

# Morphological Stability of a Cylinder

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The stability of the shape of a solid cylinder crystallizing in a supercooled liquid is treated. The effects of solute diffusion, slightly anisotropic surface tension and interface kinetics are included. The resulting stability equations are applied to the specific case of ice cylinders.

Key Words: Cylinder; ice; interface kinetics; morphological stability; solute diffusion; surface tension.

## 1. Introduction

The stability of the shape of a solid growing by diffusion or heat flow was first studied by Mullins and Sekerka [1]<sup>1</sup>, who determined the stability criteria for a sphere. The cylindrical geometry has been treated by Coriell and Parker [2] and Kotler and Tiller [3]. These studies assumed that the interface properties were isotropic. Cahn [4] has taken account of slightly anisotropic surface free energy for the sphere. Recently, we have made experimental studies [5, 6] of the morphological stability of ice cylinders. In connection with this experimental program, it appears desirable to work out the stability of a cylindrical shape taking account of slightly anisotropic surface tension and interface kinetics. In this paper we treat the case of a cylinder crystallizing from a binary melt; it is assumed that the surface tension and kinetic coefficient are slightly anisotropic. For the case of isotropic interface properties, the problem reduces to the case previously treated by Kotler and Tiller [3].

Although our calculation is general, we are particularly interested in the case of an ice cylinder growing from pure water and from water with impurities added. We present some specific calculations for these cases.

## 2. Formulation and Calculation

We wish to solve Laplace's equation [1, 2] for the temperature  $T$  and concentration  $C$  of impurity. The subscripts  $S$  and  $L$  denote solid and liquid, respectively, while a subscript  $I$  denotes that the quantity

is to be evaluated at the interface. We assume a slightly perturbed cylinder of shape [2]

$$r = R + \delta e^{ik\Phi} e^{i2\pi z/\lambda}, \quad (1)$$

where  $r$ ,  $\Phi$ , and  $z$  are the usual cylindrical coordinates,  $R$  is the radius of the unperturbed cylinder,  $\delta$  is the amplitude of the perturbation ( $\delta/R \ll 1$ ), and  $k$  and  $\lambda$  determine the shape of the perturbation. Neglecting diffusion in the solid, the equations to be solved are

$$\nabla^2 T_L = \nabla^2 T_S = \nabla^2 C_L = 0 \quad (2)$$

with the auxiliary equations

$$T_L(R_b) = T_b \quad (3a)$$

$$T_S(0) - \text{finite} \quad (3b)$$

$$T_{LI} = T_{SI} \quad (3c)$$

$$C_L(R_c) = C_b \quad (3d)$$

$$v = \frac{k_S}{L_v} \left( \frac{\partial T_S}{\partial r} \right)_I - \frac{k_L}{L_v} \left( \frac{\partial T_L}{\partial r} \right)_I \quad (3e)$$

$$v = \left\{ \sum_k \mu_k e^{ik\Phi} \right\} \{ T_e - T_{LI} \} \quad (3f)$$

$$v = \frac{-D_L}{C_{LI}(1-j)} \left( \frac{\partial C_L}{\partial r} \right)_I \quad (3g)$$

$$v = \dot{R} + \dot{\delta} e^{ik\Phi} e^{i2\pi z/\lambda} \quad (3h)$$

$$T_e = T_M - T_M K \left\{ \sum_k \Gamma_k e^{ik\Phi} \right\} - m C_{LI} \quad (3i)$$

<sup>1</sup> Figures in brackets indicate the literature references at the end of this paper.

and

$$K = \frac{1}{R} + \frac{\delta}{R^2} (k^2 + k_z^2 - 1) e^{ik\Phi} e^{i2\pi z/\lambda}. \quad (3j)$$

