Thermal stress reduction for a Czochralski grown single crystal

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Abstract. In this paper an optimal control approach for thermal stress reduction inside a Czochralski grown single crystal is presented. Using the lateral heat flux as a control variable, an optimal control formulation for minimizing thermal stress with a given crystal shape is derived. Since thermal stress is also affected by the lateral shape of crystals during growth, the level of the stress can be reduced by growing crystals into a suitable shape. Using the lateral shape as a control variable, a similar optimal control formulation for stress reduction is derived. In both cases, von Mises stress is used as an objective function for the constrained optimization problem. Euler-Lagrange equations are derived using calculus of variations and Lagrange multipliers. Various stress reduction strategies are explored by solving the Euler-Lagrange equations numerically.

Key words: calculus of variations, crystal, Czochralski technique, optimal control, von Mises stress

1 Introduction

Czochralski (Cz) technique is one of the most common methods for growing single semiconductor crystals. The quality of Cz grown crystals is affected greatly by crystalline defects formed during the growth process. It is well-known that defect density is directly related to the thermal stress caused by temperature variation inside the crystal [1, 2]. Therefore, it is important to find a systematical way to control the temperature variation during growth.

Several researchers have utilized optimal control approaches to find favorable growth conditions with properly chosen objective functions for growing cylindrical silicon crystals with constant radii. For example, Bornside *et al.* [1] used the von Mises stress as a measure of thermal stress to find optimum growth conditions and system configurations for dislocation-free Cz grown silicon crystals. They applied an integrated numerical analysis model to search for optimal growth conditions. Using carefully selected target temperature distribution, Müller [3] showed that the optimum growth conditions for the silicon Czochralski process can be found by optimizing the geometry of hot zone heat shields and cooling devices. Jeong *et al.* [4] obtained optimal conditions by using crystal surface temperature distribution as an objective function. All of the above studies assume cylindrical crystals and none has discussed the effect of crystal lateral shape on thermal stress distribution.

By comparison, much less attention has been paid to compound crystal growth where controlling the appearance of crystalline defects is more difficult. Care must be taken to control the lateral shape of the crystal as well as the thermal environment. It has been shown that the lateral shape needs to be controlled carefully to avoid the appearance of excessive defects [5, 6]. This is mainly due to their low resistance to resolved shear stress which is responsible for causing crystalline defects [7, 6]. In practice, "magic shapes" are obtained by trial-and-error, based on experience of the grower [7]. The main objective of this paper is to investigate stress reduction strategies using a more systematical approach. We will set up an optimal control approach which searches for favorable conditions automatically, by exploring the inter-play between thermal environment (lateral heat flux) and crystal shape.

Following [5], we derive an explicit formula for von Mises stress and use it as a primary measure to set up a constrained optimization problem under the framework of optimal control. In the first approach, we use lateral heat flux as a control variable while the shape of the crystal is fixed, normally a cone. In the second approach, crystal radius is used as a control variable while the lateral heat flux is assumed to be given. This two-step approach is not a mathematical necessity since a combined approach with two control variables (heat flux and radius) can be attempted and the mathematical setup is almost identical. It is adopted from a practical point of view since a complete control of the lateral heat flux may not be achievable. A more common strategy is to partially control the lateral heat flux, e.g. using a heat shields, while adjusting the withdrawal rate and heater power supply so that the crystal grows into a desirable shape.

While a full numerical approach using stress as a objective function [1] provides an accurate control over the stress level, it is normally computationally intensive due to the iterative nature of search algorithms during minimization. Furthermore, the results tend to be problem specific which may not be readily applicable to other growth processes with different setups. Using temperature as a control target is computationally more efficient as demonstrated in [8, 4, 9, 10, 3]. However, the control of the thermal stress is not directly imposed unless a priori knowledge of the growth process is available. In general, the relationship between the temperature and stress is not readily available for a complex growth process such as Cz growth.

In this paper, we use an alternative approach by utilizing mathematical models to predict the temperature-stress relationship, under proper simplifications. In [5], a semi-analytical thermal stress model is obtained using a perturbation approach with the Biot number (the non-dimensional heat flux through the lateral crystal surface) as a small parameter, similar to previous work [11, 12, 13, 14, 15]. In this study, we extend the model in [5] to allow for a variable heat flux between the crystal lateral surface and the ambient gas in the growth chamber. In principle, the lateral heat flux to the ambient gas can only be determined by solving the coupled heat transfer problem involving the crystal, melt, gas, the configuration and heat supply of the grower. However, it can be altered or controlled using devices such as heat shields [3]. Therefore, it is reasonable to treat the lateral heat flux as a control variable in the optimization process. The shape of a Cz grown crystal is determined by the motion of the triple phase point, which can be controlled by the heater power supply and extracting rate. We can use it as another control variable. In practice, the dynamics of the triple point may be important due to stability of its radial motion [16, Chapter 2] . However, this effect can be minimized by using a feedback control device [8].

To keep our problem manageable, we have decoupled the melt as in [5]. We assume that the heat flux from the melt is uniform in the radial direction, based on the observation that the crystal-melt interface is almost flat for Cz grown InSb crystals [7]. In reality, the heat flux at the interface is influenced by the melt flow inside the crucible and is controlled indirectly by the heater. Additional control may be realized by using electromagnetic fields to alter the melt flow pattern and its stability [17].

The rest of the paper is organized as follows. In Section 2, we present a semi-analytical model for the (von Mises) thermal stress and the setup of the optimal control problems. In Section 3, we

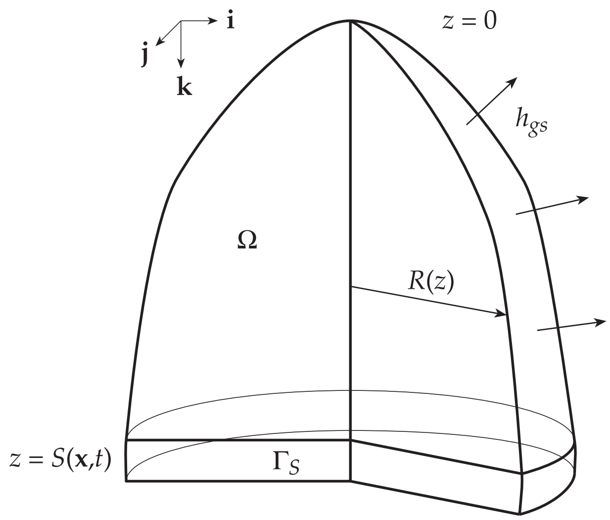


Figure 1: A typical crystal at some time t during a growth run with a newly solidified portion at $z = S(\mathbf{x}, t)$ [5].

introduce a variational formulation and the Euler-Lagrange equations are derived using calculus of variations, Lagrange multipliers and penalty functions. Numerical results and discussions are presented in Section 4 and we finish the paper with a short conclusion in Section 5.

2 Problem description

The problem setup and derivations of asymptotic solution for temperature and thermal stress are similar to those in [5]. For completeness, however, we will briefly describe the model and present the temperature and stress solutions before setting up our optimal control problems. For detailed description of the model and perturbation solution of the temperature, we refer interested readers to [5] and references therein.

2.1 A semi-analytical model

Following [5], we assume that the crystal is axis-symmetric and the coordinate system is fixed to the top of the growing crystal at $z = 0$, the final length of the crystal is denoted L and the crystal radius is denoted $R(z)$. The growth starts with a seed crystal with radius of order $R_0 = 5 \times 10^{-3}$ m and length $Z_0 = 3 \times 10^{-2}$ m. Figure 1 illustrates the geometry of a typical crystal. Within the crystal Ω, the temperature $T(\mathbf{x}, t)$ satisfies the heat equation

$$\rho_s c_s \frac{\partial T}{\partial t} = k_s \nabla^2 T, \qquad \mathbf{x} \in \Omega,\ t > 0 \tag{1}$$

where ρ_s, c_s and k_s are the density, specific heat and thermal conductivity of the crystal (solid phase), respectively. The lateral surface of the crystal is denoted by Γ_g. Temperature boundary condition at the lateral surface is given as

$$-k_s \frac{\partial T}{\partial \mathbf{n}} = h_{gs}(T - T_g) + r_c(T^4 - T_g^4), \qquad \mathbf{x} \in \Gamma_g. \tag{2}$$

where h_{gs} and r_c are the heat transfer at the lateral surface due to convective cooling by the gas and radiative heat loss, respectively and T_g the ambient gas temperature. Alternatively, we can model both convective and radiative effects through a simple Newtonian cooling law:

$$-k_s \frac{\partial T}{\partial \mathbf{n}} = h_{gs}(T - T_g), \qquad \mathbf{x} \in \Gamma_g. \tag{3}$$

Here we assume that the heat transfer coefficient, h_{gs}, incorporates both convective and radiative heat transfer (via linearization). At the top of the crystal we invoke a Newtonian cooling law

$$k_s \frac{\partial T}{\partial z} = h_{ch}(T - T_{ch}), \qquad z = 0, \tag{4}$$

in the case that the radius at $z = 0$ is assumed to be non-zero. Here h_{ch} represents the heat transfer coefficient for the seed-chuck connection and T_{ch} is the chuck temperature.

The crystal-melt interface is denoted by Γ_S where $T = T_m$ (melting temperature). The solidus isotherm is thus implicitly defined by the temperature field. Explicitly we denote the solidus isotherm by

$$z - S(\mathbf{x}, t) = 0, \qquad \mathbf{x} \in \Gamma_S. \tag{5}$$

The motion of the solidus isotherm is governed by the Stefan condition

$$\rho_s L_h \, |\vec{v}_n| = k_s \frac{\partial T}{\partial \mathbf{n}}\bigg|_{z \to S^-} - q_{l,n} \tag{6}$$

where $|\vec{v}_n|$ is the speed at which the interface moves in the direction of the outward unit normal $\mathbf{n}$, L_h is the latent heat and $q_{l,n}$ is the heat flux from the melt normal to the interface.

The crystal radius is determined by the motion of the melt-solid-gas triple point, which is given by

$$\frac{\partial R}{\partial t}\bigg|_{z=S} = \tan(\theta - \theta_c) \frac{\partial S}{\partial t}\bigg|_{r=R} \tag{7}$$

where θ_c is the contact angle formed by the wetting fluid (melt) and the crystal and θ is the angle formed by the meniscus with the vertical z-axis.

For the size of crystals under consideration here, the effect of the surface tension in the azimuthal direction can be neglected. Furthermore, if we neglect the dynamic effect of the melt flow, the shape of the meniscus is determined by the surface tension through the Laplace-Young equation and the capillary height ζ_0 at the triple junction is approximately [18]

$$\zeta_0 = \sqrt{\alpha \, (1 - \sin \theta)} \tag{8}$$

where $\alpha = 2\sigma/\rho_l g$, σ the surface tension coefficient, ρ_l the density of melt, and g the gravitational acceleration. Figure 2 shows the schematic diagram of the meniscus. From mass conservation, change of the capillary height ζ_0 at the triple junction is

$$\frac{d\zeta_0}{dt} = v_p + v_m - \frac{\partial S}{\partial t} \tag{9}$$

where v_p is the pulling rate and v_m is the rate at which the melt/gas surface drops, which is given by

$$v_m = \frac{\rho_s R^2}{\rho_l R_c^2} \frac{\partial S}{\partial t}$$

where R_c is the radius of the crucible. Thus the meniscus shape is determined by its height, which is controlled by the heat fluxes and crystal extracting (pulling) rate.

