Morphological Stability Analysis of the Self-Similar Solidification Front in the Case of Thermodiffusion.

Part II. The Stability Criterion

I. V. Alexandrova

Laboratory of Multi-Scale Mathematical Modeling, Ural Federal University
Lenin ave., 51, Ekaterinburg, 620000, Russian Federation

I. G. Nizovtseva

Ural Federal University
Office 607, Turgeneva str. 4, Ekaterinburg, 620075, Russia

Abstract

In the current paper series, consisting of two parts, we are treating the morphological stability and performing the analysis for the self-similar solidification of a binary melt. Also we getting new results on graphical studies of stability curves, representing neutral stability curve and the self-similar solution, plotted in accordance with obtained expressions. If the melt thermodiffusion (the temperature dependence of the diffusion coefficient) is negligible, the neutral stability curve for morphological perturbations does not intersect the self-similar branch of solutions for solidification, that is, the regime under consideration is always stable and the constitutional supercooling instead of instability analysis is responsible for the origination of a two-phase zone. In the opposite case, when the melt thermodiffusion is important the latter is responsible for the morphological instability of the planar front. Moreover, this unstable solidification stage occurs before a point of constitutional supercooling. In other words, thermodiffusion effects are responsible for the origination of a two-phase (mushy) layer.

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Keywor
ds: Solidification, Instability, Thermodiffusion

1 Linear analysis of morphological stability

We shall now concentrate on the linear analysis of morphological stability and perturb self-similar temperature and concentration profiles (13) (see Part I for the previous sets of equations), as well as the front position $\Sigma = 0$. In fact it means that because of the various perturbations in the system (e.g., some temperature field small oscillations, ingot mold small mechanical oscillations e.t.c.), the self-similar temperature and concentration profiles get the time and $y$-dependent small additions (as well as the front rate and position $g$) in the following way:

$$T_s' = T_s - T_{ss}, \quad C_s' = C_s - C_{ss}, \quad T_L' = T_L - T_{ls}, \quad C_L' = C_L - C_{ls}, \quad \Sigma = \Sigma'.$$

Taking into account these expressions for perturbations we should get back to Eqs. (9) from Part I and expand the boundary conditions (10)-(12) in Taylor's series at $\eta = 0$. Let us rewrite it only for linear terms in perturbations:

$$(\tau/2) \partial C_s / \partial \tau = \left( \frac{dC_s(T_{ls})}{d\eta} + \frac{1}{2}(\eta + \lambda) \frac{d^2 C_s}{d\eta^2} + D_L(T_{ls}) \frac{d^2 C_s}{d\eta^2} + \tau^2 \frac{\partial^3 C_s}{\partial \eta^3} + \frac{d}{d\eta} \left( D_L(T_{ls}) \frac{dC_s}{d\eta} \right) \right).$$

with similar equations for the perturbations $T_s'$ and $C_s'$, and

$$T_s' = T_L' + H_s(\lambda) \Sigma' = 0, \quad H_s(\lambda) = dT_{ss} / d\eta - dT_{ls} / d\eta, \quad \eta = 0,$$  \hspace{1cm} (19)

$$T_L' - mC_L' + H_s(\lambda) \Sigma' - \Gamma T_m \tau \Sigma' / \partial \Sigma' / \partial \tau = 0, \quad \eta = 0,$$  \hspace{1cm} (20)

$$K_s \partial T_s' / \partial \eta - K_s \partial T_s' / \partial \eta + H_s(\lambda) \Sigma' - (L_\nu / 2) \tau \Sigma' / \partial \Sigma = 0, \quad \eta = 0,$$  \hspace{1cm} (21)

$$C_s' = kC_s' + H_s(\lambda) \Sigma' = 0, \quad \eta = 0.$$  \hspace{1cm} (23)

For the sake of simplicity, we shall introduce the following notations (and do not substitute the self-similar distributions (13):

$$H_s(\lambda) = dT_{ls} / d\eta - m dC_{ls} / d\eta, \quad \eta = 0,$$

$$H_s(\lambda) = K_s d^2 T_{ss} / d\eta^2 - K_s d^2 T_{ls} / d\eta^2 - L_\nu / 2, \quad \eta = 0.$$
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\[ H_s(\lambda) = D_L(T_{LS}) d^2 C_{LS} / d\eta^2 + (1-k)C_{LS} / 2 + (1-k)(\lambda/2)dC_{LS} / d\eta - D_s d^2 C_{SS} / d\eta^2, \quad \eta = 0. \]

\[ H_s(\lambda) = dC_{SS} / d\eta - k dC_{LS} / d\eta, \quad \eta = 0. \]