In the above equations  $R_b$  is the bath radius,  $T_b$  (a constant) is the temperature at  $R_b$ ,  $R_c$  is the radius at which the concentration of impurity is  $C_b$  (a constant),  $v$  is the velocity of the interface,  $k_s$  and  $k_L$  are the thermal conductivities of the solid and liquid, respectively,  $L_v$  is the latent heat per unit volume of the solid, the  $\left(\sum_k \mu_k e^{ik\Phi}\right)$  is the slightly anisotropic

linear kinetic coefficient,<sup>2</sup>  $T_e$  is the equilibrium temperature of the interface,  $D_L$  is the solute diffusion coefficient,  $j$  is the partition coefficient (the ratio of the equilibrium concentration of solute on the solid side of the interface to that on the liquid side of the interface),  $\dot{R}$  and  $\delta$  are the rate of growth of the unperturbed cylinder and the perturbation, respectively,  $T_M$  is the melting point of a flat interface,  $K$  is the curvature of the perturbed cylinder, the  $\left(\sum_k \Gamma_k e^{ik\Phi}\right)$  is the

slightly anisotropic capillary constant,  $m$  is the freezing point lowering constant, and  $k_z = 2\pi R/\lambda$ . The solution of Laplace's equation in cylindrical coordinates is of the form  $C_1 + C_2 \ln r + e^{ik\Phi} e^{i2\pi z/\lambda} [C_3 I_k(k_z r/R) + C_4 K_k(k_z r/R)]$  where the  $C$ 's are constants and  $I_k$  and  $K_k$  are modified Bessel functions. A solution of this form is written for  $T_L$ ,  $T_S$ , and  $C_L$  and the constants are determined by the boundary conditions. Since we are treating small perturbations and slightly anisotropic surface tension and interface kinetics, we neglect all terms that are greater than first order in  $\delta$ ,  $\mu_k$ , and  $\Gamma_k$  ( $k \neq 0$ ), i.e., we omit terms containing  $\delta^2$ ,  $\delta\mu_k$ ,  $\delta\Gamma_k$ ,  $\Gamma_k\mu_k$ , etc., ( $k \neq 0$ ). Although the calculation is tedious, it is straightforward and we will not reproduce the details here. We define  $A_b = \ln(R_b/R)$ ,  $A_c = \ln(R_c/R)$ ,

$$\Delta T = T_M - (T_M \Gamma_0/R) - T_b, \quad \Delta \theta = \Delta T - m C_b$$

$$\beta_A = (k_L/RL_v \mu_0) + A_b, \quad \xi = [(1-j)k_L A_c]/[DL_v]$$

$$A_1 = [K_k(k_z R_b/R)/I_k(k_z R_b/R)],$$

$$D_1 = [K_k(k_z R_c/R)/I_k(k_z R_c/R)],$$

$$J_A = -k_z \{ [K'_k(k_z) - A_1 I'_k(k_z)] / [K_k(k_z) - A_1 I_k(k_z)] \},$$

$$J_D = -k_z \{ [K'_k(k_z) - D_1 I'_k(k_z)] / [K_k(k_z) - D_1 I_k(k_z)] \},$$

<sup>2</sup> The kinetic coefficient and the capillary constant can be written in this form since we are only considering first order terms in the perturbation amplitude and in the anisotropy. In general, for example, the kinetic coefficient  $\mu$  can be expanded as

$$\mu = \sum_k \int dk_z \mu_k(k_z) e^{ik\Phi} e^{ik_z z'/R}$$

where  $\Phi'$  and  $z'$  give the orientation of the surface. For a slightly perturbed cylinder, the orientational coordinates  $z'$  and  $\Phi'$  are related to the space coordinates  $z$  and  $\Phi$  by  $z' = 0 + \delta a_1$  and  $\Phi' = \Phi + \delta a_2$ , where  $a_1$  and  $a_2$  are functions of  $\Phi$  and  $z$ . Substituting these expressions in the expansion of  $\mu$ , expanding the exponentials, neglecting terms of the order  $\mu_k(k_z)\delta$ , and performing the integral over  $k_z$  we obtain  $\mu = \sum_k \mu_k e^{ik\Phi}$ , where  $\mu_k = \int dk_z \mu_k(k_z)$ . A similar procedure gives  $\Gamma = \sum_k \Gamma_k e^{ik\Phi}$ ; here  $\Gamma$  is  $(1/L_v)(\gamma + d^2\gamma/d\Phi^2)$  where  $\gamma$  is the surface tension.