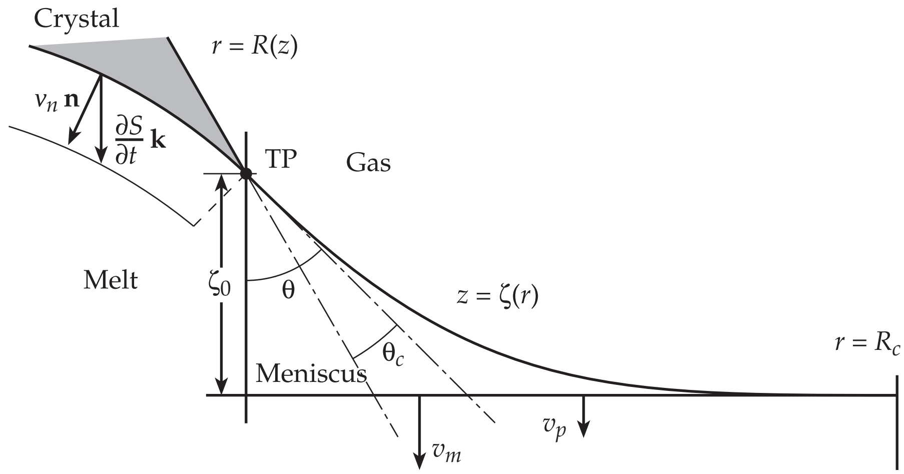


Figure 2: Schematic diagram of the meniscus $z = \zeta(r)$ with capillary height ζ_0 [5].

2.1.1 Asymptotic solution

Define the Biot number by

$$\epsilon = \frac{\bar{h}_{gs}\overline{R}}{k_s}$$

where $\bar{h}_{gs}$ is the mean heat transfer coefficient defined by

$$\bar{h}_{gs} = \frac{1}{L}\int_0^L h_{gs}(z)\,dz,$$

where L is the final length of the crystal.

In the same spirit as in [5], we now non-dimensionalize the equation and boundary conditions using the following scalings

$$r = \overline{R}\hat{r}, \quad \epsilon^{1/2}z = \overline{R}\hat{z}, \quad R(z) = \overline{R}\hat{R}(\hat{z}), \quad \epsilon^{1/2}S(r,t) = \overline{R}\hat{S}(\hat{r},\hat{t}),$$
$$T = T_g + \Delta T\Theta, \quad \Delta T = T_m - T_g, \quad t = \frac{\mathrm{S_T}\overline{R}^2\rho_s c_s}{k_s\epsilon}\hat{t}, \quad \mathrm{S_T} = \frac{L_h}{c_s\Delta T}.$$

Here variables with hats (ˆ) are the non-dimensional ones. In the following, the hats will be dropped for brevity. The nondimensional equation and boundary conditions are

$$\frac{\epsilon}{\mathrm{S_T}}\Theta_t = \frac{1}{r}(r\Theta_r)_r + \epsilon\Theta_{zz}, \qquad \mathbf{x}\in\Omega,\ t>0 \tag{10}$$

$$\begin{aligned} -\Theta_r + \epsilon\Theta_z R'(z) = \epsilon\left[1+\epsilon(R'(z))^2\right]^{1/2}(\beta\Theta + f(\Theta)), &\qquad \mathbf{x}\in\Gamma_g,\\ \Theta = 1, &\qquad \mathbf{x}\in\Gamma_S,\\ \Theta_z = \delta_0(\Theta-\Theta_{ch}), &\qquad z=0, \end{aligned} \tag{11}$$

where $\beta(z) = h_{gs}(z)/\bar{h}_{gs}$, $f(\Theta) = r_c/\bar{h}_{gs}\left(\Delta T^3\Theta^4 + 4T_g\Delta T^2\Theta^3 + 6T_g^2\Delta T\Theta^2 + 4T_g^3\Theta\right)$ and $\delta_0 = \epsilon^{1/2}h_{ch}/\bar{h}_{gs}$. The solidus advances according to the Stefan condition (6) which in non-dimensional coordinates becomes

$$\Theta_z - \frac{1}{\epsilon} S_r \Theta_r = \left(1 + \frac{S_r^2}{\epsilon}\right)^{1/2} (\gamma + S_t), \qquad \gamma = \frac{q_l \overline{R}}{\epsilon^{1/2} k_s \Delta T} \tag{12}$$

where q_l and γ are the dimensional and dimensionless heat fluxes in the liquid across the crystal/melt interface in the axial direction. As discussed in [5], we assume that the solidification is driven by the heat loss from the crystal lateral surface and the rate of solidification defines the relevant time scale. Equation (12) simply states that the motion of the interface is determined by the heat conduction in the solid as well as in the melt. The heat conduction in the crystal is an order one quantity, after the rescaling. Therefore, the rescaled heat flux from the melt γ must be of order one at most.

To find asymptotic solution for the temperature, we expand both Θ and S in terms of ϵ as in [5]. Equations (10) and (11) suggest that the temperature Θ is independent of r to leading order. Consequently, the crystal/melt interface S is also independent of r to leading order. Based on these observations, we expand both Θ and S as follows:

$$\begin{aligned} \Theta &\sim \Theta_0(z,t) + \epsilon\Theta_1(r,z,t) + \epsilon^2\Theta_2(r,z,t) + \ldots \\ S &\sim S_0(t) + \epsilon S_1(r,t) + \epsilon^2 S_2(r,t) + \ldots \ . \end{aligned} \tag{13}$$

If we substitute the expansion into the scaled model (10) and (11), collect the terms at the zeroth and first order of ϵ, and apply the solvability conditions, we can derive the equations for the leading and higher order solutions. The detailed derivations can be found in [5, 19]. Similar and more detailed asymptotic analysis for cylindrical crystals can be found in [11, 12, 15].

The zeroth order temperature solution satisfies

$$\frac{1}{\mathrm{S_T}} \Theta_{0,t} = \Theta_{0,zz} + \frac{2}{R}\left[R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right], \qquad 0 < z < S_0(t),\ t > 0, \tag{14a}$$

$$\Theta_{0,z} = \delta_0(\Theta_0 - \Theta_{ch}), \qquad z = 0, \tag{14b}$$

$$\Theta_0 = 1, \qquad z = S_0(t) \tag{14c}$$

with an initial condition $\Theta_0(z, 0) = g(z) \leq 1$ compatible with the boundary conditions.

The advance of $S_0(t)$ is coupled to the thermal gradients via

$$\gamma + S_{0,t} = \Theta_{0,z}\big|_{z=S_0(t)}, \tag{15}$$

where $S_0(0) = Z_0$.

Since there is a one-to-one relationship between growth time t and the size of the crystal (given by S_0), we use S_0 as the main variable, instead of t. In addition, as the lateral size of the crystal is determined by the motion of the triple junction, which can be controlled by the pulling rate and thermal flux from the melt, we will work with the crystal radius directly. In some of the computations reported in this study, we have used the nondimensional version of equation (7) for the radial motion of the triple junction where the value of θ is given by (8).

2.1.2 Thermal stress

Based on the plane strain assumption, thermal stress can be obtained as

$$\sigma_{rr} = \frac{1}{4}\overline{\sigma}\left(R(z)^2 - r^2\right), \quad \sigma_{\theta\theta} = \frac{1}{4}\overline{\sigma}\left(R(z)^2 - 3r^2\right), \quad \sigma_{zz} = \frac{1}{2}\overline{\sigma}\left(R(z)^2 - 2r^2\right) \tag{16}$$

where $\overline{\sigma} = \epsilon\Theta_1^1(z,t)$. The von Mises stress is

$$\begin{aligned}\sigma_{\mathrm{VM}} &= \frac{1}{\sqrt{2}}\left[(\sigma_{rr}-\sigma_{\theta\theta})^2+(\sigma_{rr}-\sigma_{zz})^2+(\sigma_{\theta\theta}-\sigma_{zz})^2\right]^{1/2} \\ &= \frac{1}{4}\epsilon\left|\Theta_1^1(z,t)\right| R(z)^2\left[1-4\left(\frac{r}{R(z)}\right)^2+7\left(\frac{r}{R(z)}\right)^4\right]^{1/2}. \qquad (17)\end{aligned}$$

In the pseudo-steady case $\Theta_{0,zz} = -4\Theta_1^1$ so that equation (17) becomes

$$\begin{aligned}\sigma_{\mathrm{VM}} &= \frac{1}{16}\epsilon\left|\Theta_{0,zz}(z)\right| R(z)^2\left[1-4\left(\frac{r}{R(z)}\right)^2+7\left(\frac{r}{R(z)}\right)^4\right]^{1/2} \\ &= \frac{1}{8}\epsilon\left|\left(R'(z)\Theta_{0,z}(z)-\beta(z)\Theta_0(z)-f(\Theta_0)\right)\right| R(z) \\ &\quad \left[1-4\left(\frac{r}{R(z)}\right)^2+7\left(\frac{r}{R(z)}\right)^4\right]^{1/2}. \qquad (18)\end{aligned}$$

Remark. If we use (3) as the temperature boundary condition, we can obtain asymptotic solution and thermal stress by setting $f(\Theta_0) = 0$ in the above equations.

2.2 Stress minimization

We now discuss the main objective of this paper. We will use the analytical formula for the von Mises stress (18) and set up our optimization problems. Since the von Mises stress depends on heat transfer coefficient h_{gs} (or more precisely $\beta(z)$) as well as crystal shape $R(z)$, we will discuss two problems. In the first approach we use $\beta(z)$ as a control variable while keeping the crystal shape $R(z)$ fixed. In the second problem we use $R(z)$ as a control variable with $\beta(z)$ fixed.

2.2.1 Problem I: Optimal $\beta(z)$

The original mathematical statement of the problem at hand is

$$\min_{\beta(z)} \max_{Z_0\le S_0\le L, 0\le z\le S_0, 0\le r\le R(z)} \sigma_{\mathrm{VM}}\left(r,z;R,S_0,\beta\right)$$

subject to constraints, where Z_0 and L are the length of the seed and the final length of the crystal, respectively. However this set up is not easy to handle numerically. We seek an approximation for this problem as follows

$$\min_{\beta(z)}\left[\int_0^L \sigma_{\mathrm{VM}}^2(z)dz+\omega_1\int_0^L\left(\beta(z)-\beta_0\right)^2-\omega_2\int_0^L\Theta_{0,z}^2(z)dzdz\right]$$

subject to

$$\Theta_{0,zz} + \frac{2}{R}\left[R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right] = 0, \qquad 0 < z < L, \tag{19a}$$

$$\Theta_{0,z} = \delta_0(\Theta_0 - \Theta_{ch}), \qquad z = 0, \tag{19b}$$

$$\Theta_0 = 1, \qquad z = L, \tag{19c}$$

$$\int_0^L \beta(z)dz = \beta_0 L, \tag{19d}$$

$$\beta(z) \geq 0, \qquad 0 \leq z \leq L \tag{19e}$$

where $0 \leq \omega_k \leq 1$ are weighting parameters and β_0 a given parameter. The first term of the objective functional is the L^2 version of the original functional. The second term is a penalty term added to avoid drastic variation in β, which could be impractical to implement. The last term is also term which penalizes slow growth, which may also be undesirable from practical point of view. The integral constraint on β is for comparison purpose since the stress reduction due to smaller mean heat flux should be considered as a real benefit. Obviously the heat transfer coefficient must remain positive from the physics point of view. Here we have used the pseudo-steady approximation. In this case the von Mises stress becomes

$$\sigma_{\text{VM}} = \frac{1}{8}\epsilon\left|R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0)\right| R(z)\left[1 - 4\left(\frac{r}{R(z)}\right)^2 + 7\left(\frac{r}{R(z)}\right)^4\right]^{1/2}$$

which reaches its maximum when $r = R$, i.e.,

$$\sigma_{\text{VM}} \leq \frac{1}{4}\epsilon\left|R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0)\right| R(z).$$

Thus our optimization problem can be changed into

$$\min_{\beta(z)}\left[\int_0^L \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0)\right) R(z)\right]^2 dz + \omega_1\int_0^L (\beta(z)-\beta_0)^2\, dz - \omega_2\int_0^L \Theta_{0,z}^2(z)dz\right] \tag{20}$$

with the constraints.