Here we write down only equations for the temperature and concentration perturbations in the liquid. We shall now concentrate on the neutral stability curve in a plane of any operating parameters for morphological perturbations. In this situation, there are no terms, dependent on \( \tau \), so it can be seen, that all perturbations can depend only on \( y \) as linear functions, i.e. \( T'_y = T_{L1}(\eta) + T_{LS}(\eta) y \), \( T'_y = T_{S1}(\eta) + T_{SS}(\eta) y \), \( C'_y = C_{L1}(\eta) + C_{LS}(\eta) y \), \( C'_y = C_{S1}(\eta) + C_{SS}(\eta) y \) (see Eqs. (18) and their analogs for \( T'_y \) and \( C'_y \)), and boundary conditions at \( \eta = 0 \). Substituting these functions into Eqs. (18) and similar equations for \( T'_y \) and \( C'_y \), we obtain

\[ (\eta + \lambda) dT'_y / d\eta + 2\kappa_L d^2 T'_y / d\eta^2 = 0, \]

\[ (\eta + \lambda) dT'_y / d\eta + 2\kappa_s d^2 T'_y / d\eta^2 = 0, \quad (24) \]

\[ (\eta + \lambda) dC'_y / d\eta + 2D_s d^2 C'_y / d\eta^2 = 0. \]

\[ \left( \frac{dD(T_{LS})}{d\eta} + \frac{1}{2}(\eta + \lambda) \right) \frac{dC'}{d\eta} + D_L(T_{LS}) \frac{d^2 C'}{d\eta^2} + \frac{d}{d\eta} \left( D_L(T_{LS}) \frac{dC_{LS}}{d\eta} \right) = 0. \quad (25) \]

Taking into consideration boundary conditions from the Part I, namely (6) and (7) that imply \( T'_y \rightarrow 0 \) and \( C'_y \rightarrow 0 \) as \( \eta \rightarrow -\lambda \); \( T'_y \rightarrow 0 \) and \( C'_y \rightarrow 0 \) as \( \eta \rightarrow \infty \), we can rewrite the solutions of Eqs. (24), (25) at the neutral stability curve in the following form:

\[ T'_y = h_y \text{erf}[(\eta + \lambda)/\sqrt{4\kappa_L}], \quad C'_y = h_y \text{erf}[(\eta + \lambda)/\sqrt{4\kappa_s}], \quad (26) \]

\[ T'_y = h_y \text{erf}[(\eta + \lambda)/\sqrt{4\kappa_L}], \quad C'_y = h_y \left( 1 - \frac{A(\eta)}{A(\infty)} \right) + h_y \left( \frac{B(\infty)}{A(\infty)} A(\eta) - B(\eta) \right), \quad (27) \]

where

\[ B(\eta) = \int_0^{\infty} \left[ \exp \left( -\frac{1}{2} \frac{x + \lambda}{D_L(T_y(s))} \right) \exp \left( -\frac{1}{2} \frac{x + \lambda}{D_L(T_y(s))} \right) \frac{\partial}{\partial y} \left( D_L(T_y(s)) \frac{dC_{LS}}{dy} \right) dy \right] dz, \]

\[ h_1 = h_1 + h_2 y, \quad h_2 = h_1 + h_2 y, \quad h_3 = h_3 + h_2 y, \quad h_4 = h_4 + h_2 y. \]

In these expressions coefficients \( h_i \) stand for the perturbation amplitudes and coefficients \( h_i \) represent the arbitrary constants.

We shall point out here, that (26) and (27) are the solutions, which satisfy to
the boundary conditions (19)-(23) at the neutral stability curve (nothing depends on \( \tau \)) only in the case if

\[
\Sigma' = h_y = \Sigma_1 + \Sigma_2 y, \tag{28}
\]

where \( \Sigma_1 \) and \( \Sigma_2 \) are constants.

If we now shall regard the set of boundary conditions (19)-(23) together with solutions (26)-(28), it will be revealed, that afore-mentioned coincide with similar ones for dynamic perturbations at the neutral stability curve, when nothing depends on \( y \) and \( \tau \) [1]. The only difference is in the amplitudes \( h_i \ (i=1,2,3,4,5) \), which are the functions of \( y \) in the case of morphological instability, whereas these amplitudes are constants in the case of dynamic instability. But this does not affect to the situation in principle since when substituting the perturbations (26)-(28) into boundary conditions (19)-(23), we got the set of five linear equations for various amplitudes \( h_1, h_2, h_3, h_4 \) and \( h_5 \). When equating to zero the determinant, which consist of the amplitude coefficients, we shall come to the expression, which will describe the neutral stability curve, in other words, nothing depends on the amplitudes \( h_i \).

