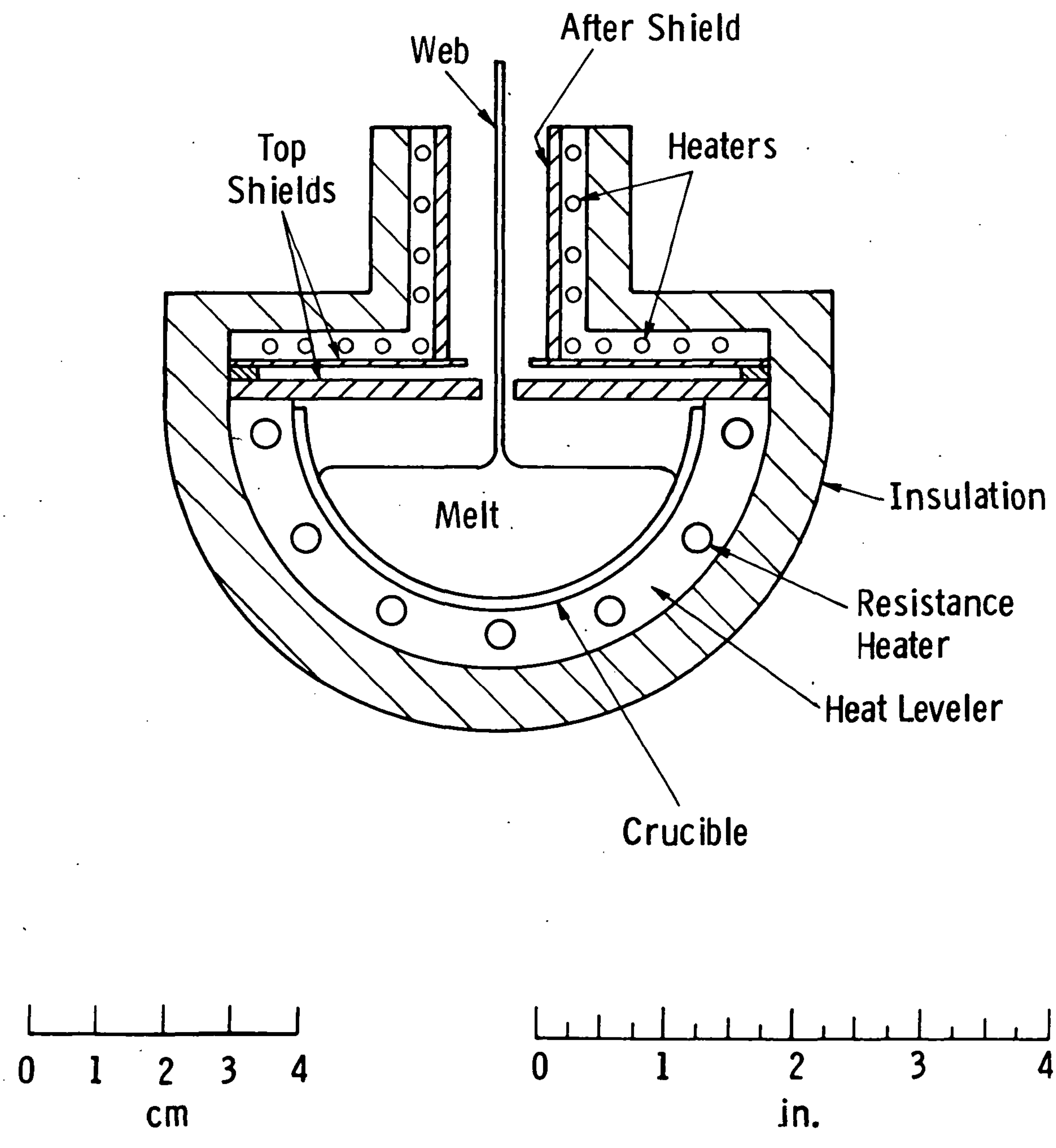

Fig. 11. End view of resistance heated heterocrucible concept.

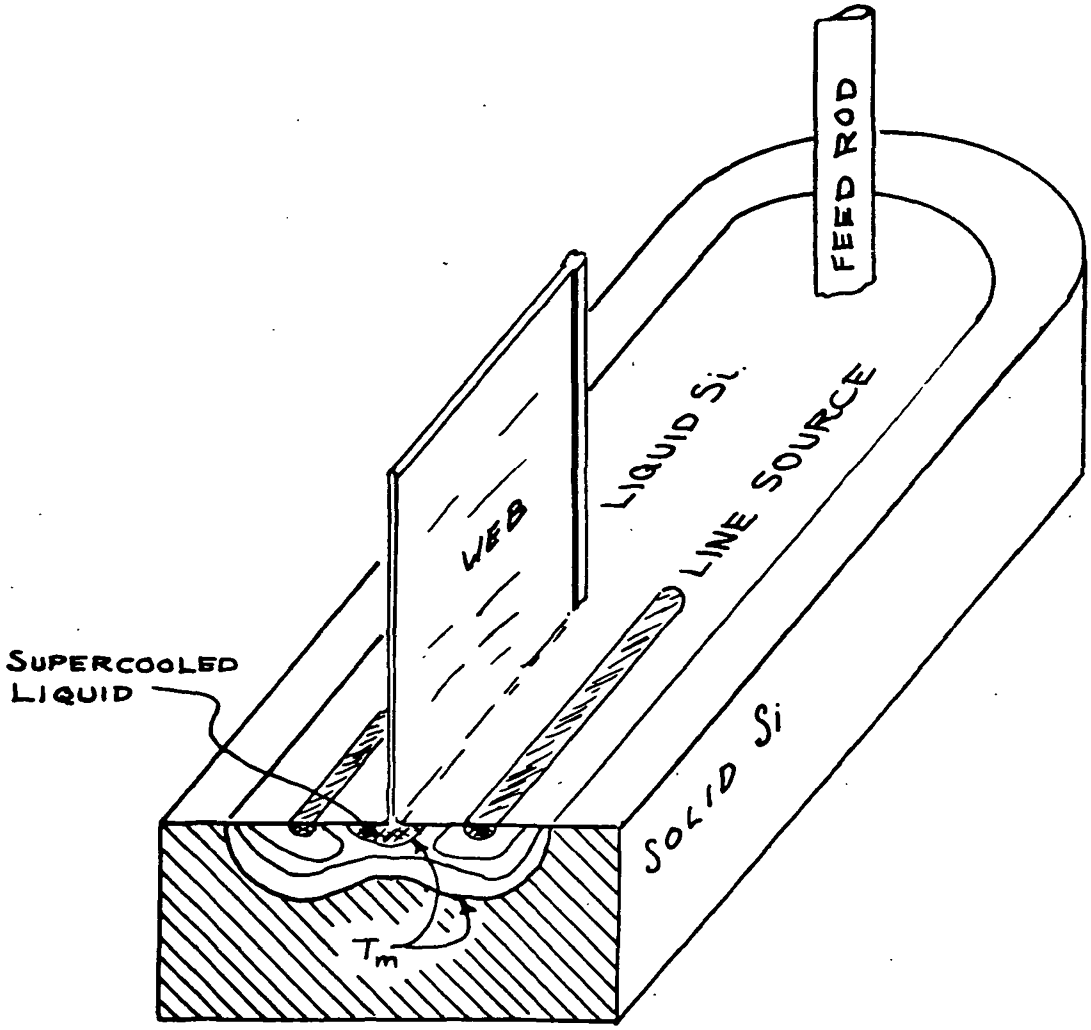

Fig. 12. Schematic homocrucible concept.

create a large enough gradient to stabilize the solid-liquid interface. Otherwise, a slight change in the overall temperature would drastically change the amount of material melted. This problem is aggravated by the fact that the emissivity of liquid silicon is smaller than the emissivity of solid silicon. Thus, as solid silicon melts, the power required to maintain a constant temperature distribution in the system decreases. The configuration is thus inherently unstable and requires all power control to be vested in an external control loop. The whole problem of stability and control requires additional, careful examination.