2.2.2 Problem II: Shape optimization

Similarly we seek an approximation and set up the problem as follows

$$\min_{R(z)}\left[\int_0^L \sigma_{\text{VM}}^2(z)dz + \omega_1\int_0^L \left(R'(z) - R'_0\right)^2 dz - \omega_2\int_0^L \Theta_{0,z}^2(z)dz\right],$$

or

$$\min_{R(z)}\left[\int_0^L \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right) R(z)\right]^2 dz + \omega_1\int_0^L \left(R'(z) - R'_0\right)^2 dz - \omega_2\int_0^L \Theta_{0,z}^2(z)dz\right], \tag{21}$$

subject to

$$R\Theta_{0,zz} + 2\left(R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right) = 0, \qquad 0 < z < L, \tag{22a}$$

$$\Theta_{0,z} = \delta_0(\Theta_0 - \Theta_{ch}), \qquad z = 0, \tag{22b}$$

$$\Theta_0 = 1, \qquad z = L, \tag{22c}$$

$$\int_0^L R^2(z)dz = L, \tag{22d}$$

$$R = R_0, \qquad z = 0, \tag{22e}$$

$$R(z) \geq R_0, \qquad 0 \leq z \leq L \tag{22f}$$

where R_0 and R_0' are given parameters and ω_k are penalty parameters. The first and last term in the objective functional are the same as before. The second term is added to penalize deviation from the cone shape. From practical point of view, drastic variation of the crystal shape should be avoided since it may reduce the usable amount of the material. In addition, there exists a critical growth angle for some compound crystals beyond which twinning may happen during growth [20].

3 Derivation of the Euler-Lagrange equations

We now discuss the Euler-Lagrange equations for the two optimization problems using calculus of variation.

3.1 Problem I

Since the constraints include both equality constraints (19a), (19d) and inequality (19e), we use the method of Lagrange multipliers and the penalty function method. The augment Lagrangian objective function is defined by

$$\begin{aligned} J_1 \;=\; & \int_0^L \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right) R(z)\right]^2 dz \\ & + \int_0^L \lambda(z) \left[\Theta_{0,zz}(z) + \frac{2}{R(z)} \left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)\right] dz \\ & + \omega_1 \int_0^L (\beta(z) - \beta_0)^2 dz - \omega_2 \int_0^L \Theta_{0,z}^2(z) dz \\ & + \mu\left(\beta_0 L - \int_0^L \beta(z)dz\right) + \frac{\rho}{2}\int_0^L \beta^2(z) H(-\beta(z))dz \end{aligned} \tag{23}$$

where $\lambda(z)$ and μ are Lagrange multipliers, ρ is penalty parameter which is a sufficiently large positive number, and $H(.)$ is Heaviside function.

Using the calculus of variation, we can derive the necessary conditions based on first order variations. For the optimization problem given by (23) with constraints (19a)-(19e), we obtain the following Euler-Lagrange equations (see Appendix A for the detailed derivation)

$$\Theta_{0,zz} + \frac{2}{R}\left[R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right] = 0, \tag{24a}$$

$$\int_0^L \beta(z)dz = \beta_0 L, \tag{24b}$$

$$2R^2\left(R'\Theta_{0,z} - \beta\Theta_0\right)\Theta_0 + 2\lambda\Theta_0/R - 2\omega_1(\beta - \beta_0) + \mu - \rho\beta H(-\beta) = 0, \tag{24c}$$

$$\begin{aligned}\lambda_{zz} - \left(\frac{2R'\lambda}{R}\right)_z - 2\lambda\beta/R - 2\beta R^2\left(R'\Theta_{0,z} - \beta\Theta_0\right)& \\ -2\left(R^2R'\left(R'\Theta_{0,z} - \beta\Theta_0\right)\right)_z - 2R^2\left(R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right)f'(\Theta_0)& \\ +2R^2\beta f(\Theta_0) - 2\left(2RR'^2 f(\Theta_0) + R^2R'' f(\Theta_0) + R^2R' f'(\Theta_0)\Theta_{0,z}\right)& \\ -2\lambda f'(\Theta_0)/R + 2\omega_2\Theta_{0,zz} &= 0.\end{aligned} \tag{24d}$$

The boundary conditions for the above equations are

$$\Theta_{0,z} = \delta_0(\Theta_0 - \Theta_{ch}), \qquad z = 0, \tag{25a}$$

$$\Theta_0 = 1, \qquad z = L, \tag{25b}$$

$$\lambda_z - \left(\delta_0 + \frac{2R'}{R}\right)\lambda - 2R^2R'\left[\left(R'\delta_0 - \beta\right)\Theta_0 - R'\delta_0\Theta_{ch}\right] - 2R^2R' f(\Theta_0) = 0, \qquad z = 0, \tag{25c}$$

$$\lambda = 0, \qquad z = L. \tag{25d}$$

This is a system of coupled nonlinear second order ordinary differential equations for temperature Θ and Lagrange multiplier λ. It has to be solved numerically in general.

3.2 Problem II

Similarly we use Lagrange multipliers to derive the Euler-Lagrange equations. The augment Lagrangian objective function is defined by

$$\begin{aligned} J_2 = & \int_0^L \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)R(z)\right]^2 dz \\ & + \omega_1 \int_0^L \left(R'(z) - R'_0\right)^2 dz - \omega_2 \int_0^L \Theta_{0,z}^2(z)dz \\ & + \int_0^L \lambda(z)\left[R(z)\Theta_{0,zz}(z) + 2\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)\right]dz \\ & + \mu\left[L - \int_0^L R^2(z)dz\right] + \frac{\rho}{2}\int_0^L \left(R(z) - R_0\right)^2 H(R_0 - R(z))dz \end{aligned} \tag{26}$$

where $\lambda(z)$ and μ are Lagrange multipliers, $\rho > 0$ is penalty parameter and $H(.)$ is Heaviside function.

Using calculus of variation, we can derive necessary conditions for the optimization problem given by equation (26) with constraints (22a)-(22f). We obtain the following Euler-Lagrange equations (details in Appendix B)

$$R\Theta_{0,zz} + 2\left(R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right) = 0 \quad (27a)$$

$$\int_0^L R^2(z)dz = L \quad (27b)$$

$$\begin{aligned} 6RR'^2\Theta_{0,z}^2 - 12\beta\Theta_0 RR'\Theta_{0,z} + 6\beta^2 R\Theta_0^2 - 2R^2R''\Theta_{0,z}^2 + 2\beta' R^2\Theta_0\Theta_{0,z} - 2\omega_1 R'' \\ +2\beta R^2\Theta_{0,z}^2 - \lambda\Theta_{0,zz} - 2\lambda'\Theta_{0,z} - 2\mu R + \rho\left(R - R_0\right)H(R_0 - R) \\ -4RR'\Theta_{0,z}f(\Theta_0) + 6R^2f^2(\Theta_0) + 8R\beta\Theta_0 f(\Theta_0) + 2R^2f'(\Theta_0)\Theta_{0,z}^2 &= 0 \quad (27c) \\ 2\beta^2R^2\Theta_0 + 2R\left(\beta R\Theta_0 - 2RR'\Theta_{0,z}\right)R'' + 2\beta' R^2R'\Theta_{0,z} + R\lambda'' - \lambda R'' - 2\beta\lambda \\ +2R^2\beta\Theta_0 f'(\Theta_0) + 2R^2f(\Theta_0)f'(\Theta_0) + 4RR'2f(\Theta_0) + 2R^2R''f(\Theta_0) \\ +2R^2\beta f(\Theta_0) - 2\lambda f'(\Theta_0) + 2\omega_2\Theta_{0,zz} &= 0. \quad (27d) \end{aligned}$$

The boundary conditions are

$$\Theta_{0,z} = \delta_0(\Theta_0 - \Theta_{ch}), \qquad z = 0, \qquad (28a)$$

$$\Theta_0 = 1, \qquad z = L, \qquad (28b)$$

$$\left(\frac{\lambda}{R}\right)' + 2R'\left[\beta\Theta_0 - \delta_0 R'\left(\Theta_0 - \Theta_{ch}\right)\right] + 2\omega\delta_0\left(\Theta_0 - \Theta_{ch}\right) + 2R^2R'f(\Theta_0) = 0, \qquad z = 0, \qquad (28c)$$

$$\lambda = 0, \qquad z = L, \qquad (28d)$$

$$R = R_0, \qquad z = 0, \qquad (28e)$$

$$2R^2\left(R_z\Theta_{0,z} - \beta\right)\Theta_{0,z} + 2\lambda\Theta_{0,z} + 2\omega_1\left(R' - R_0'\right) - R^2 f(\Theta_0)\Theta_{0,z} = 0, \qquad z = L. \qquad (28f)$$

3.3 Numerical implementation

The system of equations (24a)-(24d) with boundary conditions (25a)-(25d) and equations (27a)-(27d) with boundary conditions (28a)-(28f) are nonlinear and coupled. In order to solve them, we use finite difference method to discrete differential equations and use Trapezoidal rule for the integrals. The discrete system is also nonlinear system and is solved using MATLAB. MATLAB used Gauss-Newton method and trust-region dogleg method to solve the nonlinear systems of equations. Gauss-Newton method uses a nonlinear least-squares solver which employs a line search procedure and a quasi-Newton method to solve the equations. Newton's method is used in trust-region dogleg method to find the search direction.

4 Results

Two sets of computations are carried out, one corresponding to temperature boundary condition (3) and the other with temperature boundary condition (2). Using the optimality system of equations (24a)-(24d) with boundary conditions (25a)-(25d), we can find optimal heat transfer coefficient h_{gs} for given crystal shapes. The optimal crystal shape is obtained using equations (27a)-(27d) with boundary conditions (28a)-(28f). The zero order temperature and von Mises stress are computed using equation (14a)-(15) and (18). A typical set of parameters used in our computations is listed in Table 1. For all computations, we assume that the top of the crystal is insulated from the chuck ($\delta_0 = 0$) and the mean heat transfer coefficient is $\overline{h}_{gs} = 4$ W/m^{2}K.

Table 1: A summary of parameters for InSb growth.

Name	Symbol	Value
Ambient gas temperature	T_g	600 K
Melting temperature	T_m	798.4 K
Solid density	ρ_s	5.64×10^3 kg/m^3
Liquid density	ρ_l	6.47×10^3 kg/m^3
Thermal conductivity	k_s	4.57 W/m K
Heat capacity	$\rho_s c_s$	1.5×10^6 J/m^3 K
Surface tension coefficient	σ	0.434 J/m^2
Latent heat of fusion	L_h	2.3×10^5 J/kg
Mean crystal radius	$\bar{R}$	0.03 m
Crucible radius	R_c	0.1 m
Equilibrium growth angle	θ_c	25°

4.1 Combined convective and radiative heat transfers

We assume that a desirable crystal shape can be produced by adjusting the pulling rate and the non-dimensional heat flux of melt is $\gamma = -0.1$. The negative γ means that the melt region near the crystal/melt interface is a supercooling region. This γ can be got through controlling the melt flow. The assumption on a constant heat flux from the melt is based on the observation from the manufacturing process for InSb using the Cz method [7]. It has been observed that the bottom of the crystals is almost flat (except at the triple point) and the power supply stays stable (indicating a steady heat flux).

Before we present our main results, we will discuss briefly the case of cylindrical crystals in section 4.1.1. The main purpose is to show that our procedure produces consistent results compared to the ones reported in the literature for silicon [4]. As noted earlier, it is extremely difficult to grow InSb crystals in cylindrical shape [7] and it has been shown in our previous paper [5] that stress level is much higher in cylindrical crystals than that in conic ones. Therefore, we will focus our discussions on conic crystals in section 4.1.2 before discussing shape optimization in section 4.1.3.