$$H_I = k_z I'_k(k_z)/I_k(k_z),$$

where the prime indicates the derivative with respect to  $k_z$ . The results of the calculation are

$$\dot{R} = (k_L/RL_v A_b) (DT), \quad (4a)$$

where  $(DT)$  is given by

$$(DT) = A_b \left[ \frac{1}{2\xi} + \frac{\Delta T}{2\beta_A} \right] - \left| A_b \left\{ \left[ \frac{1}{2\xi} + \frac{\Delta T}{2\beta_A} \right]^2 - \frac{\Delta \theta}{\beta_A \xi} \right\}^{1/2} \right|. \quad (4b)$$

For the ratio  $(\dot{\delta}/\delta)/(\dot{R}/R)$  we obtain

$$\begin{aligned} \frac{\dot{\delta}R}{\delta\dot{R}} = & \left\{ 1 + \frac{\alpha}{\mu_0} + \frac{m\alpha}{\nu} \right\}^{-1} \left\{ J_A - 1 + \frac{m\alpha}{\nu} (J_D - 1) \right. \\ & + \frac{[J_A + (k_s/k_L)H_I]A_b}{(DT)} \left[ \frac{\dot{R}R\mu_k}{\delta\mu_k^2} \right. \\ & \left. \left. - \frac{T_M\Gamma_k}{\delta} - \frac{T_M\Gamma_0(k^2 + k_z^2 - 1)}{R} \right] \right\}, \quad (4c) \end{aligned}$$

where  $\alpha = (k_L/L_v R) (J_A + k_s H_I/k_L)$

and

$$\nu = \frac{k_L A_c}{L_v R C_b \xi} \left[ 1 - \frac{\xi(DT)}{A_b} \right] \left[ J_D - \frac{\xi(DT)}{A_c A_b} \right].$$

In the above equations  $\Gamma_k$  and  $\mu_k$  ( $k \neq 0$ ) are zero if  $k_z \neq 0$ , i.e., the anisotropic terms only effect  $\Phi$ -type perturbations and have no effect on perturbations along the axis of the cylinder. This follows, for example, since there are no terms of the form  $\Gamma_{k, k_z} e^{ik\Phi} e^{i2\pi z/\lambda}$  in eq (3i) (see footnote 2).

Thus one of the important results of this calculation is that the growth rate of  $z$ -perturbations is independent of slightly anisotropic surface tension and kinetic coefficient.

## Discussion

The general results of our stability analysis are given by eqs (4a-c). In this section, we apply these equations to the special case of ice cylinders (oriented with the  $c$ -axis of ice parallel to the cylinder axis) growing in slightly supercooled water ( $\Delta T \cong 0.1^\circ \text{C}$ ). We also consider the case in which impurities have been added to the water. We are interested in the case where  $R$  lies between 0.04 cm and 0.25 cm; the bath radius  $R_b$  in most of the experimental work was 0.875 cm. Under the above conditions we can simplify the equations by neglecting interface kinetics, i.e., we take

$1 + \frac{\alpha}{\mu_0} \cong 1$ . This implies that  $(k_L/RL_v) (J_A + H_I k_s/k_L) \ll \mu_0$ ; typically  $(J_A + H_I k_s/k_L) < 100$  and the above approximation is valid if  $\mu_0 \gg .05 \text{ cm deg}^{-1} \text{ sec}^{-1}$ .

Although the interface kinetic coefficient for ice growing perpendicular to the  $c$ -axis is not known, a lower bound can be obtained from bath undercooling measurements. For example, Lindenmeyer et al. [7], observed a growth velocity of  $1 \text{ cm sec}^{-1}$  at a bath undercooling of  $2^\circ \text{C}$ . Assuming a linear law, this gives  $\mu_0 > .5 \text{ cm sec}^{-1} \text{ deg}^{-1}$  [8]. Since  $\mu_0$  may be orders of magnitude greater than this, it appears valid to neglect interface kinetics. It follows that  $\beta_A \approx A_b$ .

