Stability of Detached Solidification

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Abstract

Bridgman crystal growth can be conducted in the so-called “detached” solidification regime, where the growing crystal is detached from the crucible wall. A small gap between the growing crystal and the crucible wall, of the order of 100 micrometers or less, can be maintained during the process. A meniscus is formed at the bottom of the melt between the crystal and crucible wall. Under proper conditions, growth can proceed without collapsing the meniscus. The meniscus shape plays a key role in stabilizing the process. Thermal and other process parameters can also affect the geometrical steady-state stability conditions of solidification. The dynamic stability theory of the shaped crystal growth process has been developed by Tatarchenko [1]. It consists of finding a simplified autonomous set of differential equations for the radius, height, and possibly other process parameters. The problem then reduces to analyzing a system of first order linear differential equations for stability. Here we apply a modified version of this theory for a particular case of detached solidification. Approximate analytical formulas as well as accurate numerical values for the capillary stability coefficients are presented. They display an unexpected singularity as a function of pressure differential. A novel approach to study the thermal field effects on the crystal shape stability has been proposed. In essence, it rectifies the unphysical assumption of the model [1] that utilizes a perturbation of the crystal radius along the axis as being instantaneous. It consists of introducing time delay effects into the mathematical description and leads, in general, to stability over a broader parameter range. We believe that this novel treatment can be advantageously implemented in stability analyses of other crystal growth techniques such as Czochralski and float zone methods.

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DETACHED BRIDGMAN GROWTH

quartz ampoule

furnace

melt

crystal

contact angle

growth angle

Dynamic Growth Stability –
V.A.Tatarchenko – Shaped Crystal Growth
Linear response of perturbed crystal radius and height

\[
\delta \dot{R} = A_{RR} \delta R + A_{Rh} \delta h
\]

\[
\delta \dot{h} = A_{hR} \delta R + A_{hh} \delta h
\]

Stable growth if

\[
A_{RR} + A_{hh} < 0
\]

\[
A_{RR} A_{hh} - A_{Rh} A_{hR} > 0
\]
Young-Laplace equation

\[
\frac{z''}{(1 + z'^2)^{3/2}} + \frac{z'}{r(1 + z'^2)^{1/2}} = a - b z(r)
\]

\[
a = \frac{\Delta P_m r_c}{\gamma}
\]
\[
b = \frac{\rho g_0 r_c^2}{\gamma}
\]
\[
\Delta P_m = \left( P_{top} + \rho g \right) H - P_{bot}
\]
CAPILLARY COEFFICIENTS - THEORY

\[ A_{RR} = -V \frac{\partial \beta}{\partial R} \quad A_{Rh} = -V \frac{\partial \beta}{\partial h} \]

\[ V - \text{pulling rate} \]

\[ \frac{\partial \beta}{\partial R} = \frac{(aR - \cos \alpha_{gr}) \sin(\alpha_{gr})^2}{Rs \left( i\alpha_{gr} \right)^3 - 1b \left( 5 - R \right)^2 \left( 2 - 4/R - 8 \right)} \]

\[ \frac{\partial \beta}{\partial h} = -\frac{r_c^2 \rho g}{2\gamma Rs \left( i\alpha_{gr} \right)^3 - 1b \left( 5 - R \right)^2 \left( 2 - 4 + R - 8 \right)} \left( 1 - R^2 \right) \sin \alpha_{gr}^2 \]
\[ \theta = 156^\circ, \alpha_g = 11^\circ, \]
\[ b = 3.311927 \]
Boundary condition at the interface

\[ V_C L = k_S \frac{\partial T_S}{\partial z} - k_L \frac{\partial T_L}{\partial z} \]

Growth rate

\[ V_C = V + \frac{\partial h}{\partial t} \]