Another, more subtle, problem which occurs in this configuration is related to convection. The hottest and least dense liquid is located under the line heat sources whereas the coldest and densest liquid is in the supercooled region near the web. The hotter material will tend to rise and spread across the surface of the melt while the colder material will tend to flow directly downward and then spread laterally. The combined effect will be to set up a convective flow pattern where liquid moves across the melt surface toward the growing web and then downward.

This flow pattern has two adverse effects. First, the average temperature near the web is increased which reduces the possible growth velocity. Second, the supercooled region penetrates deeper into the liquid. This raises the possibility of a "plume" of supercooled liquid coming into contact with solid silicon followed by a rapid freeze out of the entire melt. This problem, too, needs a more detailed examination.

Figure 13 shows a more detailed version of this homocrucible concept. The line heat source is provided by a quartz-halogen tungsten lamp enclosed in an elliptical reflector. The silicon block which serves as the crucible is mounted in thermal insulation to reduce heat losses. Depending on the results of a more detailed analysis, it may be necessary to use auxiliary heaters to compensate for the peripheral heat losses from the system.

Dwg. 6244A39

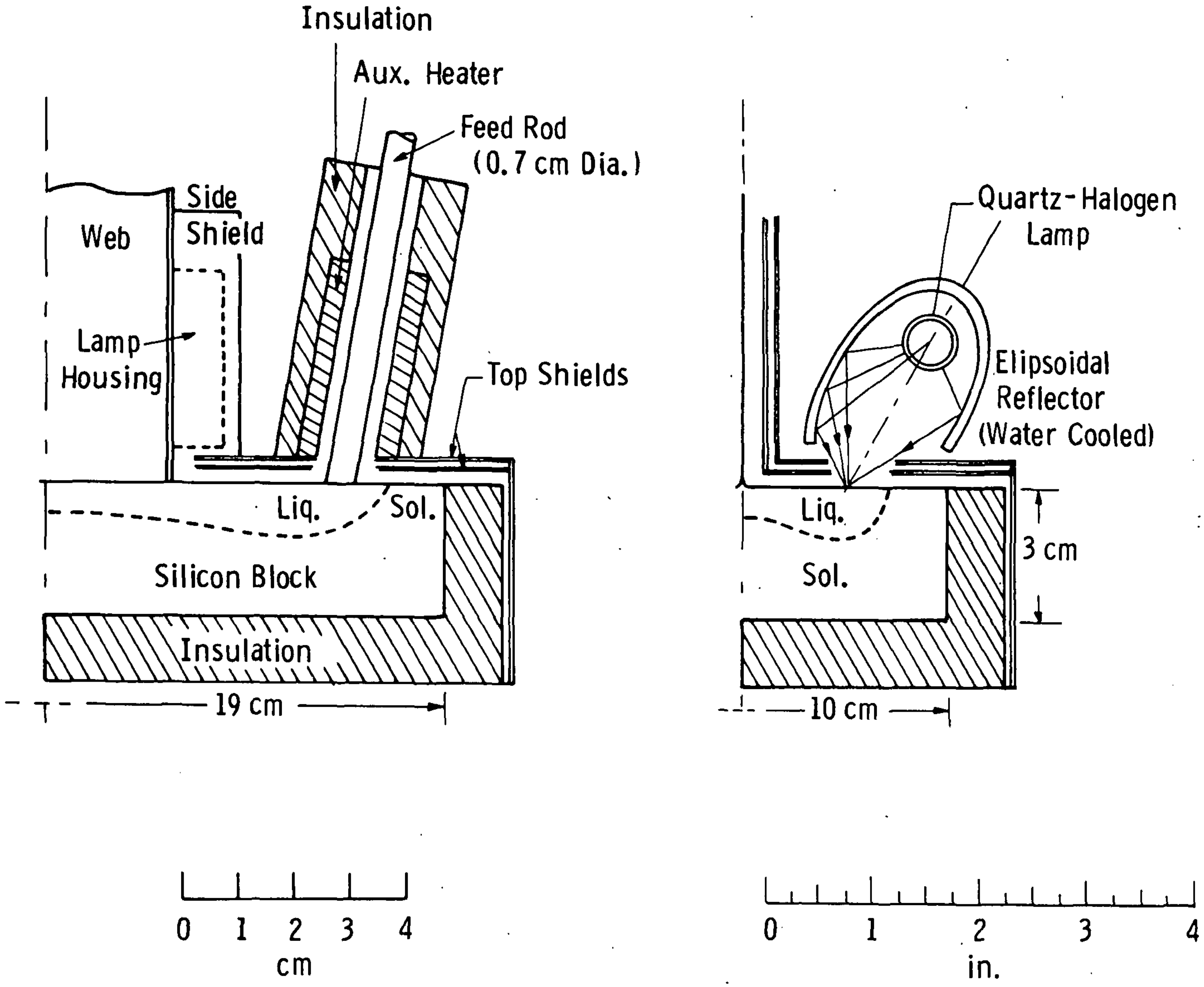

Fig. 13. Side and end sections of homocrucible concept.

4.2 Final Crucible/Susceptor Design

Evaluation of the various conceptual designs for crucible systems led to the choice of an induction heated susceptor with a fused quartz crucible. The choice of induction heating over resistance heating was based more on practical considerations than on technical superiority although there may be some slight advantage of the induction design in heating the crucible ends to melt the feed rods. The practical considerations were based on the reduced number of power feed-throughs required and the immediate availability of the generator and control system. Further, the operating life of resistance heating elements in a silicon atmosphere is not known.

The one major concern with the design was the suitability of a fused quartz crucible for the extended operating times inherent in the quasi-continuous operation concept. This concern, of course, applied to any hetero-crucible system and to settle the question a crucible life test was set up.

The first experiment performed used a graphite susceptor and a small fused quartz crucible. This assembly, with auxiliary shielding, was mounted in a vacuum-tight crystal pulling furnace equipped with a 10 kHz induction work coil. Before attempting to melt down a silicon charge the susceptor was thoroughly outgassed; first to 1200°C in vacuum and then to 1500°C in high purity argon. A charge of about 15 gm of silicon was then added to the crucible and the furnace chamber evacuated and backfilled with argon.

When the charge was melted, two unexpected phenomena occurred: first, the liquid silicon was visibly agitated and pulled to the center of the crucible, and second, a brown "smoke" was evolved which settled as a precipitate on the cooler regions of the crucible assembly. The agitation of the melt was obviously due to the penetration of the electromagnetic field through the susceptor, but the "smoke" was not so easily understood. Later experiments suggest that the stirring of the melt greatly accelerated the reaction of the silicon with the quartz to form silicon monoxide.

The experiment was repeated with a molybdenum susceptor which has a much smaller skin depth than the graphite. As expected, the melt stirring was eliminated as far as any visible agitation; however, a moderate amount of silicon monoxide was evolved. This led to the supposition that the silicon itself might be at fault. In both the experiments, chunks of polycrystalline, vapor deposited silicon were used for the charge, and there was the possibility that some occluded gas might have been present in the charge. To test this possibility, a final experiment was performed using a piece of single crystal silicon as the starting material.

In the final run, the same operating procedure was used as in the previous runs. The results, however, were much different when the charge was melted. Very little silicon monoxide was evolved during the first twenty-four hours or so. The amount of material depositing on the shields and crucible assembly would not have interferred with any growth operation. As the test continued, however, the deposit built up and was moderately dense after 120 hours of operation. At that time, the temperature was slowly lowered and the melt frozen out. Examination of the crucible showed that it was still intact, although it had devitrified and was severely attacked, especially near the top of the melt. The fact that the attack was least at the bottom of the crucible leads to the hypothesis that the reaction rate is controlled principally by the rate at which the reaction product, SiO, can escape. This is consistant with the rapid rate of evolution observed in the violently stirred melt. The effect of the silicon source material is suggestive, but by no means conclusive. What can be said is that with this experiment configuration, the single crystal silicon produced less deposit in a given time than did the polycrystalline starting material.