Taking into account \( T_w(\lambda) \) equating the result to the right hand side of relation (14) (minding \( c_w = kC_{w0} \)), we get a point of intersection of the neutral stability curve and the self-similar solution (of course, only in case if this point exists), writing down the equation in the final form as:

\[
\frac{Z(\lambda)}{\sqrt{\pi}A(x)} \left[ K_L(T_w + mC_{i1} - T_{i1}) \exp\left(-\frac{\lambda^2}{4\kappa_1}\right) \right] \left[ \frac{\lambda}{\sqrt{\pi}K_L} \operatorname{erfc}\left(\frac{\lambda}{2\sqrt{\kappa_L}}\right) - \exp\left(-\frac{\lambda^2}{4\kappa_L}\right) \right] \times
\]

\[
\times \operatorname{erfc}\left(-\frac{\lambda}{2\sqrt{4\kappa_L}}\right) \left[ \pi\kappa_L \operatorname{erfc}\left(\frac{\lambda}{2\sqrt{4\kappa_L}}\right) - \lambda \operatorname{erfc}\left(-\frac{\lambda}{2\sqrt{4\kappa_L}}\right) \right] -
\]

\[
- \exp\left(-\frac{\lambda^2}{4\kappa_L}\right) \left[ \pi \sqrt{\kappa_L \kappa_S} \right] \right] + T_1(\lambda) \right] = m \right] T_2(\lambda) \operatorname{erf}\left(\frac{\lambda}{4D_2}\right) \times
\]

\[
\times \left[ \frac{C_{i1} - C_{i1}}{A(x)D_1(T_1(0))} \frac{\partial D}{\partial T} T_m + mC_{i1} - T_{i1} \right] \exp\left(-\frac{\lambda^2}{4\kappa_L}\right) +
\]

\[
+ \frac{D_3}{\pi} T_3(\lambda) \exp\left(-\frac{\lambda^2}{4D_3}\right) \left[ \frac{C_{i1} - C_{i1}}{A(x)} \left( \frac{1}{D_2} - \frac{k}{D_1(T_1(0))} \right) \right] +
\]

\[
+ \frac{B(x)}{A(x)} \operatorname{erf}\left(\frac{\lambda}{4D_3}\right) \left[ \frac{K_L}{\pi \sqrt{K_L \kappa_L}} \left( T_m + mC_{i1} - T_{i1}\right) \right] \times
\]
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\[ \times \left( \frac{\exp(-\lambda^2/4\kappa_s) \exp(-\lambda^2/4\kappa_s)}{\text{erfc}(\lambda/4\kappa_s)} + \frac{\lambda}{2} \frac{L_v}{\sqrt{\pi\kappa_s}} \exp \left( -\lambda^2/4\kappa_s \right) \right) + \]

\[ + \frac{\lambda}{2} K_L \left( \frac{1}{\kappa_L} - \frac{1}{\kappa_s} \right) \left( T_m + mC_s - T_c \right) \text{erf} \left( \frac{\lambda}{\sqrt{4\kappa_s}} \right) \text{erfc} \left( \frac{\lambda}{\sqrt{4\kappa_s}} \right) \times \]

\[ \times \exp \left( -\lambda^2/4\kappa_s \right) + \frac{L_v}{2\kappa_s} \left( \lambda^2 + 2\kappa_s \right) \text{erf} \left( \frac{\lambda}{\sqrt{4\kappa_s}} \right) \right]. \]  

(29)

Here the following positive functions for \( \lambda > 0 \) are introduced:

\[ T_1(\lambda) = \frac{L_v}{4\kappa_s} \text{erf} \left( \frac{\lambda}{\sqrt{4\kappa_s}} \right) \left( 2\kappa_s + \lambda^2 \right) \text{erfc} \left( -\frac{\lambda}{\sqrt{4\kappa_s}} \right) + 2\lambda \kappa_s \exp \left( -\lambda^2/4\kappa_s \right) / \sqrt{\pi}, \]

\[ T_2(\lambda) = \frac{K_L}{\sqrt{\pi\kappa_s}} \exp \left( -\lambda^2/4\kappa_s \right) \text{erfc} \left( -\frac{\lambda}{\sqrt{4\kappa_s}} \right) + \frac{K_L}{\sqrt{\pi\kappa_s}} \exp \left( -\lambda^2/4\kappa_s \right) \text{erfc} \left( \frac{\lambda}{\sqrt{4\kappa_s}} \right) + \lambda \kappa_s \exp \left( -\lambda^2/4\kappa_s \right) \text{erf} \left( \frac{\lambda}{\sqrt{4\kappa_s}} \right). \]