It is almost always (and certainly under the experimental conditions of interest) valid to neglect the  $A_1$  and  $D_1$  terms in the definition of  $J_A$  and  $J_D$ . Defining  $H_K = -k_z K'_k(k_z)/(K_k(k_z))$ , we then have  $J_A \approx H_K \approx J_D$ . With this approximation and the neglect of interface kinetics, eqs (4b) and (4c) can be written

$$(DT) = \frac{1}{2}([A_b/\xi] + \Delta T) - \frac{1}{2}([A_b/\xi] + \Delta T)^2 - 4\Delta\theta A_b/\xi^{1/2} \quad (5a)$$

and

$$\frac{\dot{\delta}R}{\delta\dot{R}} = H_K - 1 + \frac{[H_K + (k_S/k_L)H_I]A_b}{(1 + m\alpha/\nu)DT} \times \left[ \frac{\dot{R}R\mu_k}{\delta\mu_0^2} - \frac{T_M\Gamma_k}{\delta} - \frac{T_M\Gamma_0(k^2 + k_z^2 - 1)}{R} \right] \quad (5b)$$

For pure water, there is further simplification, viz, taking  $m = 0$  and  $\xi \rightarrow 0$ ,

$$(DT) = \Delta T \quad (6a)$$

and

$$\frac{\dot{\delta}R}{\delta\dot{R}} = H_K - 1 + \frac{[H_K + (k_S/k_L)H_I]A_b}{DT} \times \left[ \frac{\dot{R}R\mu_k}{\delta\mu_0^2} - \frac{T_M\Gamma_k}{\delta} - \frac{T_M\Gamma_0(k^2 + k_z^2 - 1)}{R} \right] \quad (6b)$$

Recalling that for a  $k_z \neq 0$  perturbation  $\mu_k = 0$  and  $\Gamma_k = 0$  ( $k \neq 0$ ), eq (6b) becomes

$$\frac{\dot{\delta}R}{\delta\dot{R}} = H_K - 1 - \frac{T_M\Gamma_0(k^2 + k_z^2 - 1)A_b[H_K + (k_S/k_L)H_I]}{R(DT)} \quad (6c)$$

This equation has been used in references 5 and 6 to analyze the experimental data.

We now give a brief discussion of the choice of the bath boundary conditions, i.e., the choice of  $R_b$  and  $R_c$ ; we also discuss the use of Laplace's equation. For an infinite bath, the choice of these parameters has been previously discussed [2]. For an infinite bath  $R_b = R_\lambda$  where  $R_\lambda = R/(1.33\lambda)$  and  $\lambda$  satisfies

$$\lambda^2 e^{\lambda^2} Ei(-\lambda^2) + S = 0.$$

In the above equation  $Ei$  is the exponential integral function and for heat flow  $S = S_T = C_v(T_l - T_b)/L_v$  and for impurity diffusion [9]  $S = S_c = (C_{LI} - C_b)/C_{LI}(1 - j)$ . In these equations  $T_b$  and  $C_b$  are the temperature and concentration at infinity, respectively, and  $C_v$  is the specific heat per unit volume of the liquid. In order for Laplace's equation to be valid, it is necessary that

$S_T \ll 1$  and  $S_c \ll 1$ . Since  $S_T \approx \left(\frac{1}{73}\right) \Delta T$ , it is clear that for  $\Delta T < 1$ ,  $S_T \ll 1$ . Using the solution of the diffusion equation, we may rewrite  $S_c$  as  $S_c = (A_c/A_b)(k_L/DL_v)DT$ . Taking  $k_L = 1.33(10^{-3}) \text{ cal cm}^{-1} \text{ deg}^{-1} \text{ sec}^{-1}$ ,  $L_v = 73.4 \text{ cal cm}^{-3}$ ,  $D = 10^{-5} \text{ cm}^2 \text{ sec}^{-1}$ , and  $(A_c/A_b) \approx 1$ , we have  $S_c \approx 1.8 (DT)$ . For  $DT = 0.1$ ,  $S_c \approx 0.2$ , and Laplace's equation is a reasonable approximation. For larger  $(DT)$  and for smaller diffusion constants, there may well be deviations from Laplace's equation.