One may further conclude that quartz crucibles can be used to contain silicon for long periods of time; however, 120 hours of operation is probably marginal if this experiment can be taken at face value.

4.2.1 Mechanical Configuration Details

The final crucible/susceptor mechanical design is shown schematically in Fig. 14 and in more detail in Fig. 15. The crucible would initially be fabricated by hand from 48 mm OD x 45 mm ID fused quartz tubing and the susceptor machined from arc cast molybdenum.

The shield assemblies would be fabricated from molybdenum sheet rolled from arc cast material. The construction of these assemblies would be such that gas flow between the shields would be minimal to avoid the deposition of any SiO, silicon, etc. which could degrade the performance. Although not shown in the drawings, it may be desirable to have a small gap or channels between the top shield assembly and the susceptor to promote a gas flow over the melt to reduce the deposition of material in the grow slot area. This would be a simple modification to the design which could be implemented easily if warranted by experiment.

The lower shield assembly would be less complex than the top assembly. Again, molybdenum sheet or foil would be used for the elements. The elements would simply be stacked on the susceptor mounting posts. It is not known whether or not gas circulation must be minimized with the lower shields; however, this could be done easily if required.

4.2.2 Design Analysis

The analysis of the final heater/crucible design followed the same procedure discussed in detail in Section 3.3. The only difference in the general treatment was the coding of the finite element program (WECAN) to reflect the translational symmetry of the new design. The geometry analyzed was the crucible/susceptor cross-section shown as Section B-B in Fig. 15. The initial analysis was performed with isothermal boundary conditions; later, the analysis was repeated with boundary flux conditions appropriate to an induction work coil centered with respect to the susceptor.

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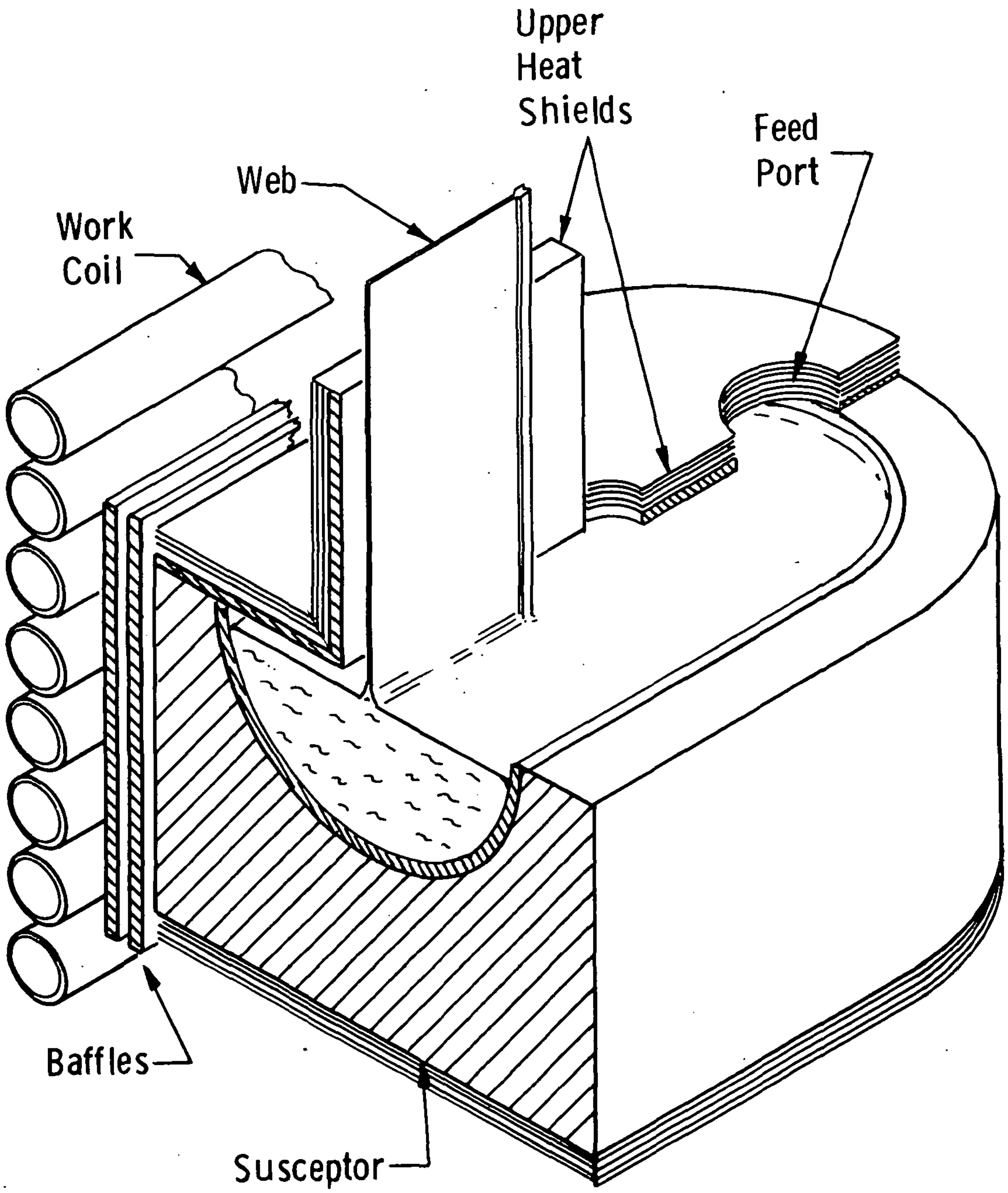

Fig. 14. New dendritic web design.