Height perturbation – Tatarchenko’s model

\[
\frac{\partial \delta h}{\partial t} = L^{-1} \left( k_S \frac{\partial G_S}{\partial h} - k_L \frac{\partial G_L}{\partial h} \right) \delta h + L^{-1} \left( k_S \frac{\partial G_S}{\partial R} - k_L \frac{\partial G_L}{\partial R} \right) \delta R
\]

\[ \delta G_S \neq \frac{\partial G_S}{\partial R} \delta R \]

questionable
ONE-DIMENSIONAL LUMP HEAT MODEL

For melt
\[
\frac{\partial^2 T_L}{\partial z^2} + \frac{V}{\kappa_L} \frac{\partial T_L}{\partial z} - \frac{\mu_L}{k_L r_C} \frac{2}{r_C} (T_L - T_e) = 0
\]

For crystal
\[
\frac{\partial^2 T_S}{\partial z^2} + \frac{V}{\kappa_S} \frac{\partial T_S}{\partial z} - \frac{\mu_S}{k_S} \frac{2}{R + \delta(z)} (T_S - T_e) = 0
\]

\[
A_{hh} = -L^{-1} k_L \frac{T_{hot} - T_0}{H^2} - L^{-1} k_S \frac{T_0 - T_{cold}}{l^2}
\]

Perturbation of thermal gradient at the interface due to crystal radius variation (convolution):
\[
\delta G_S = -\frac{2 \mu_S (T_0 - T_{cold}) V}{R^2 k_S} \int_0^t \delta R (t') e^{-V(t-t')} \sqrt{\frac{V}{\kappa_S}}^2 + \frac{8 \mu_S}{k_S R} dt'
\]
GENERALIZED RESPONSE

\[ \delta \dot{h} = \int_{0}^{t} \delta R(t') G(t - t') \, dt' + A_{hh} \delta h \]

\[ \delta \dot{R} = A_{RR} \delta R + A_{Rh} \delta h \]

Laplace transform solution

\[ h(s) = \frac{G(s) \delta R(0) + s(\delta h(0) - A_{RR})}{s^2 - s(A_{RR} + A_{hh}) + A_{RR}A_{hh} - A_{Rh}G(s)} \]

\[ R(s) = \frac{\delta R(0)(s - A_{hh}) + A_{Rh} \delta h(0)}{s^2 - s(A_{RR} + A_{hh}) + A_{RR}A_{hh} - hG(s)A_{h}} \]
Tatarchenko model gives

\[ A_{hR} = - \frac{2V \mu_s (T_0 - T_e)}{LR^2 s_0} \]

Our model

\[ G(s) = \frac{A_{hR}s_0}{s + s_0} \]

\[ s_0 = V \sqrt{\left( \frac{V}{\kappa S} \right)^2 + \frac{8\mu_s}{k_s R}} \]

Response for the induced perturbation can be studied by analyzing the roots of the following polynomial

\[ s^2 \left( s + s_0 \right) - s \left( s + s_0 \right) \left( A_{RR} + A_{hh} \right) + A_{RR} A_{hh} \left( s + s_0 \right) - A_{Rh} A_{hR} s_0 = 0 \]
**MODIFIED STABILITY CRITERION**

Conditions for stable growth

**Our model**

\[
A_{RR}A_{hh} - A_{Rh}A_{hR} > 0 \\
A_{RR} + A_{hh} < s_0 \\
A_{RR}A_{hh} - s_0(A_{RR} + A_{hh}) > 0
\]

**Tatarchenko model**

\[
A_{RR}A_{hh} - A_{Rh}A_{hR} > 0 \\
A_{RR} + A_{hh} < 0
\]
MODEL APPROXIMATIONS

- Environmental temperature is constant
- One-dimensional lump type approximation
- Capillarity – zero order model – no meniscus motion effects, no triple point effects
CONCLUSION

- Capillarity coefficients display singularity
- Thermal response for the radius perturbation is of the convolution type – this modified model is applicable for other types of shaped crystal growth: Czochralski, Float Zone, etc.