4.1.1 Optimal β for cylindrical crystals

Even thought our main focus in this paper is on InSb crystals, our model is also applicable to other crystals. We start by applying our procedure to cylindrical silicon crystals so that a qualitative comparison can be made to the results obtained in [4]. Our computations are carried out by using the following parameter values. Density of the silicon crystal=2420 kg/m^3; heat capacities=1000 J /kg K; thermal conductivities k_s=22 W/m K; Poisson's ratio=0.25; crystal radius=0.1 m; melting temperaturre=1683 K; T_g=600 K; surface tension coefficient=0.72 J/m^2; latent heat of fusion=1.8$\times 10^6$ J/kg; specific heat c_s=1000 J/kg K; mean heat transfer coefficient h_{gs}= 8 W/m^3 K and length of crystal = 0.6 m.

Figure 3(a) shows the profiles for optimal and constant h_{gs}. Following parameters are used to find the optimal solution: $\rho = 60$; $\omega_1 = 0.2$; $\omega_2 = 0.3$; $\beta_0 = 1.0$. Figure 3(b) shows the nondimensional temperature along the lateral surface at the end of growth. The history of nondimensional maximum von Mises stress corresponding to the optimal and constant heat transfer coefficient h_{gs}

in Figure 3(c). Figure 3(d) shows the nondimensional von Mises stress along the lateral surface at the end of growth.

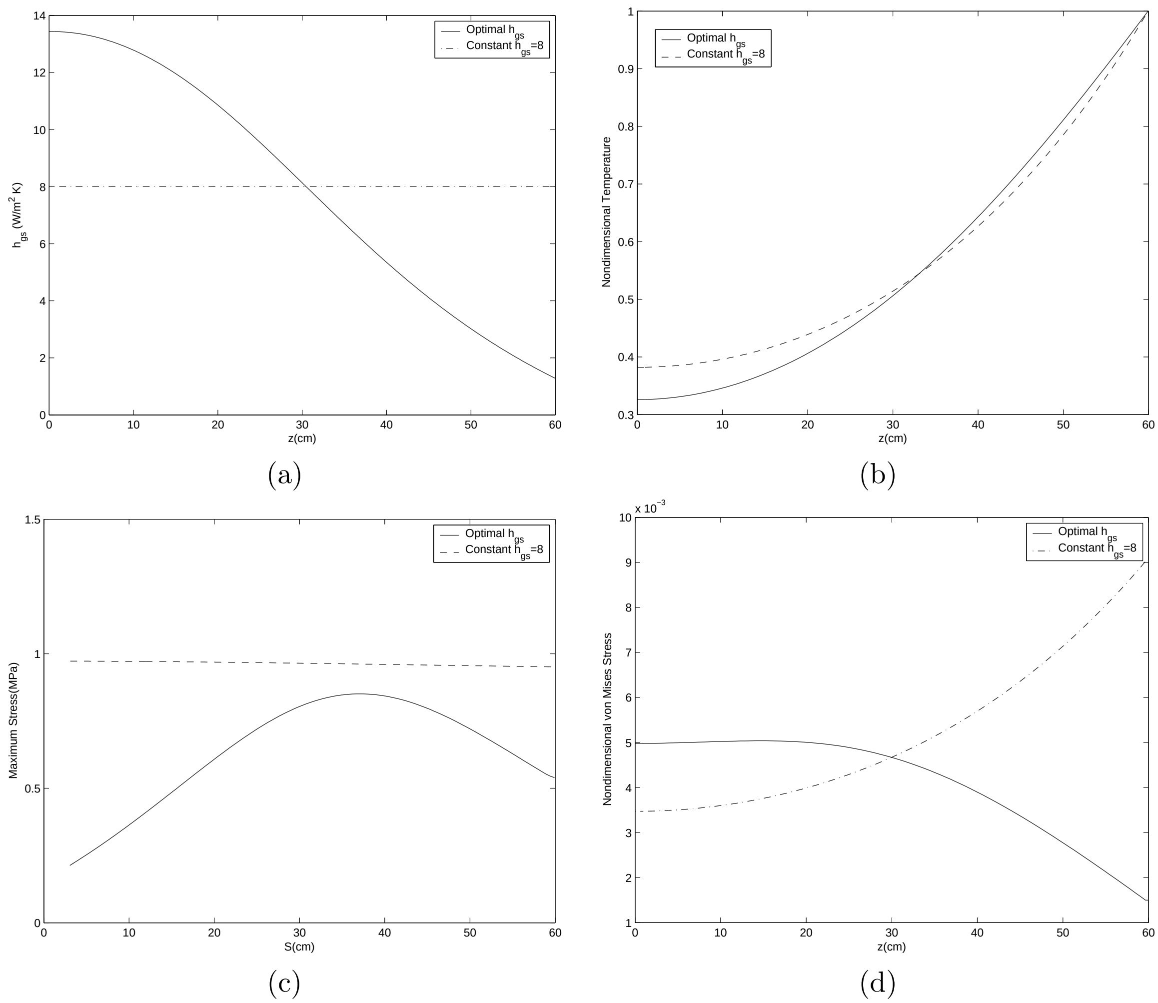


Figure 3: (a) The optimal and constant heat transfer coefficient h_{gs} for a silicon crystal. (b) The temperature along the lateral surface at the end of growth. (c) The history of nondimensional maximum von Mises stress corresponding to the optimal and constant heat transfer coefficient h_{gs}. (d) The nondimensional von Mises stress along the lateral surface at the end of growth.

The results show that the stress level can be reduced by almost 50%, when the heat transfer coefficient profile is optimized. The results also show that for the optimal case, lateral temperature variation is smaller near the crystal/melt interface, which is consistent with the solution in [4] when the surface temperature itself is used as a control variable in the optimization process.

For an InSb crystal we assume that the mean radius of the crystal is 0.03 m. Figure 4(a) shows the corresponding optimal heat transfer coefficient. The following parameters are used to find the optimal solution: $\rho = 60$; $\omega_1 = 0.2$; $\omega_2 = 0.3$; $\beta_0 = 1.0$. Figure 4(b) shows the nondimensional temperature along the lateral surface at the end of growth. The history of maximum von Mises stress corresponding to the optimal and constant heat transfer coefficient h_{gs} in Figure 4(c). Figure 4(d) shows the von Mises stress along the lateral surface at the end of growth. From Figure 4(c) it can be seen that the thermal stress can be greatly reduced, similar to the results for silicon crystals.

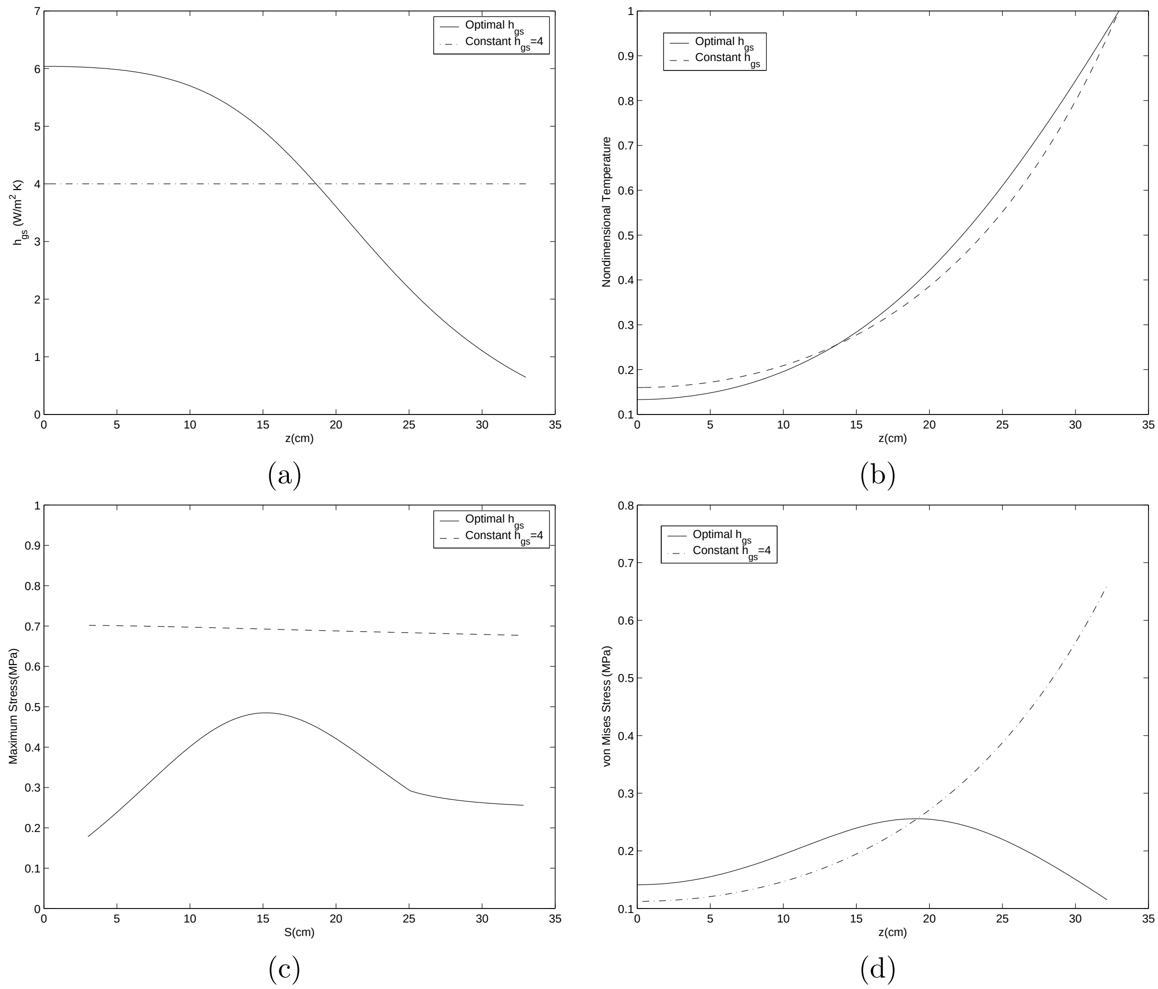


Figure 4: (a) The optimal and constant heat transfer coefficient h_{gs} for an InSb crystal. (b) The temperature along the lateral surface at the end of growth. (c) The history of maximum von Mises stress corresponding to the optimal and constant heat transfer coefficient h_{gs}. (d) The von Mises stress along the lateral surface at the end of growth.

4.1.2 Optimal β for conical crystals

The following results are for a conical crystal 33.5 cm long and 6 cm in diameter (largest) so that the mean radius is comparable to the cylindrical one in the previous section. We assume that $\beta_0 = 1$. To begin, we present some numerical results for three heat transfer coefficients: constant, optimal, and experimental heat transfer coefficient $h_{gs}(z)$. The experimental heat transfer coefficient was estimated and communicated to us by the engineers we have been working with [7]. The optimal heat transfer coefficient is calculated when $\omega_1 = 0$, $\omega_2 = 0.3$, and $\rho = 20$. In Figure 5 we plotted the shape of β (or h_{gs}) used for comparison: constant, one fitted from experimental data and the optimal function (obtained by solving the Euler-Lagrange equations derived in the previous section).