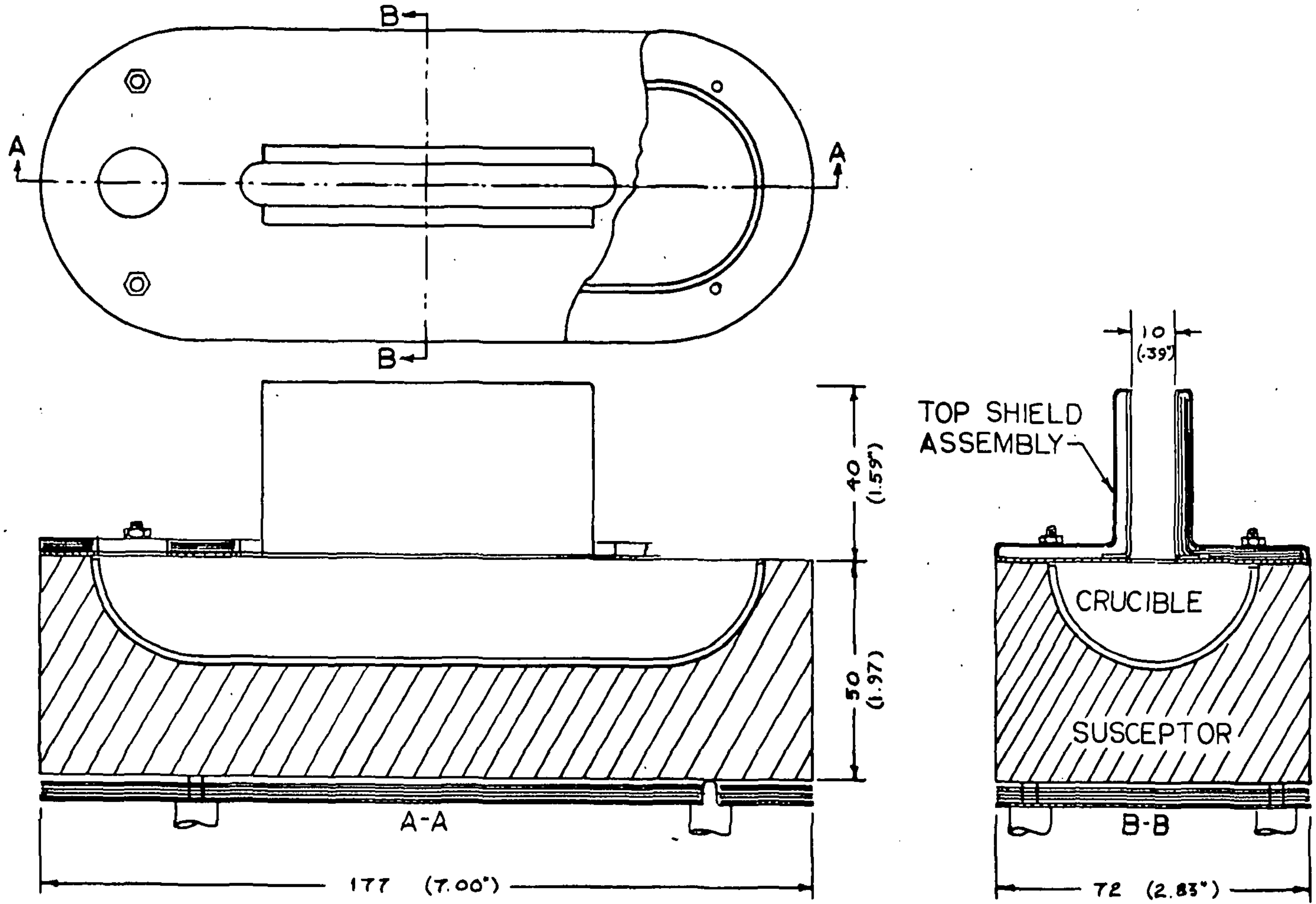

Fig. 15. New susceptor/crucible design.

Isothermal Boundary Condition

The thermal analysis of the crucible/susceptor design was first run with the side wall temperature specified at a constant value (1720°K). The resulting temperature distribution in the liquid silicon is shown in the left-hand side of Fig. 16. The general shape of the isotherms approximates the shape of the crucible ensuring that the liquid in contact with the walls will be hotter than the melting temperature when the axial liquid is supercooled. The magnitude of the axial gradient is about 5.5 K/cm which is about twice as large as desired. This gradient can be reduced to the desired magnitude by two means: increasing the effectiveness of the top shields and changing the position of the induction work coil.

The specific design of the top lid considered in the analysis assumed four molybdenum shields in the assembly. The number of shields can easily be increased by a factor of two or more by using thin (12 μm) molybdenum foil interstitial shields between the heavier main shields. The effect of the work coil position will be discussed in the next section.

Eddy Current Analysis and Heat Generation Boundary Conditions

Experience in growing dendritic web crystals has shown that the relative position of the induction work coil and susceptor is very important in achieving proper growth conditions. In order to understand the effect of coil position, a field mapping program was used to calculate the square of the eddy current density (power density) as a function of position in the susceptor. Various coil lengths and positions were investigated and a wide variation in power profiles could be generated. These power profiles could then be compared with the boundary heat fluxes concommitant with the isothermal boundary conditions analyzed in Section 4.2.2.1. When the work coil was about 50 percent longer than the height of the susceptor, the power generation rate and heat flux

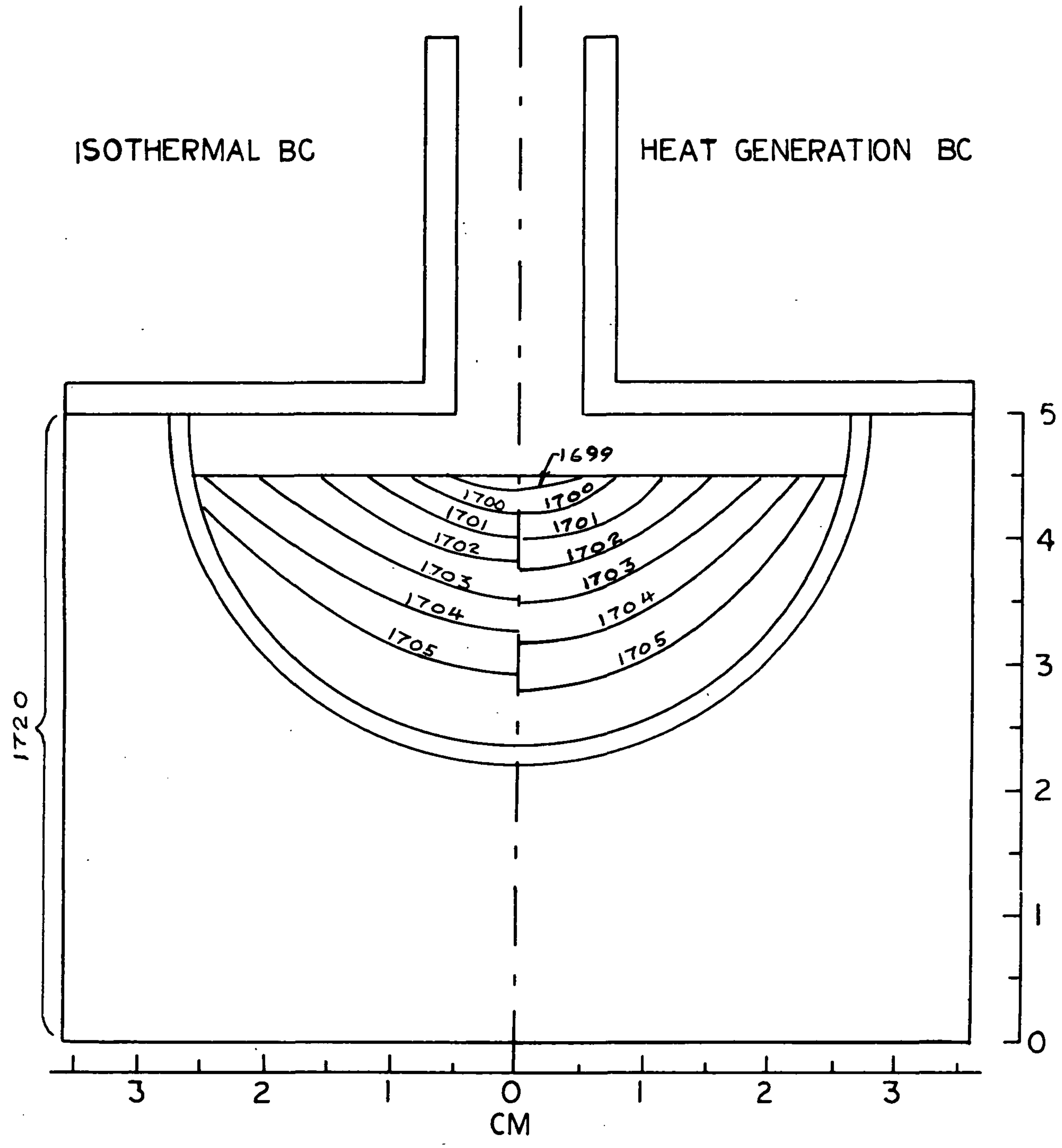

Fig. 16. Isotherms in silicon melt with two different boundary conditions (BC).

profiles were very similar. This general coil size was also very similar to the one actually used for growing silicon dendritic web.

The normalized heat generation rates for two positions of an 8.8 cm long work coil are shown in Fig. 17. The boundary heat flux for a crucible with isothermal walls is also shown for comparison. Although neither of the induced power profiles agrees exactly with the flux from the thermal analysis, the general agreement is reasonable. The remaining comparison was to use the calculated heat generation rate as the boundary condition for the thermal analysis.