In the actual experiments the temperature is maintained at  $T_b$  at some radius, say  $R_a$ . If  $R_a < R_\lambda$ , it seems reasonable to take  $R_b = R_a$  and this has been done [5, 6]. If  $R_a > R_\lambda$ , it is better to take  $R_b = R_\lambda$ . If this were not done, i.e., if instead we let  $R_b = R_a$  when  $R_a > R_\lambda$ , then from eq (4a) we would predict that  $\dot{R}$  is larger when  $T_L(\infty) = T_b$  than when  $T_L(R_a) = T_b$ . This is obviously wrong and hence for  $R_a > R_\lambda$  we take  $R_b = R_\lambda$ . When  $R_a < R_\lambda$ , there is a certain error in taking  $R_b = R_a$ , but this error lies within the experimental error in measuring  $\Delta T$  and  $\dot{R}$ . The choice of  $R_c$  is even more complicated since the average concentration in the liquid changes as solute is rejected from the ice. Fortunately for many cases the results are not very sensitive to the choice of  $R_c$  and for calculational purposes we take  $R_c = R_a$ . The proper choice of  $R_c$  and  $R_b$  can be studied experimentally by comparing the observed growth rate  $\dot{R}$  of the unperturbed cylinder with the theoretical  $\dot{R}$ .

We present some calculations of  $\delta$  as a function of  $R$  for the special case where  $(\dot{\delta}R/\delta\dot{R})$  is given by eq (6c). Following Cahn [4] we use the relationship  $(\dot{\delta}R/\delta\dot{R}) = (R/\delta)(d\delta/dR)$ . Denoting the right hand side of eq (6c) by  $f(R)$ , we have

$$\frac{d \ln \delta}{dR} = \frac{f(R)}{R} \quad (7a)$$

Integrating yields

$$\ln(\delta/\delta_0) = \int_{R_0}^R \frac{f(x)}{x} dx, \quad (7b)$$

where  $\delta_0$  is the value of  $\delta$  at  $R = R_0$ . The integral has been evaluated numerically for various values of the parameters occurring in  $f(R)$ . In many cases over small ranges of  $R$ ,  $\ln \delta$  is to a good approximation linear with  $R$ . This is illustrated in table 1 which gives  $(d \ln \delta/dR)$  as a function of  $R$ . From table 1, it is seen that  $(d \ln \delta/dR)$  changes very slowly with  $R$ . Since in the experimental measurements of  $\delta$  versus  $R$ ,  $R$  varies by about 0.02 to 0.03 cm and  $R$  is greater than 0.120 cm, it is to be expected that plots of the experimental data in the form  $\ln \delta$  versus  $R$  will appear linear.



Of interest is the wavelength dependence of the  $\delta$  versus  $R$  curves. This is shown in table 2 for three different wavelengths. We can consider  $\delta/\delta_0$  as an amplification factor, that is, it gives the relative magnitude of  $\delta$  at  $R$  as compared to  $\delta$  at  $R_0$ . As seen from the table, these amplification factors are very large. Experimentally values of  $\delta$  of  $3(10^{-3})$  cm have been observed at  $R \approx 0.175$  cm. Extrapolating to  $R = 0.05$  cm gives a value of  $\delta \approx 10^{-7}$  cm as the size of the initial perturbation. It is clear from table 2 that the amplification factor is a function of wavelength. For small  $R$ , perturbations with  $\lambda = 0.045$  cm are amplified slightly more than perturbations corresponding to the other wavelengths of the table. For larger  $R$ , however, the amplification is greatest for  $\lambda = 0.055$ . Calculations similar to these will be useful in attempting to predict the wavelength of the perturbation which appears on a growing cylinder. However, such a prediction requires some assumption about the initial distribution of the amplitudes of the perturbations of various wavelengths.

We wish to discuss the effect of solute on the stability equations. It is interesting that for no anisotropy and zero surface tension, the addition of solute has no effect on  $(\delta/\delta)/(R/R)$ , i.e., eqs (5b) and (6b) are identical. The addition of solute changes the unperturbed growth rate by changing  $DT$ . For a bath undercooling  $\Delta T = 0.1$ , the value of  $(DT)$  for various values of  $mc_b$  are given in table 3.