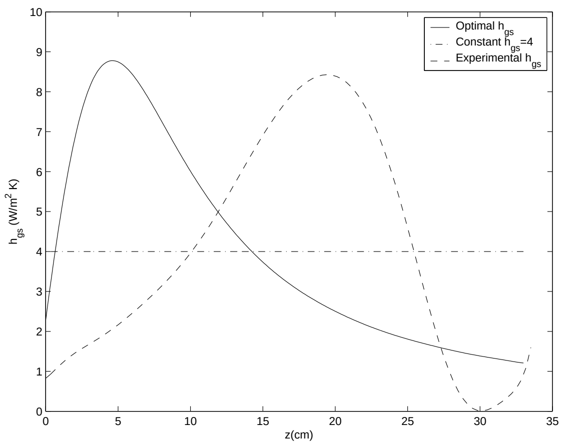
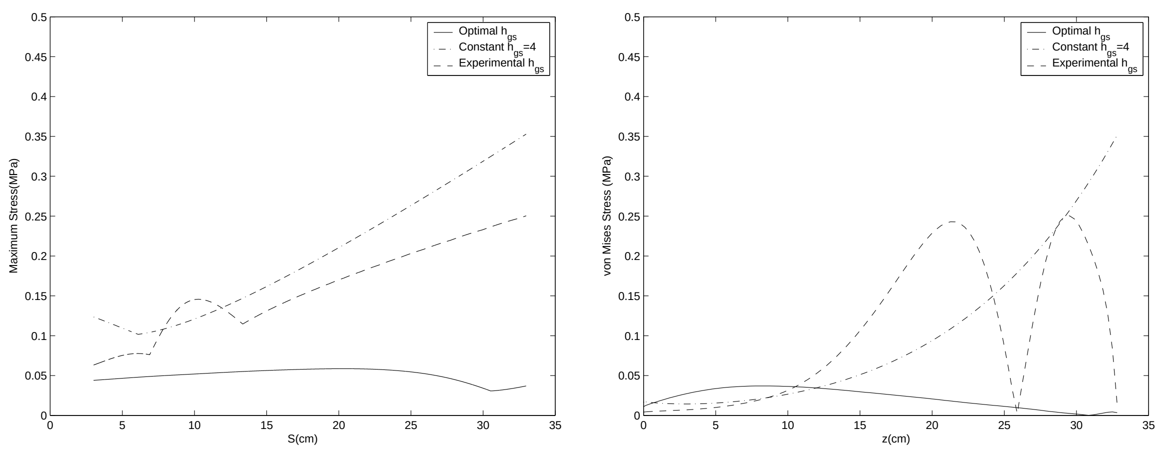


Figure 5: The three heat transfer coefficients.

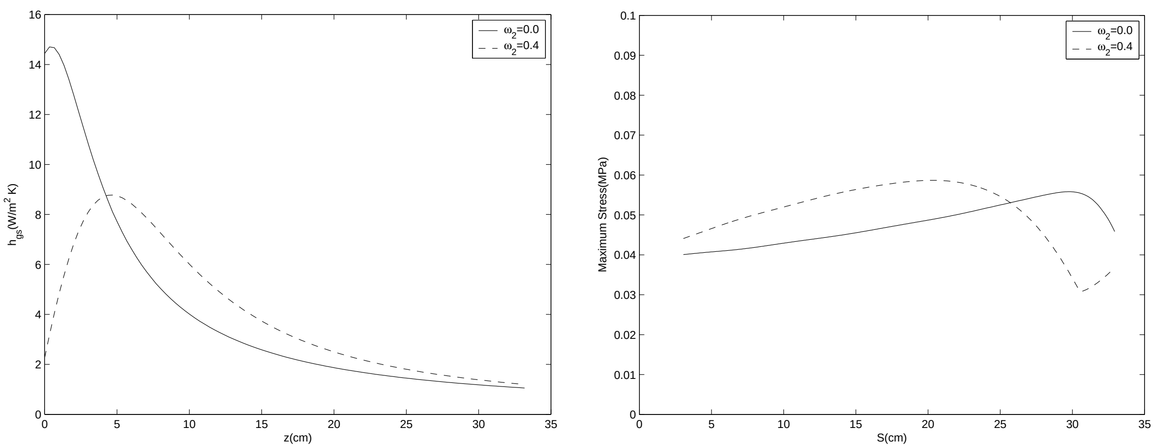
In Figure 6(a) the maximum von Mises stress during growth are plotted. The parameters for the optimization problem are $\rho = 20$, $\omega_1 = 0$ and $\omega_2 = 0.3$. The stress at the end of the growth is plotted in Figure 6(b). It can be seen that the reduction of the stress using the optimal h_{gs} is quite dramatic.



(a) (b)

Figure 6: (a) Maximum von Mises stress during the growth. (b) von Mises stress along the crystal lateral surface at the end of the growth. For the optimal heat transfer coefficient the following parameters are used: $\rho = 20$, $\omega_1 = 0$ and $\omega_2 = 0.3$.

Next we investigate the effect of the parameters in the optimization setup on the solutions. For the penalty parameter ρ, we find that the optimization problem converges to same solution when $\rho \geq 20$ for given ω_1 and ω_2 values. Figure 7(a) shows two optimal solutions of h_{gs} for $\omega_2 = 0$ and $\omega_2 = 0.4$ when $\omega_1 = 0$ and $\rho = 20$. Figure 7(b) shows the maximum von Mises stress during growth for these two cases. We found that the von Mises stress with $\omega_2 = 0$ is slightly lower than that with $\omega_2 = 0.4$ case. Since the penalty associated with ω_2 determines the growth speed, the gain on stress reduction is at the expense of a longer growth time. For example, the growth time for $\omega_2 = 0$ (33.47 hours) is longer than that for for $\omega_2 = 0.3$ case (30.68 hours).



(a) (b)

Figure 7: (a) Optimal heat transfer coefficients; (b) Maximum von Mises stress during the growth for parameters values are $\omega_2 = 0$ and $\omega_2 = 0.4$, respectively. The other parameter values are $\omega_1 = 0$ and $\rho = 20$.

In order to investigate the robustness of the optimal solution we present a set of calculations corresponding to a perturbation of the heat flux from the melt γ. We used the same optimal heat transfer coefficient shown in Figure 5 and computed the maximum von Mises stress σ_{max} corresponding to $\gamma = -0.1$, $\gamma = -0.09$ and $\gamma = -0.11$, denoted by $\sigma_{max}^{-0.09}$, $\sigma_{max}^{-0.1}$, $\sigma_{max}^{-0.11}$, respectively. We define the ratio of the fluctuations as

$$R_{dec} = \frac{\sigma_{max}^{-0.11} - \sigma_{max}^{-0.1}}{\sigma_{max}^{-0.1}}, \qquad R_{inc} = \frac{\sigma_{max}^{-0.09} - \sigma_{max}^{-0.1}}{\sigma_{max}^{-0.1}}.$$

From Figure 8, it can be seen that the ratio of the fluctuation is less than 0.6% for a 10% deviation of the heat flux, which is small compared to the reduction of the stress by using the optimal h_{gs}.

Finally, we note that while the optimization process works well in reducing the stress level, it is also evident that in general the stress level is much lower in a conic crystal than in a cylindrical one of similar size, regardless of the optimization process. In the following we explore the possibility of further reducing thermal stress by shape optimization.

4.1.3 Optimal shape

The following calculations are for a crystal with a length of 33.5 cm and a seed crystal radius of $R_0 = 0.5$ cm. The mean crystal radius is $\overline{R} = 0.03$ m. We fix the slope of the crystal shape around

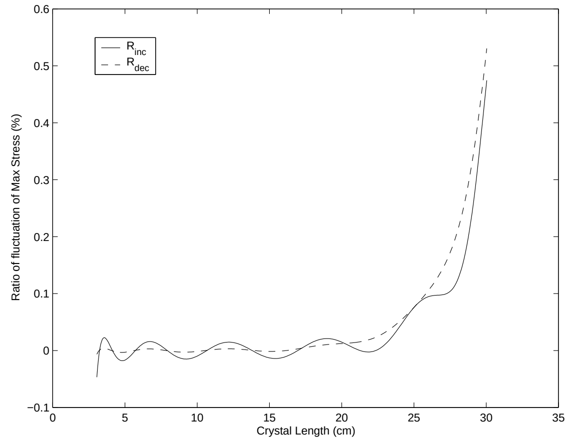


Figure 8: The ratio of the fluctuation of the history of the maximum von Mises stress with an increase of 10% of the heat flux from -0.1 and a decrease of 10% of the heat flux from -0.1. The optimal shape used for the calculations is presented in Figure 5.

$\tan 15°$. The parameters used for the optimization are $\rho = 20$, $\omega_1 = 0.1$, and $\omega_2 = 0.5$.

Figure 9(a) shows the linear h_{gs} used for the computation. Figure 9(b) is the optimal shape obtained by solving the Euler-Lagrange equations and a conical shape. Figure 9(c) shows the maximum von Mises stress corresponding to the optimal shape and the conical shape, respectively. Figure 9(d) shows the angle of the crystal shape in the (z, r) coordinate system corresponding the optimal shape. We can see that the reduction in thermal stress is significant while the slope of the crystal shape remains smooth with a slight increase of growth angle.

For the next set of computation we used an optimal h_{gs} obtained for a conical crystal. Figure 10(a) shows the shape of the optimal h_{gs}. Figure 10(b) shows the optimal shape obtained using $\rho = 20$, $\omega_1 = 0.1$, and $\omega_2 = 0.5$ with the conical shape. Figure 10(c) shows the maximum von Mises stress corresponding to the optimal shape and the conical shape, respectively. Figure 10(d) shows the angle of the optimal crystal shape in the (z, r) coordinate system. In this case, the reduction of the stress is not as significant, which shows that h_{gs} is nearly optimal even though it is obtained with a given conical shape.

We also carried our parameter studies and found that nearly the same solution is obtained for $\rho \geq 15$; $R_0' = \tan 15°/\sqrt{\epsilon}$ and $R_0' = \tan 6°/\sqrt{\epsilon}$. Figure 11 shows the effect of parameter ω_1 on the optimal solution. The linear profile of h_{gs} is shown in Figure 11(a). Figure 11(b) shows the optimal shape affected by parameter ω_1. Figure 11(c) shows the effect on maximum von Mises stress. Figure 11(d) shows the angle of the crystal shape in the (z, r) coordinate system affected by ω_1. In general, the effect of ω_1 is small.

In order to study the robustness of our optimal solution, we use the optimal shape calculated using the given linear heat transfer coefficient (Figure 9) for the calculations. We assume the perturbation of the heat flux γ is 10% use the same notation as in Section 4.1.2. Figure 12 shows that the fluctuation in stress is less than 0.07%.

Remark. We have shown that significant stress reduction can be achieved by optimizing either the