In order to apply the eddy current mapping to the thermal analysis, the total induced power was normalized to agree with the total power requirements of the isothermal boundary. The resulting distribution was then imposed on the thermal model and a new temperature distribution calculated. The result for a centered work coil is shown in the right-hand side of Fig. 16. The isotherm geometry is very similar to the isothermal result; however, the horizontal gradient is slightly larger while the vertical gradient is slightly smaller. This is in the desired direction and indicates that some adjustment in the thermal geometry can be made with coil positioning.

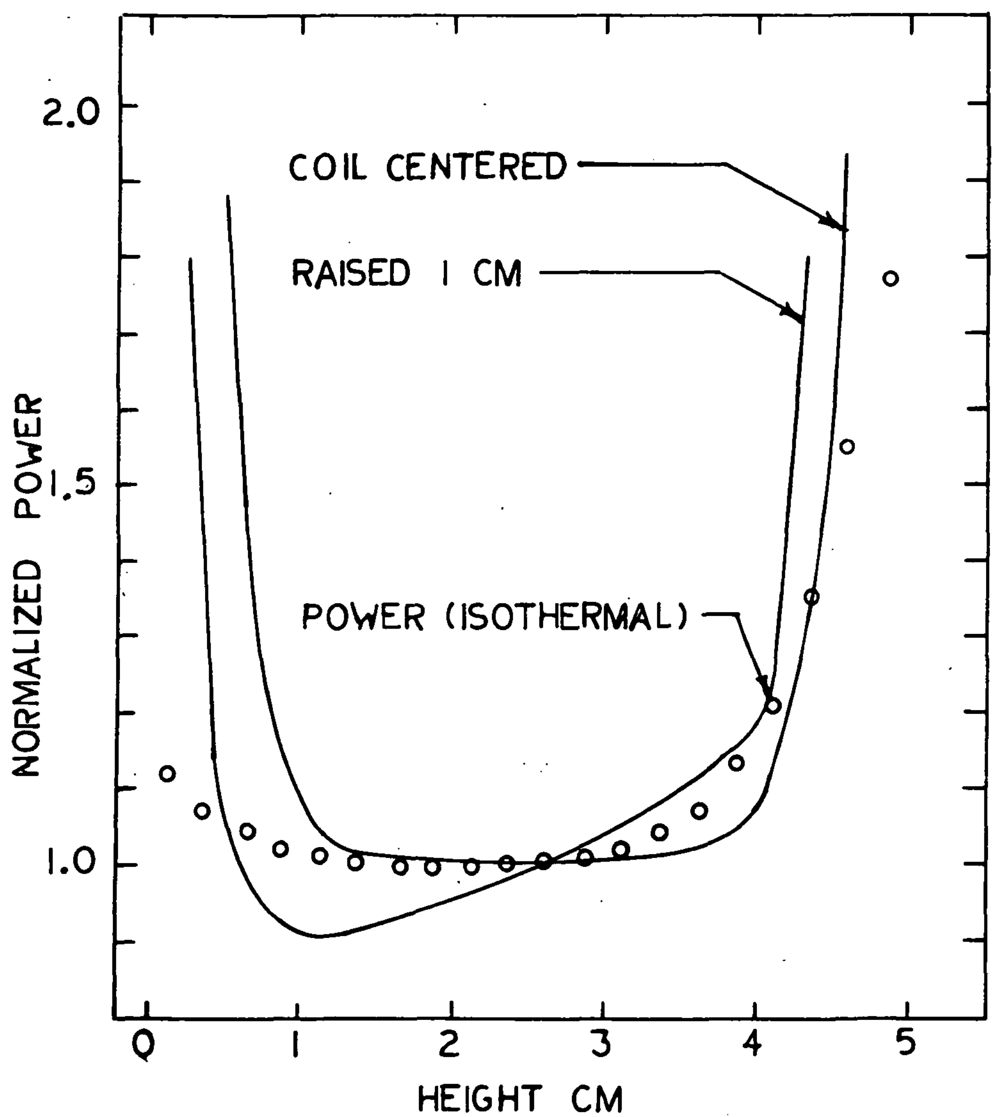

Fig. 17. Power generation profiles in susceptor wall for two work coil positions and for an isothermal wall.

4.3 System Design Concepts

4.3.1 Design Objectives

A major objective of this program was to generate a conceptual design for apparatus which would satisfy the web growth requirements as were identified during the program study tasks. Although the intent was to carry the design only to the conceptual stage, during the course of the program it was found to be advantageous to develop certain design concepts in much greater detail than had been originally intended. This need became apparent in order that some features of the design could be shown to be practical. Some of these drawings are not yet in fully developed final detail. Hence, as an aid to understanding the features of this design, a simplified representation is shown in Figs. 18, 19, and 20.

Three key design objectives, each of which is satisfied by the conceptual design, are discussed briefly as follows.

Demonstrable Capability. Apparatus constructed according to the design must be capable of demonstrating, by means of actual web growth, the ability to produce silicon dendritic web as predicted by the study conclusions of this program.

Provide Design Data. The facility defined by this design is best described as a laboratory prototype apparatus. As such, one of its purposes will be to have sufficient versatility and complexity that it can utilize and evaluate a variety of alternative methods or design