Also of interest is the factor  $1 + (m\alpha/\nu)$  appearing in eq (5b). It can be shown that

$$(m\alpha/\nu) \approx (k_L/DL_0)(mc_b)(1 + k_S/k_L) \approx 9 (mc_b).$$

Thus for  $mc_b < 0.1$  deg.,  $1 + (m\alpha/\nu)$  varies from 1 to 2. Since  $DT$  varies more rapidly than this, the main effect of adding solute is to decrease  $DT$  and thus to increase the magnitude of the last term in eq (5b). Thus for fixed  $\Delta T$  adding solute makes  $(\delta/\delta)/(R/R)$  smaller, thus stabilizing the cylinder. On the other hand for fixed  $(DT)$ , which corresponds to not changing the unperturbed velocity  $\dot{R}$ , the addition of solute increases  $(\delta/\delta)/(R/R)$  and consequently makes the cylinder more unstable. The above statements assume that the  $\Gamma_0$  term is the dominant term inside the bracket of eq (5b) and that the addition of solute does not change the physical properties such as the surface tension. If solute is adsorbed at the interface, one expects a lowering of the surface tension. One method of studying such effects is to add a very small amount of solute, e.g.,  $10^{-4}$  M NaCl in which case  $mc_b \approx 4(10^{-4})$ . From the preceding calculations, it is clear that  $DT \approx \Delta T$  and  $1 + (m\alpha/\nu) \approx 1$  so that eqs (6a) and (6b) can be used. Thus any difference between experimental results for distilled water and water containing  $10^{-4}$  M of impurities should probably be attributed to a change in the surface tension due to adsorption at the interface.

In summary, we have analyzed the stability of a solid cylinder growing by heat flow into a binary melt and have taken account of any small anisotropy in the interface properties. In particular, we have provided a theoretical framework for the experimental study of the stability of ice cylinders.

TABLE 1.  $(d \ln \delta/dR)$  as a function of  $R$  for  $\lambda = 0.055$  cm;  $\gamma = 0.018$  J/m<sup>2</sup> and  $\Delta T = 0.1$  deg.

$R(\text{cm})$	$(d \ln \delta/dR) (\text{cm}^{-1})$
0.100	85.8
.110	84.3
.120	82.9
.130	81.7
.140	80.7
.150	79.7
.160	78.9
.170	78.1
.180	77.4
.190	76.8
.200	76.3

TABLE 2. Effect of wavelength on  $\delta/\delta_0$  for  $\gamma = 0.018$  J/m<sup>2</sup> and  $\Delta T = 0.1$  deg.

$R(\text{cm})$	$(\delta/\delta_0)$		
	$\lambda = 0.045$ cm	$\lambda = 0.055$ cm	$\lambda = 0.065$ cm
0.050	1.00	1.00	1.00
.075	1.08(10 <sup>1</sup> )	1.07(10 <sup>1</sup> )	9.88
.100	9.85(10 <sup>1</sup> )	9.70(10 <sup>1</sup> )	8.16(10 <sup>1</sup> )
.125	7.80(10 <sup>2</sup> )	7.91(10 <sup>2</sup> )	6.07(10 <sup>2</sup> )
.150	5.53(10 <sup>3</sup> )	5.99(10 <sup>3</sup> )	4.22(10 <sup>3</sup> )
.175	3.59(10 <sup>4</sup> )	4.28(10 <sup>4</sup> )	2.80(10 <sup>4</sup> )
.200	2.17(10 <sup>5</sup> )	2.93(10 <sup>5</sup> )	1.80(10 <sup>5</sup> )

TABLE 3. Value of effective undercooling  $(DT)$  as a function of added solute for bath undercooling  $\Delta T = 0.1$  °C ( $\xi/A_b = 1.8$  deg<sup>-1</sup> used in the calculation)

$mc_b$	$DT$
0.001	0.0988
.01	.0881
.02	.0768
.04	.0556
.06	.0359
.08	.0174

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