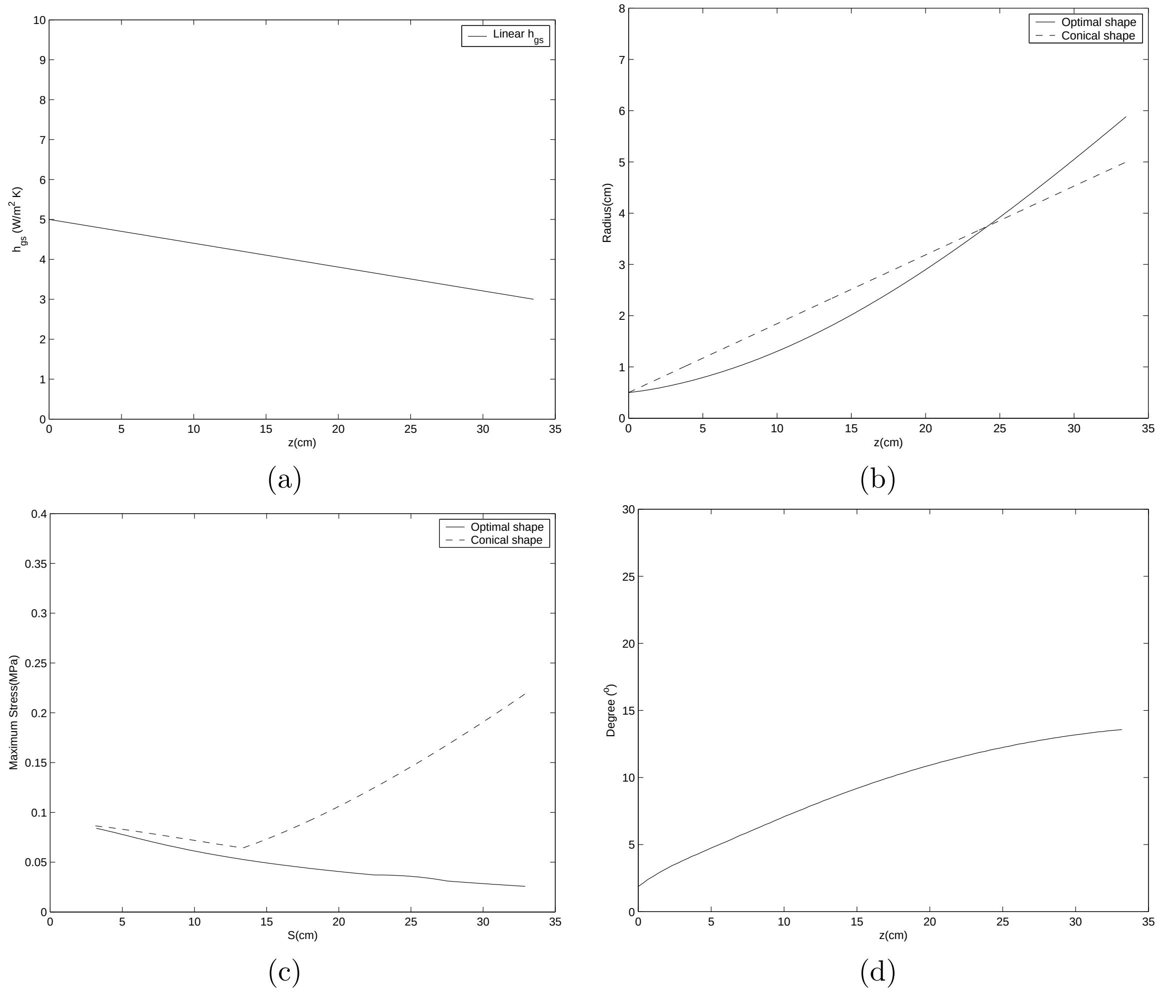


Figure 9: (a) The given linear h_{gs}. (b) The optimal shape corresponding to the given h_{gs} when $\rho = 20$, $\omega_1 = 0.1$, and $\omega_2 = 0.5$ and a conical shape. (c) The history of maximum von Mises stress corresponding to the optimal shape and the conical shape. (d) The degree of the crystal profile in the (z, r) coordinate system for the optimal shape.

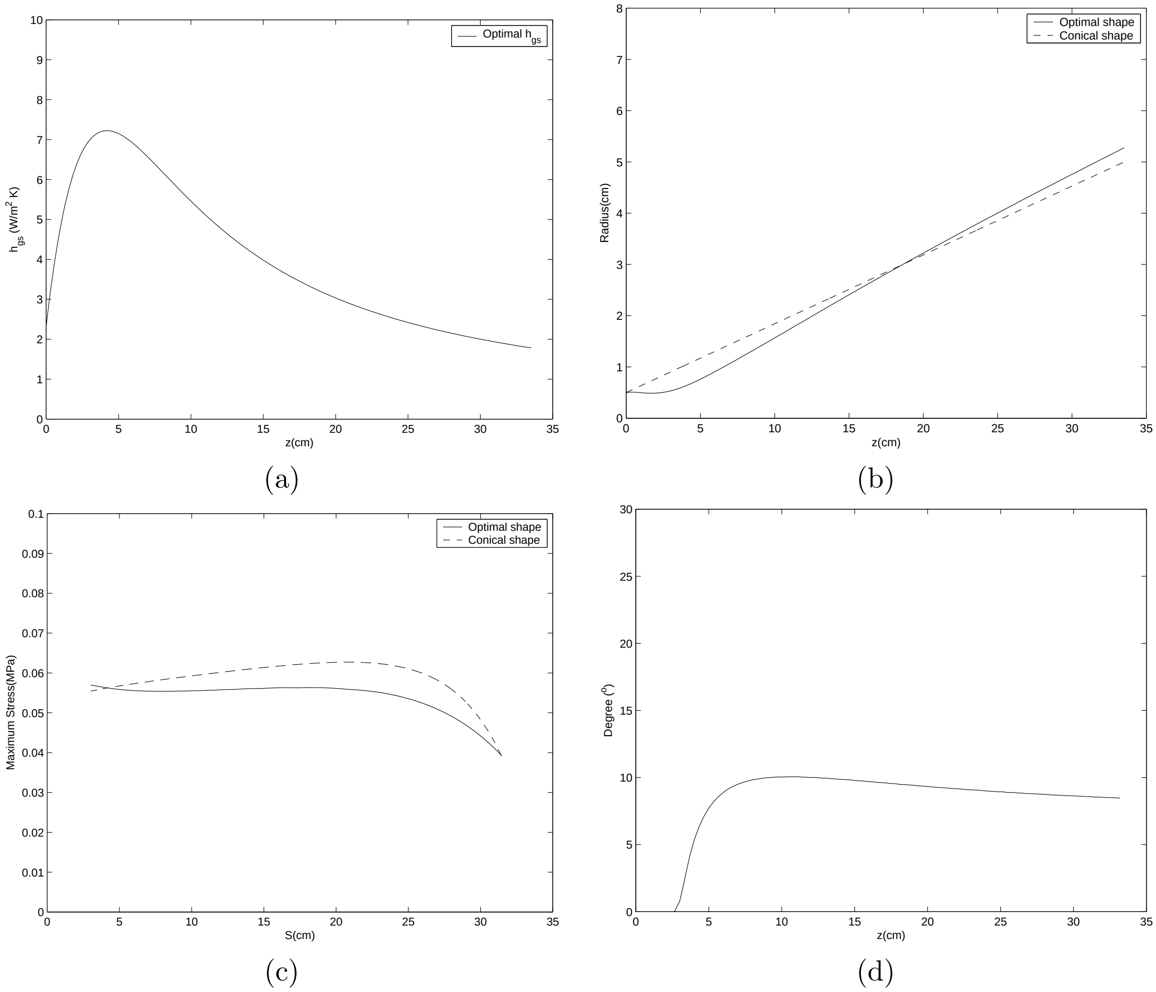


Figure 10: (a) The given optimal h_{gs}. (b) The optimal shape corresponding to the given h_{gs} when $\rho = 20$, $\omega_1 = 0.1$, and $\omega_2 = 0.5$ and a conical shape. (c) The history of maximum von Mises stress corresponding to the optimal shape and the conical shape. (d) The degree of the crystal profile in the (z, r) coordinate system for the optimal shape.

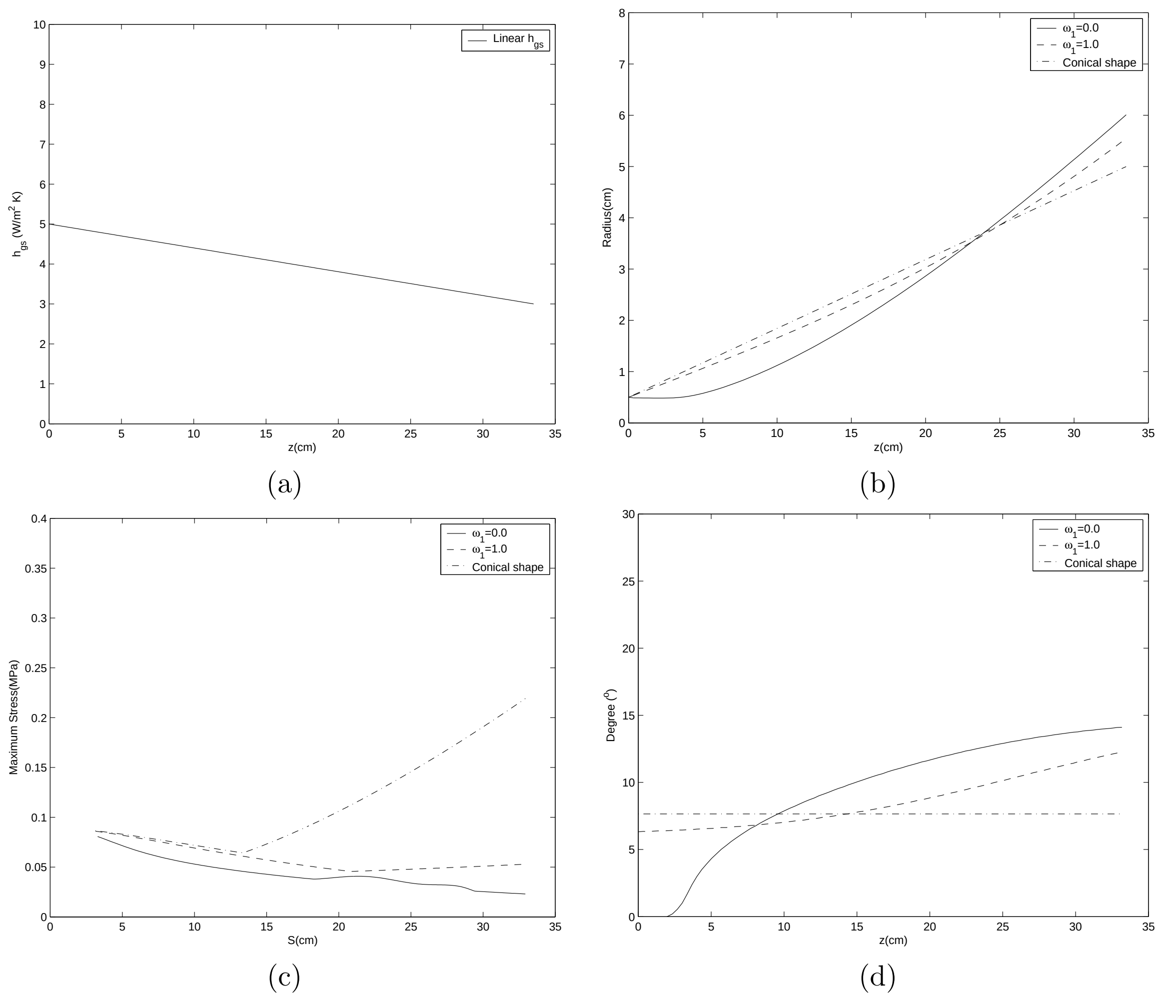


Figure 11: (a) The given linear h_{gs}. (b) The two optimal crystal shapes for $\omega_1 = 0.0$ and $\omega_1 = 1.0$. (c) The history of maximum von Mises stress during the growth of the crystal for $\omega_1 = 0.0$, $\omega_1 = 1.0$ hen $\rho = 20$, $R_0' = \tan 15^\circ/\sqrt{\epsilon}$, and $\omega_2 = 0.1$ and conical shape. (d) The degree of the crystal profile in the (z, r) coordinate system for $\omega_1 = 0.0$ and $\omega_1 = 1.0$.

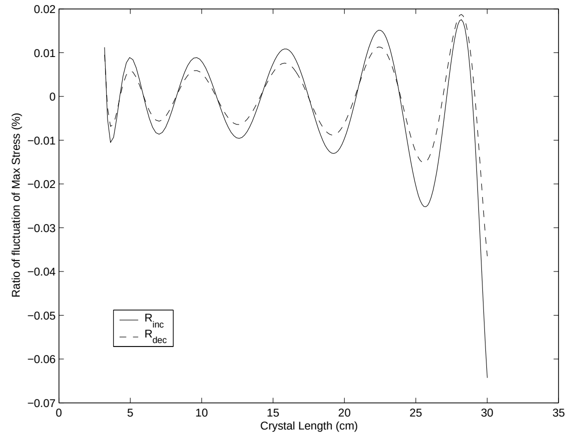


Figure 12: The ratio of the fluctuation of the history of the maximum von Mises stress with an increase of 10% of the heat flux from -0.1 and a decrease of 10% of the heat flux from -0.1. The optimal shape used for the calculations is prsented in Figure 9.

heat transfer coefficient or the crystal shape. This is due to the fact that there are two contributing factors in the thermal stress, as shown in (18). By taking advantages of the inter-play of these two contributing factors, we can search for the best conditions so that these two components cancel out each other. To show this, we have plotted the components of the stress. Figure 13(a) shows $R'\Theta_{0,z}$ component and $\beta\Theta_0$ component of von Mises stress $\sigma_{\rm VM}$ and the combination of the two components along the crystal profile at the end of growth of the crystal for the conical crystal shape. Figure 13(b) shows $R'\Theta_{0,z}$ component and $\beta\Theta_0$ component of von Mises stress $\sigma_{\rm VM}$ and the combination of the two components along the crystal profile at the end of growth of the crystal for the optimal crystal shape. The heat transfer coefficient used for the calculations is assumed to be linear.