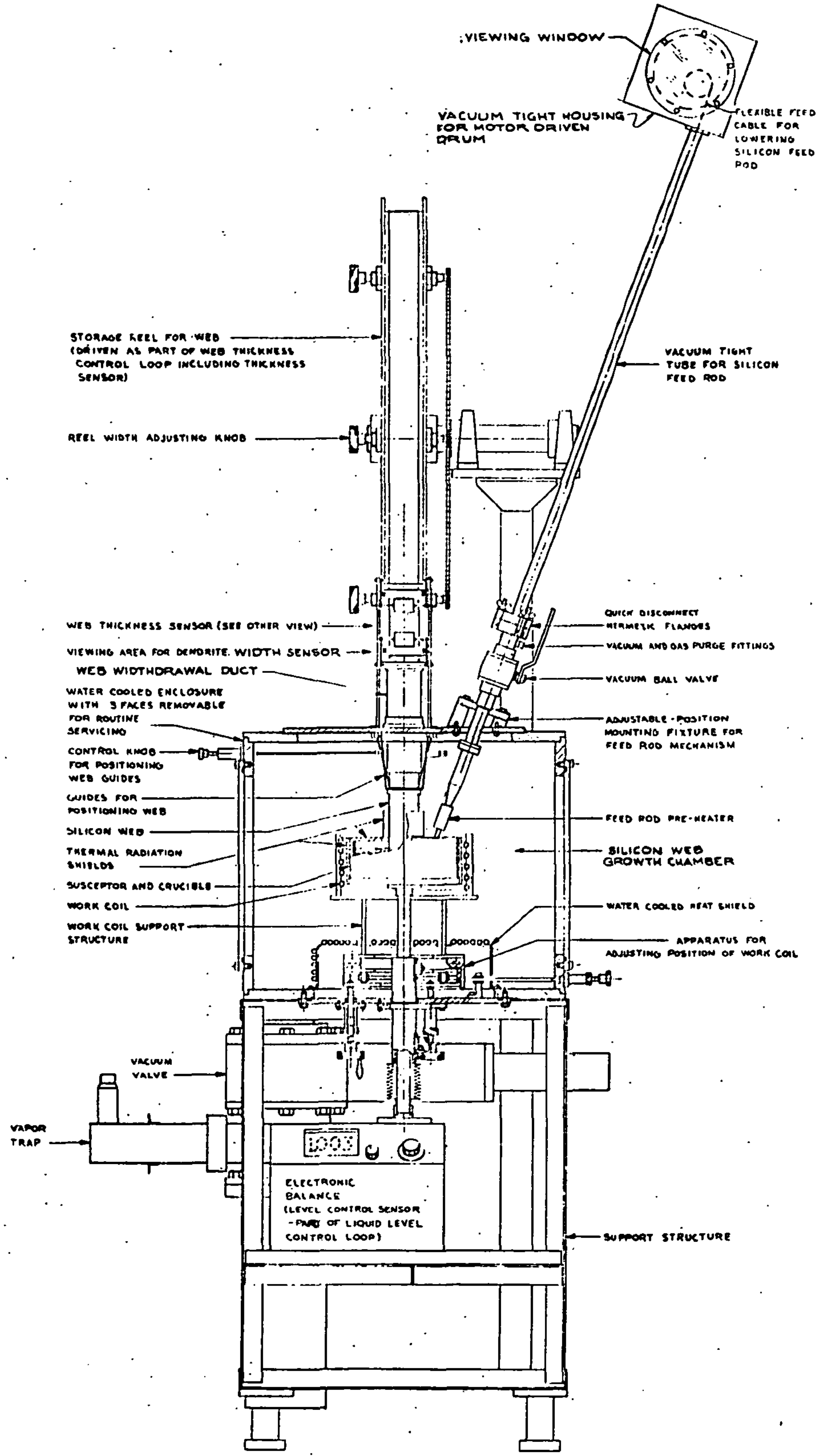

FIG. 18
SILICON WEB FURNACE
CONCEPTUAL DESIGN
FRONT VIEW

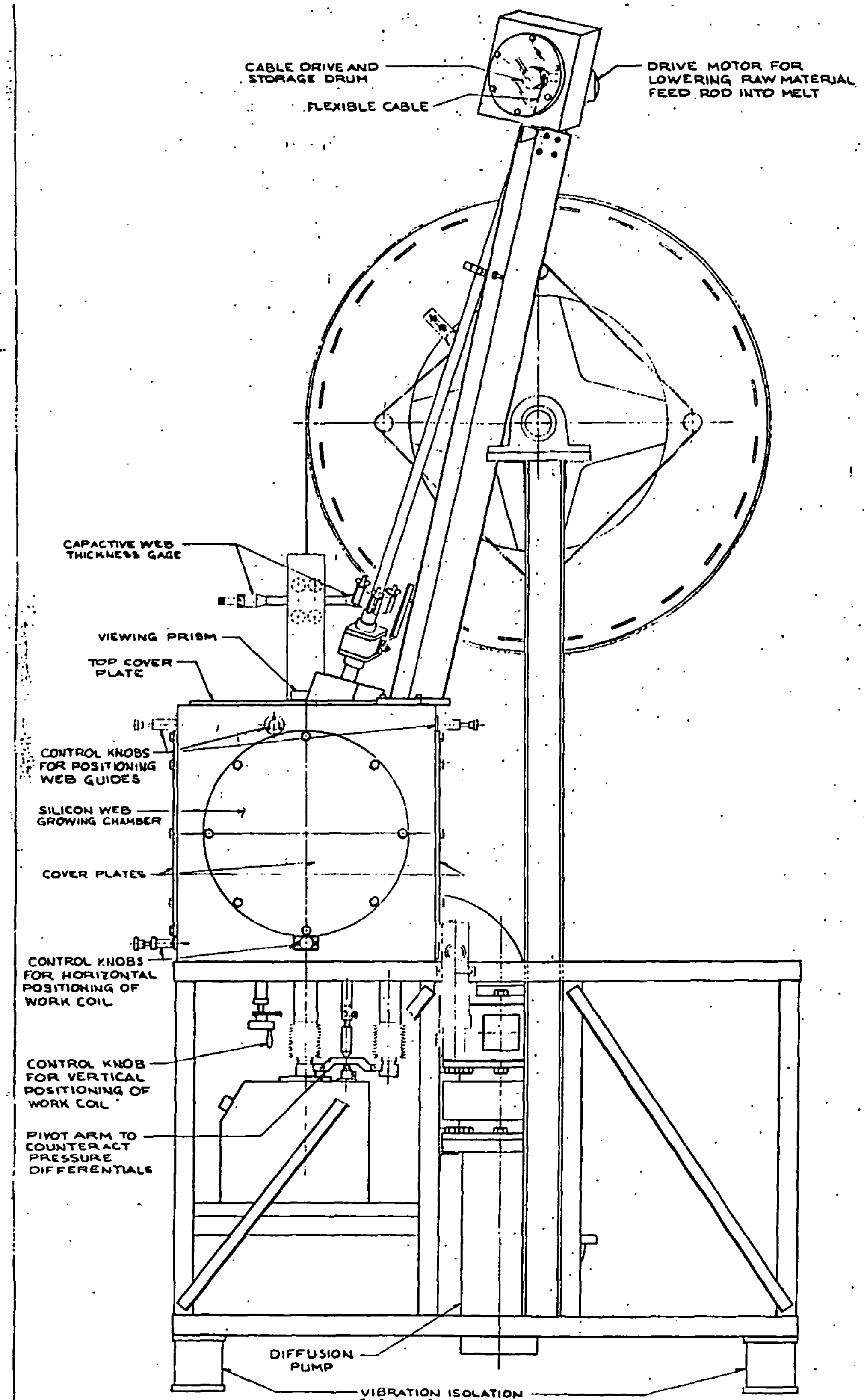

FIG. 19
SILICON WEB FURNACE
CONCEPTUAL DESIGN
SIDE VIEW

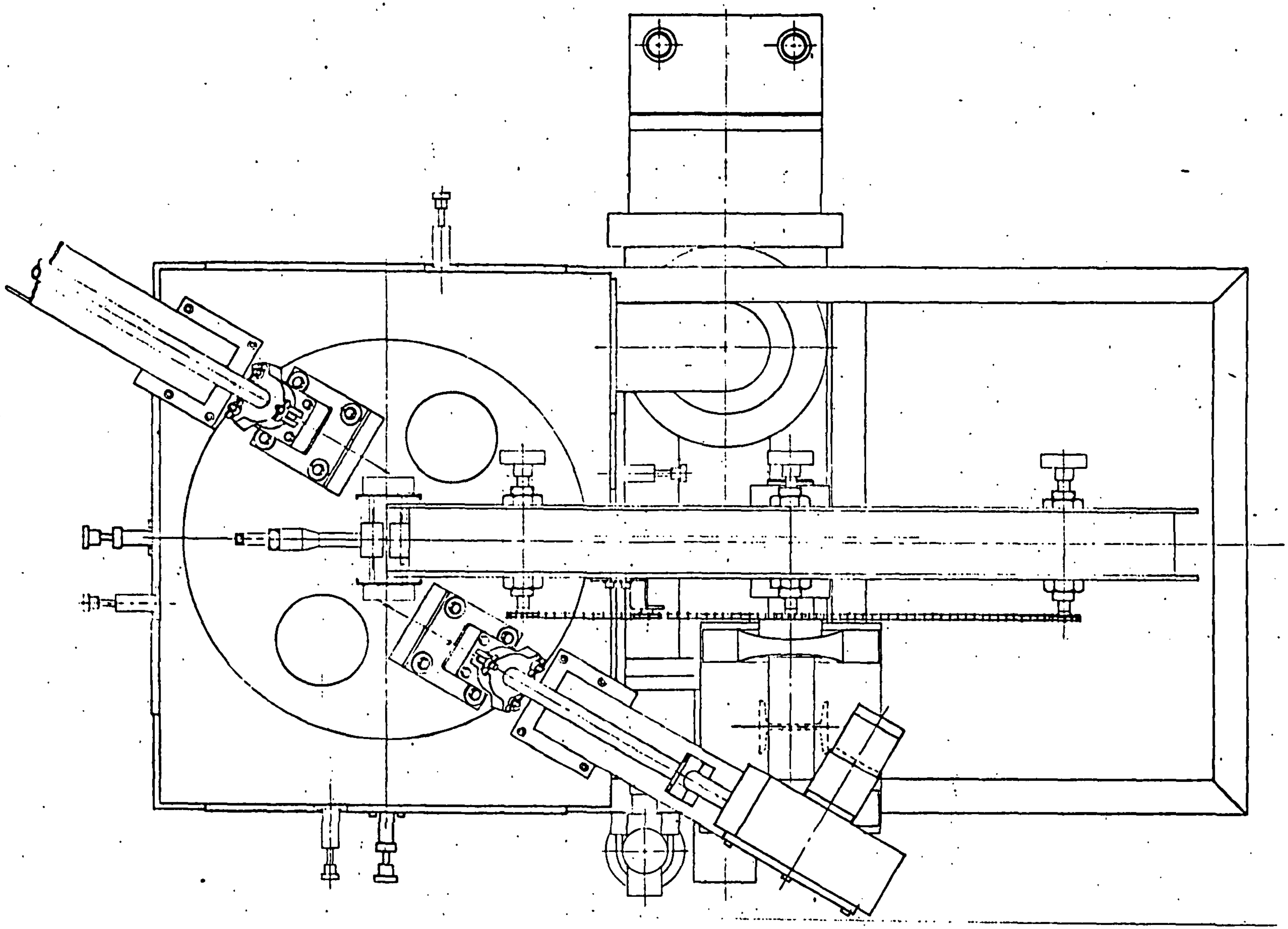

FIG. 20.
SILICON WEB FURNACE
CONCEPTUAL DESIGN
TOP VIEW

features. This capability will serve a dual purpose. It will allow the facility to be tailored to fit growth requirements and it will identify the design data needed to develop a detailed design for an efficient, relatively simple and economical production prototype web growth facility.

Provide Cost Data. The versatility of the laboratory prototype facility built according to this design will allow use of selected process features which will provide data essential to an economical evaluation of the web growth process.

4.3.2 Basic Approach to Design

As noted above, wide versatility and flexibility are major features of this design. To obtain these features the main housing of the growth chamber was selected as a water-cooled, stainless steel rectangular envelope of 18 x 18 x 24 inches. Each of the six faces has a 14-inch diameter opening which is hermetically closed by a readily removable cover plate. The cover plates provide access for a majority of the required envelope penetrations (water, gas, power and instrumentation leads, web, raw material feed, etc.). Also, much of the required internal and external mechanism is mounted on the cover plates. Thus, it is not only possible but, more importantly, relatively convenient to exchange a large portion of the operating features of the facility. Thus, both major and minor design modifications and adjustments may be efficiently implemented. Another advantage of this design is excellent accessibility -- some covers are blank -- which enhances modifications, adjustments, cleaning and set-up for operation.

4.3.3 Structural Features

Growth Chamber. The basic approach to the design and the functional features of the growth chamber were discussed above. The growth chamber will be permanently affixed to a heavy metal frame which will also support supplementary apparatus such as the vacuum pumping system and the web withdrawal and storage reel. The frame will be heavily weighted in the lowermost portion of its structure as a means of lowering the center of gravity and reducing vibration. Space is provided for shock mounting if necessary.

Crucible and Susceptor. The crucible and susceptor were described previously and will be located at roughly the geometric center of the growth chamber. The space provided is generously adequate to allow for any anticipated crucible/susceptor design modification and for modifications of the work coil and adjacent thermal insulation. In the event that direct resistance heating should be required, the space and access is more than adequate. Initially, the crucible and susceptor will be heated inductively by 10 kilohertz power.

Work Coil Positioning. The work coil mounting and positioning structure is mounted on the bottom cover plate inside the lower region of the growth chamber. Three-direction position adjustment is provided by way of externally located manually operated control knobs. Horizontal positioning is needed as a means for obtaining thermal balance within the crucible; vertical positioning is useful as a means for adjusting the vertical gradient within the crucible.

Web Withdrawal Route and Mechanism. A 36-inch diameter speed-controlled reel will serve the dual purpose of a storage and a withdrawal mechanism for web as it grows. The web will be routed vertically through a withdrawal duct which is provided with sufficient gas flow to prevent harmful entry of air into the growth chamber. Adjustable positioning guides for the web will be located inside the growth chamber below the withdrawal duct. Above the withdrawal duct additional guides will automatically center the web as it leaves the duct and passes through control loop sensors. The storage and withdrawal reel is adjustable for a wide range of web widths and automatically centers for all width adjustments.