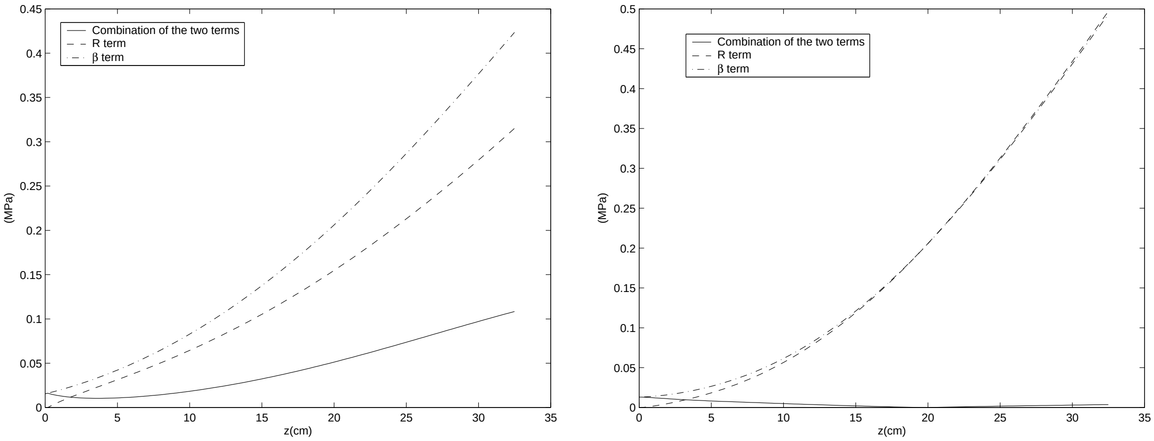
4.2 Effect of radiative transfer

We now treat the radiative heat transfer separately from the convective heat transfer. We assume that only the convective heat transfer coefficient can be optimized. The numerical results presented in this section are obtained using the radiation model (2). We assume that a desirable crystal shape can be produced by adjusting the heat flux of the melt with a given pull rate of 2.2 cm/hour and the radiative transfer coefficient is $r_c = 5.24 \times 10^{-9}$ W/(m^{2}K^2).

4.2.1 Optimal β

For a given conical crystal shape we assume the radius of the seed crystal is $R_0 = 0.5$ cm, the length and the maximum radius of the crystal at the end of growth are 33.5 cm and $R_{\rm max} = 5.7$ cm, respectively. The following parameters are used for all optimization calculations: $\rho = 20$; $\omega_2 = 0.2$; $\beta_0 = 1.0$.

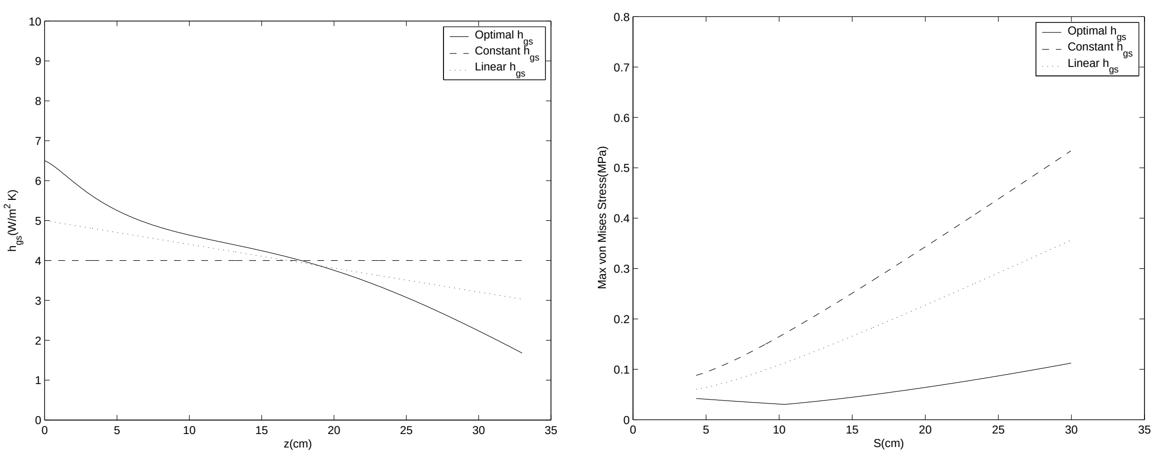
Figure 14(a) shows the shape of three heat transfer coefficients. We used $\omega_1 = 0.3$ for the optimization problem. Figure 14(b) shows the maximum von Mises stress during the growth of



(a) (b)

Figure 13: (a) $R'\Theta_{0,z}$ component and $\beta\Theta_0$ component of von Mises stress σ_{VM} and the combination of the two components along the crystal profile at the end of growth of the crystal for the conical crystal shape. (b) $R'\Theta_{0,z}$ component and $\beta\Theta_0$ component of von Mises stress σ_{VM} and the combination of the two components along the crystal profile at the end of growth of the crystal for the optimal crystal shape when $R_0' = \tan 15°/\sqrt{\epsilon}$, $\rho = 20$, $\omega_1 = 0.5$, and $\omega_2 = 0.2$.

the crystal for these three h_{gs}. Again, the reduction of the thermal stress using the optimal h_{gs} is significant.



(a) (b)

Figure 14: (a) Three heat transfer coefficients; (b) Maximum von Mises stress during the growth. For the optimal h_{gs} we used the following parameters: $\rho = 20$; $\omega_1 = 0.3$; $\omega_2 = 0.2$; $\beta_0 = 1.0$.

We also performed parameter studies and found that different parameter values produce the same solution when ρ is greater than critical values. Robustness tests also yield similar results as before.

4.2.2 Optimal shape

We present the following optimal solutions for the given heat transfer coefficients h_{gs}. For all the calculations we assume that the crystal length is 33.5 cm, the radius of the seed crystal is $R_0 = 0.005$ m, and the mean crystal radius is $\overline{R} = 0.03$ m. We assume the slope of the crystal profile is around $\tan 10°$. The parameters used for the optimization are $\rho = 20$ and $\omega_2 = 0.6$.

Figure 15(a) shows the heat transfer coefficient used for finding the optimal shape. Figure 15(b) shows a given conic shape and the optimal shape. The parameters used for finding the optimal shape are as follows: $\omega_1 = 0.2$, $\omega_2 = 0.6$, $\rho = 20$, and $R_0' = \tan 10°/\sqrt{\epsilon}$. Figure 15(c) compares the maximum von Mises stress and the stress at the final length for the two shapes. Reduction in thermal stress is again apparent when the optimal shape is used.

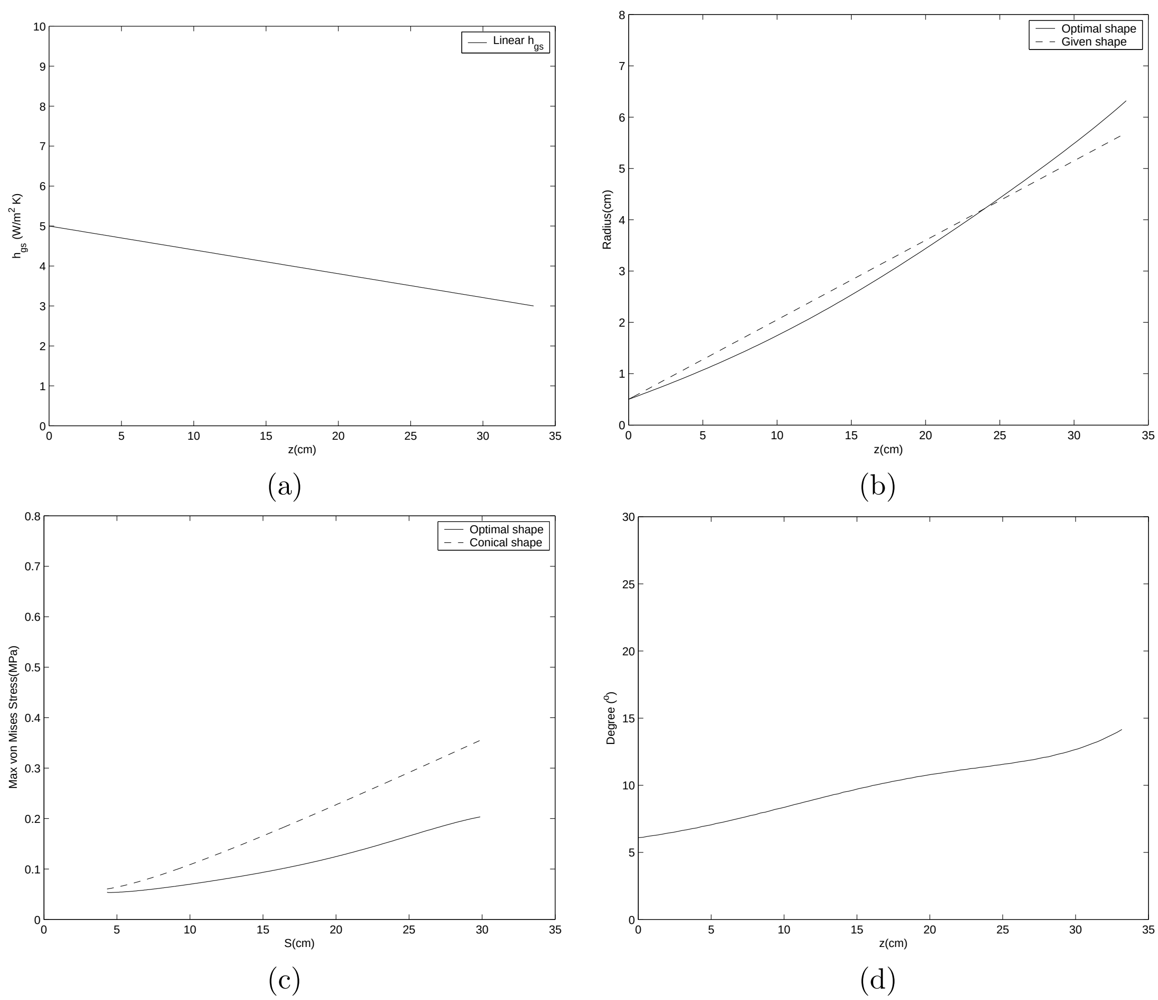


Figure 15: (a) The given linear h_{gs}. (b) A given conic shape and the optimal shape. The parameters used for finding the optimal shape are as follows: $\omega_1 = 0.2$, $\omega_2 = 0.6$, $\rho = 20$, and $R_0' = \tan 10°/\sqrt{\epsilon}$. (c) The maximum von Mises stress for the two shapes. (d) The degree of the crystal profile in the (z, r) coordinate system for the optimal shape.

We have carried out parameter study, sensitivity analysis (robustness) and computation of combined β and shape optimizations. The numerical results are similar to those obtained earlier for the combined convective-radiative heat transfer case. It suggest that our optimization procedure is relatively robust. Even when part of the heat transfer (radiative) can not be optimized, the procedure we used seem to be able to find the suitable crystal shape which can reduce the stress significantly.

5 Conclusions

One of the main concerns of using Czochralski technique to grow single compound crystals is the difficulty of controlling the appearance of crystalline defects. In practice, one has to find suitable thermal environment and "magic shapes" (defined by the axial variation of the lateral surface) by trial-and-error so that defect free crystals can be grown. The "shape effect" is not a serious issue for more commonly used single material crystals such as silicon where cylindrical crystals are routinely grown. For compound crystals, however, it is extremely difficult to grow defects free crystals in cylindrical shape due to their low resistance to resolve stress. As a result, shape control become a critical issue in practice [7, 6]. The combination of shape and thermal effects on defect distribution is not easy to manipulate and the control process is more delicate.

The shape effect was demonstrated in [5] where an explicit formula for the von Mises stress was derived. The thermal stress level is determined by heat flux through the lateral surface and the shape variation of the lateral surface. When the growth condition is not optimized, these two components both contribute to the overall stress. On the other hand, if the growth process are controlled carefully, it is possible to find optimal conditions so that these two components cancel each other out. However, finding the most favorable conditions to balance these two components is non-trivial.

By setting up a constrained optimization problem under the framework of optimal control, we are able to approach the stress reduction problem systematically. Optimal control methodology provides a valuable tool and has been used previous in the crystal growth literature. However, most of the previous study assumed simple cylindrical geometry for the crystal which is appropriate for growing silicon and other common single crystals. In this paper, we discuss both the thermal and shape effects. By using an semi-analytical solution for the thermal stress, the optimization process is more efficient than a full numerical simulation. We are able to show that stress can be reduced significantly by choosing a suitable thermal environment (lateral heat flux) or by growing the crystal into an optimal shape.

In order to keep our problem mathematically manageable, we have made several simplifications. As a consequence, direct application of our results may not be suitable for a complicated growth procedure. Verification of our model is necessary and work is currently underway to incorporate the effect of melt flow. Nevertheless, the results reported provide useful insights and can be used as a general guide, especially when the melt flow can be controlled using various techniques such as the electromagnetic field.

Acknowledgements

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Appendix A. Euler-Lagrange equations for optimization problem I

Using the calculus of variation, we derive the first-order necessary conditions for the optimization problem given by equation (23) with constraints (19a)-(19e).

Taking the first variation of (23) and using product rule yield

$$
\begin{aligned}
\delta J_1 \;=\;& \delta \int_0^L \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right) R(z)\right]^2 dz \\
&+\delta \int_0^L \lambda(z) \left[\Theta_{0,zz}(z) + \frac{2}{R(z)}\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)\right] dz \\
&+\omega_1 \delta \int_0^L (\beta(z) - \beta_0)^2 \, dz - \omega_2 \delta \int_0^L \Theta_{0,z}^2(z) dz \\
&+\delta \left[\mu\left(\beta_0 L - \int_0^L \beta(z) dz\right)\right] + \frac{\rho}{2}\delta \int_0^L \beta^2(z) H(-\beta(z)) dz \\
=\;& \int_0^L 2R^2(z)\left[R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right] \\
&\qquad \left[R'(z)\delta\Theta_{0,z}(z) - \beta(z)\delta\Theta_0(z) - \Theta_0(z)\delta\beta(z) - f'(\Theta_0(z))\delta\Theta_0(z)\right] dz \\
&+ \int_0^L \delta\lambda(z) \left[\Theta_{0,zz}(z) + \frac{2}{R(z)}\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)\right] dz \\
&+ \int_0^L \lambda(z) \left[\delta\Theta_{0,zz}(z) + \frac{2}{R(z)}\left(R'(z)\delta\Theta_{0,z}(z) - \delta\beta(z)\Theta_0(z) - \beta(z)\delta\Theta_0(z)\right.\right. \\
&\qquad\qquad \left.\left. - f'(\Theta_0(z))\delta\Theta_0(z)\right)\right] dz \\
&+2\omega_1 \int_0^L (\beta(z) - \beta_0)\,\delta\beta(z) dz - 2\omega_2 \int_0^L \Theta_{0,z}(z)\delta\Theta_{0,z}(z) dz \\
&+\delta\mu\left(\beta_0 L - \int_0^L \beta(z) dz\right) - \mu \int_0^L \delta\beta(z) dz + \rho \int_0^L \beta(z)\delta\beta(z) H(-\beta(z)) dz
\end{aligned}
$$

Using integration by parts for above expression and the boundary conditions for Θ_0, we obtain the following after simplifying.

$$
\begin{aligned}
\delta J_1 \quad = \quad & \int_0^L \left[\lambda_{zz} - \left(\frac{2R'\lambda}{R}\right)_z - 2\lambda\beta/R - 2\beta R^2\left(R'\Theta_{0,z} - \beta\Theta_0\right)\right. \\
& -2\left(R^2R'\left(R'\Theta_{0,z} - \beta\Theta_0\right)\right)_z - 2R^2\left(R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right)f'(\Theta_0) \\
& +2R^2\beta f(\Theta_0) - 2\left(2RR'^2 f(\Theta_0) + R^2R'' f(\Theta_0) + R^2R'f'(\Theta_0)\Theta_{0,z}\right) \\
& \left. -2\lambda f'(\Theta_0)/R + 2\omega_2\Theta_{0,zz}\right]\delta\Theta_0 dz \\
& + \int_0^L \left[\Theta_{0,zz} + \frac{2}{R}\left(R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right)\right]\delta\lambda(z)dz \\
& - \int_0^L \left[2R^2\left(R'\Theta_{0,z} - \beta\Theta_0\right)\Theta_0 + 2\lambda\Theta_0/R - 2\omega_1\left(\beta - \beta_0\right) + \mu - \rho\beta H(-\beta)\right]\delta\beta dz \\
& +\delta\mu\left(\beta_0 L - \int_0^L \beta(z)dz\right) + \frac{2R'\lambda}{R}\delta\Theta_{0,z}(L) \\
& + \left[\lambda_z - \left(\delta_0 + \frac{2R'}{R}\right)\lambda - 2R^2R'\left[\left(R'\delta_0 - \beta\right)\Theta_0 - R'\delta_0\Theta_{ch}\right] - 2R^2R'f(\Theta_0)\right]\delta\Theta_0(0)
\end{aligned}
$$

Since $\Theta_0(z)$, $\beta(z)$, $\lambda(z)$ and μ are arbitrary, the optimality conditions are obtained by setting the coefficients of $\delta\Theta_0(z)$, $\delta\beta(z)$, $\delta\lambda(z)$ and $\delta\mu$ equal to zero. The resultant conditions stated are exactly the Euler-Lagrange equations and the boundary conditions given in (24d)-(25d).

Appendix B. Euler-Lagrange equations for optimization problem II

We derive the first-order necessary conditions for the optimization problem given by equation (26) with constraints (22a)-(22f) using the calculus of variation.

Taking the first variation of (26) and using product rule yield

$$
\begin{aligned}
\delta J_2 &= \delta \int_0^L \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right) R(z)\right]^2 dz \\
&+\omega_1 \delta \int_0^L \left(R'(z) - R_0'\right)^2 dz - \omega_2 \delta \int_0^L \Theta_{0,z}^2(z) dz \\
&+\delta \int_0^L \lambda(z) \left[R(z)\Theta_{0,zz}(z) + 2\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)\right] dz \\
&+\delta \left[\mu \left(L - \int_0^L R^2(z) dz\right)\right] + \frac{\rho}{2} \delta \int_0^L \left(R(z) - R_0\right)^2 H(R_0 - R(z)) dz \\
&= \int_0^L 2\left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right) R(z)\right] \\
&\qquad \left[\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right) \delta R(z) \right. \\
&\qquad \left. + \left(\delta R'(z)\Theta_{0,z}(z) + R'(z)\delta\Theta_{0,z}(z) - \beta(z)\delta\Theta_0(z)\right) R(z)\right] dz \\
&+2\omega_1 \int_0^L \left(R'(z) - R_0'\right) \delta R'(z) dz - \omega_2 \int_0^L 2\Theta_{0,z}(z)\delta\Theta_{0,z}(z) dz \\
&+ \int_0^L \delta\lambda(z) \left[R(z)\Theta_{0,zz}(z) + 2\left(R'(z)\Theta_{0,z}(z) - \beta(z)\Theta_0(z) - f(\Theta_0(z))\right)\right] dz \\
&+ \int_0^L \lambda(z) \left[\delta R(z)\Theta_{0,zz}(z) + R(z)\delta\Theta_{0,zz}(z) \right. \\
&\qquad \left. +2\left(\delta R'(z)\Theta_{0,z}(z) + R'(z)\delta\Theta_{0,z}(z) - \beta(z)\delta\Theta_0(z) - f'(\Theta_0(z))\delta\Theta_0(z)\right)\right] dz \\
&+\delta\mu \left(L - \int_0^L R^2(z) dz\right) - \mu \int_0^L 2R(z)\delta R(z) dz \\
&+\rho \int_0^L \left(R(z) - R_0\right) H(R_0 - R(z)) \delta R(z) dz
\end{aligned}
$$

Using integration by parts for above expression and the boundary conditions for $\Theta_0(z)$ and $R(z)$, we obtain the following after simplifying.

$$
\begin{aligned}
\delta J_2 \;=\; & \int_0^L \left[R\Theta_{0,zz} + 2\left(R'\Theta_{0,z} - \beta\Theta_0 - f(\Theta_0)\right)\right]\delta\lambda dz \\
& + \int_0^L \left[2\beta^2 R^2\Theta_0 + 2R\left(\beta R\Theta_0 - 2RR'\Theta_{0,z}\right)R'' + 2\beta' R^2 R'\Theta_{0,z} + R\lambda'' - \lambda R'' - 2\beta\lambda\right. \\
& \qquad +2R^2\beta\Theta_0 f'(\Theta_0) + 2R^2 f(\Theta_0)f'(\Theta_0) + 4RR'2f(\Theta_0) + 2R^2R''f(\Theta_0) \\
& \qquad \left. +2R^2\beta f(\Theta_0) - 2\lambda f'(\Theta_0) + 2\omega_2\Theta_{0,zz}\right]\delta\Theta_0 dz \\
& + \int_0^L \left[6RR'^2\Theta_{0,z}^2 - 12\beta\Theta_0 RR'\Theta_{0,z} + 6\beta^2 R\Theta_0^2 - 2R^2R''\Theta_{0,z}^2 + 2\beta' R^2\Theta_0\Theta_{0,z} - 2\omega_1 R''\right. \\
& \qquad + 2\beta R^2\Theta_{0,z}^2 - \lambda\Theta_{0,zz} - 2\lambda'\Theta_{0,z} - 2\mu R + \rho\left(R - R_0\right)H(R_0 - R) \\
& \qquad \left. - 4RR'\Theta_{0,z} f(\Theta_0) + 6R^2 f^2(\Theta_0) + 8R\beta\Theta_0 f(\Theta_0) + 2R^2 f'(\Theta_0)\Theta_{0,z}^2\right]\delta R dz \\
& +\delta\mu\left(L - \int_0^L R^2(z)dz\right) + \lambda R\delta\Theta_{0,z}(L) \\
& + \left\{\lambda' R - \lambda R' + 2R^2R'\left[\beta\Theta_0 - \delta_0 R'\left(\Theta_0 - \Theta_{ch}\right)\right]\right. \\
& \qquad \left. + 2\omega\delta_0\left(\Theta_0 - \Theta_{ch}\right) + 2R^2R' f(\Theta_0)\right\}\delta\Theta_0(0) \\
& + \left[2R^2\left(R_z\Theta_{0,z} - \beta\right)\Theta_{0,z} + 2\lambda\Theta_{0,z} + 2\omega_1\left(R' - R_0'\right) - R^2 f(\Theta_0)\Theta_{0,z}\right]\delta R(L)
\end{aligned}
$$

Since $\Theta_0(z)$, $R(z)$, $\lambda(z)$ and μ are arbitrary, the optimality conditions are obtained by setting the coefficients of $\delta\Theta_0(z)$, $\delta R(z)$, $\delta\lambda(z)$ and $\delta\mu$ equal to zero. The resultant conditions stated are exactly the Euler-Lagrange equations and the boundary conditions given in (27a)-(28f).