Raw Material Feed Mechanism. The versatility of the conceptual design will accommodate alternative methods for continuous replenishment of the melt with raw silicon. The method which was selected will utilize slim ($\sim$ 3 to 6 mm diameter) polycrystalline rods which are mass produced as a key step in the commercial production of semiconductor grade silicon. The selected design will utilize rods of approximately one meter length and will allow re-loading as often as necessary. Dual feed mechanisms, one at each end of the crucible, will be used alternately in order to feed raw silicon by gravity at a continuous, uninterrupted rate. Raw material preheaters, located near the melt, can be used with each feed mechanism and in addition to preheating the feed rods can be used continuously in order to help maintain thermal balance in the crucible. The feed mechanisms will be mounted entirely on and through the top cover plate. If found to be desirable for the purpose of evaluation, the mechanisms can be converted to a pellet feed method.

Atmosphere. The growth chamber, feed mechanisms and the web withdrawal duct can be purged by high vacuum ($\sim 10^{-6}$ torr) prior to web growth. The design, as presently planned, will utilize an inert gas -- argon -- as an atmospheric medium during web growth. Except for the web withdrawal duct the design provides for full range of pressure from high vacuum to a maximum in excess of one atmosphere. Present intention is to temporarily seal the web withdrawal duct during purging prior to the establishment of web growth. The need of the capability of growing web under vacuum or reduced pressure has not at this time been established and is not planned. The current design will allow conversion to vacuum and reduced pressure under growth conditions, if later found to be necessary, although the cost would be relatively high.

Vacuum Pumping System. Vacuum pumping capability is a built-in part of the design. The high vacuum pumping will include a high conductance gate valve and liquid nitrogen vapor trap in conjunction with a diffusion pump and mechanical pump. Vacuum roughing will be handled by a separate mechanical pump, valve and pump line. Although not presently planned, a hot metal getter has been considered for use inside the growth chamber and can be added if later desired.

Viewing Capability. Ability to view the growth region of the melt is mandatory during the growth start-up procedure. After growth is established, viewing is essential not only for visible monitoring of the growth but additionally for adjusting the web positioning guides inside the growth chamber. To satisfy these needs viewing prisms will be located on the top access plate immediately adjacent to the withdrawal duct in order to provide viewpaths, parallel to the web, to the growth region of the melt surface. View ports will also be located in the front and in one of the side access cover plates.

4.3.4 Sensing and Control

Web growth will ultimately be governed automatically by three separate but related control loops. Although the exact design of control loops is yet to be determined, adequate space and accessibility has been provided such that final choices can be installed and evaluated without difficulty. Initially, web growth will be controlled semi-automatically with manual operation decisions based on observations as well as sensor outputs not tied into control loops. The information gained by this experience will result in the finalizing of the control loops design and, subsequently, automatic control of the full process. The three control loops are discussed in the following sections.

Dendrite Width/Melt Temperature Control Loop. Within the normal range of withdrawal rate the width of the dendrites will depend primarily upon the melt temperature. It is presently anticipated that a scanning optical sensor will be used to measure dendrite width and will be located immediately above the web withdrawal duct. For the system to function for web growth it is necessary that the melt temperature be held within a very narrow range essentially at the melting point of silicon. This will be performed by means of a relatively conventional temperature control loop system similar to those commonly used for Czochralski growth of silicon. The dendrite width sensor will be joined with the conventional system as a subsystem loop to provide control point offset as required to maintain the desired dendrite width. Because the dendrite sensing occurs several minutes after growth the response rate for correction of control point offset must be adjusted accordingly.

<u>Web Thickness/Withdrawal Rate Control Loop</u>. Assuming that the melt temperature is held within the narrow range indicated in the previous paragraph, the web thickness will depend primarily upon the withdrawal rate. An electronic capacitive thickness sensor will be located immediately above the dendrite width sensor and will be used in a control loop with the motor and mechanism which drives the withdrawal and storage reel. The response rate for this loop must also be adjusted to compensate for the fact that sensing occurs several minutes after growth.

<u>Melt Level/Material Feed Rate Control Loop</u>. An electronic balance will be employed to provide continuous sensing of the crucible weight and, consequently, the melt level. The electronic balance is located below the growth chamber and will provide a dc voltage indicative of the melt level. The control loop will combine the balance and the

motor drive units which lower raw silicon rods into the melt. The weighing system will be insensitive to growth chamber internal-external pressure changes during operation by virtue of a pressure balancing arrangement which is built-in. An optical melt level sensor has been considered as an alternative to the weight sensor and could be used as a backup if needed.

Other Sensing and Measurements. During the evaluation and development phase of furnace use, it is expected that numerous temperature measurements will be required in several parts of the system, especially the crucible and susceptor. Adequate accessibility has been provided to serve this need.

5.0 CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

The most important conclusion to be drawn from this study is that the dendritic web growth process is a practiable technique for growing wide, thin silicon ribbon. Not only can the necessary thermal conditions be met by a reasonable crucible/susceptor configuration, but the control requirements can be met by commercially available technology. Further, a preliminary analysis indicates that the growth process is self-stabilizing in several aspects which may lead to an advantage over other methods of silicon ribbon growth.

On a more technical level, a number of requirements were clarified or developed and in most instances quantitative limits could be established. The necessity for a temperature field compatible with the web geometry has been obvious; the present work established the magnitude of the temperature gradients required. The analysis led to the conclusion that latent heat at the growth front should flow to the melt as well as to the web thus permitting faster pull rates than in other techniques. The analysis further indicated that the growth should be stable in the presence of small temperature variations and thus the requirements on the control system are somewhat relaxed.

To the extent that there are strong crystallographic factors governing the web growth process, it may be even easier to control than other techniques. For example, the growth of the dendrites is strongly governed by the crystallography of the system and this in turn ensures that the web faces are extremely close to (111) planes and thus likely to grow as almost atomically flat surfaces. Further, the growth processes of the dendrites and of the web section are only very loosely coupled which permits the use of independent servo loops on melt temperature and pull speed to control the two growth modes essentially independently. In fact, by sensing the dendrite size and the web thickness, actual temperature control may be relegated to maintaining short term stability in the system, a requirement met with little difficulty with current commercial apparatus.

Two basically different crucible/heater configurations have been considered for web growth: heterocrucible systems where the silicon liquid is held in a container of foreign material, e.g. fused quartz, and homocrucible systems where the liquid silicon is contained by solid silicon. The heterocrucible designs are extensions of (successful) prior practice and are relatively simple in design. They have the disadvantage that chemical reactions can take place between the liquid silicon and the container. The homocrucible designs have the advantage that contamination problems are minimized, but suffer from the serious disadvantage that the heater system and thermal design would be extremely complex. Creating the supercooled liquid region necessary for dendritic web growth is a design problem of formidable proportions with a homocrucible concept.

Two types of material feed systems have been considered: a continuous rod feed and a continual pellet feed. Both techniques seem practicable; however, the continuous rod feed seems simpler for a laboratory apparatus. The control of the feed rate could be accomplished by several different mechanisms and both an automatic weighing technique and an optical sensing technique have been considered.

5.2 Recommendations

Based on the results of this study, it is recommended that a laboratory facility be constructed for growing silicon dendritic web from a quartz crucible/molybdenum susceptor. The initial design should incorporate a continuous rod feed for replenishing the melt controlled by an automatic crucible weighing system. The facility design should, however, be sufficiently flexible to easily accommodate a wide range of modifications which might be suggested by operating experience.

REFERENCES

1. Billig, E., "Growth of Monocrystals of Germanium from an Undercooled Melt," Proc. Roy. Soc. A 229, 346-63 (1955).

2. Bennett, A. I., and R. L. Longini, "Dendritic Growth of Germanium Crystals," Phys. Rev. 116 (1) 53-61 (1959).

3. Hamilton, D. R., and R. G. Seidensticker, "Propagation Mechanism of Germanium Dendrites," J. Appl. Phys. 31, 1165-1168 (1960).

4. Faust, J. W., Jr., and H. F. John, "Germanium Dendrite Studies: I. Studies of Twin Structures and the Seeding Mechanism," J. Electrochem. Soc. 108 (9) 855-859 (1961).

5. Faust, J. W., Jr., and H. F. John, "Germanium Dendrite Studies: II. Lateral Growth Processes," J. Electrochemical Soc. 108 (9) 860-863 (1961).

6. Faust, J. W., Jr., and H. F. John, "Germanium Dendrites Studies: III. Dislocations," J. Electrochemical Soc. 108 (9) 864-868 (1961).

7. Hamilton, D. R., and R. G. Seidensticker, "Growth Mechanisms of Germanium Dendrites: Kinetics and the Nonisothermal Interface," J. Appl. Phys. 34 (5) 1450-1460 (1963).

8. Seidensticker, R. G., and D. R. Hamilton, "Growth Mechanisms in Germanium Dendrites: Three Twin Dendrites: Experiments On and Models for the Entire Interface," J. Appl. Phys. 34 (10) 3113-3119 (1963).

9. Smith, R. G., "A Study of Growth Processes in Germanium Dendrites Using Pulse Electroplating Techniques," J. Electrochemical Soc. 108 (3) 238 (1961).

10. O'Hara, S., "Dislocations in Webs of Germanium and Silicon," J. Appl. Phys. 35 (2) 409-413 (1964).

11. O'Hara, S., and A. I. Bennett, Jr., "Web Growth of Semiconductors," J. Appl. Phys. 35 (3, Part 1) 686-693 (1964).

12. Dermatis, S. N., and J. W. Faust, Jr., "Semiconductor Sheets for the Manufacture of Semiconductor Devices," IEEE Trans. Commun. Electron. 65, 195-200 (1963).

13. Barrett, D. L., E. H. Myers, D. R. Hamilton, and A. I. Bennett, Jr., "Growth of Wide, Flat Crystals of Silicon Web," J. Electrochem. Soc. 118 (6) 953-957 (1971).

14. Stepanov, A. V., "On Growing Crystals of Predetermined Shape," Iz. AN SSSR 33 (12) 1775 (1969); Translation in: Bul. Acad. Sci. USSR 33 (12) 1775 (1969).

15. Ivantsov, G. P., "The Temperature Distribution Around a Hemisphere, a Cylinder and a Needle Shaped Crystal Growing in a Supercooled Melt," Dok. AN SSSR 58, 567 (1947).

16. Temkin, D. E., "Growth Rate of the Needle Shaped Crystal Formed in a Supercooled Melt," Dok. AN SSSR 132 (6) 1307-1310 (1960). Translation in: Proc. Acad. Sci. USSR 132 (6) 1307-1310 (1960).

17. Horvay, G., and J. W. Cahn, "Dendritic and Spheroidal Growth," Acta Met. 9, 695-705 (1961).

18. "Thermophysical Properties of High Temperature Solid Materials," Y. S. Touloukian, Ed., Vol. 1, p. 878, MacMillan, New York (1967). Referenced to M. Olette, Comp. Rend. 244, 1033-6 (1957).