2 Graphical studies of stability curves

In the case of temperature independent diffusivity (constant diffusion coefficient in the liquid) this equation was analyzed in [2]. This study shows that Eq. (29) has no any roots in the case under consideration for \( \lambda > 0 \). In other words, the neutral stability curve does not intersect the self-similar branch of solutions for solidification (\( \lambda > 0 \)) and the temperature independent diffusivity regime is absolutely (morphologically and dynamically) stable for any possible alloys.

Numerical solutions of Eq. (29) in the case of constant diffusion coefficient are shown in Fig. 1 (thermophysical properties are given in Table 1, regions \( \lambda > \lambda_s \) and \( T_w < T_{wa} \) correspond to the constitutional supercooling regime in accordance with inequalities (16) and (17)). In other words, the constitutional supercooling instead of instability analysis is responsible for the origination of a two-phase zone for the self-similar case with invariable diffusion coefficient.

![Fig. 1: The neutral stability curve (dashed curve) plotted in accordance with Eq. (29) and the self-similar solution (solid curve) plotted in accordance with expressions (14) and (15), \( \partial D/\partial T = 0 \) and \( D_t = 2.07 \cdot 10^{-5} \text{cm}^2/\text{s} \), \( \lambda_s \approx 3.7 \cdot 10^{-4} \text{cm}/\sqrt{\text{s}}, \) \( T_{wa} \approx 474.59 \text{K} \).](image-url)
Table 1. Thermophysical properties of tin-silver alloys

<table>
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<tr>
<th>Property</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Liquid thermal conductivity, $K_L$</td>
<td>0.3</td>
<td>$J/(cm \cdot s \cdot K)$</td>
</tr>
<tr>
<td>Solid thermal conductivity, $K_s$</td>
<td>0.6</td>
<td>$J/(cm \cdot s \cdot K)$</td>
</tr>
<tr>
<td>Liquid thermal diffusivity, $\kappa_L$</td>
<td>0.16</td>
<td>$cm^2/s$</td>
</tr>
<tr>
<td>Solid thermal diffusivity, $\kappa_s$</td>
<td>0.4</td>
<td>$cm^2/s$</td>
</tr>
<tr>
<td>Solid diffusion coefficient, $D_s$</td>
<td>$10^{-9}$</td>
<td>$cm^2/s$</td>
</tr>
<tr>
<td>Melting point of tin, $T_M$</td>
<td>505.1</td>
<td>$K$</td>
</tr>
<tr>
<td>Solute concentration at infinity, $C_{L\infty}$</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Heat of fusion, $L_v$</td>
<td>418</td>
<td>$J/cm^3$</td>
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<tr>
<td>Liquidus slope, $m$</td>
<td>-284</td>
<td>$K/\text{at. frac.}$</td>
</tr>
<tr>
<td>Segregation coefficient, $k$</td>
<td>0.018</td>
<td></td>
</tr>
</tbody>
</table>

A growth of the thermodiffusion influence ($\partial D/\partial T > 0$ in Figs. 2 and 3) makes closer the curves and gives a point of their intersection. Since the region of stability lies above the neutral stability curve (dashed curves), point $B$ formally divides the stable and unstable regimes (point $B$ lies on the left to point $A$ in Fig. 2). Therefore, regions of stability $0 < \lambda < \lambda_A, T_w < T_w(0)$ and instability $\lambda > \lambda_A, T_w < T_w(0)$ in Fig. 2 show that the instability occurs due to the constitutional supercooling whereas regions of stability $0 < \lambda < \lambda_A, T_w < T_w(0)$ and instability $\lambda > \lambda_A, T_w < T_w(0)$ in Fig. 3 show that instability occurs due to the temperature dependent diffusivity ($T_w(0) \approx 476.7K$ for all figures). In other words, the temperature dependent diffusivity (thermodiffusion) can be responsible for the origination of a two-phase zone [3-10].
Fig. 2: The neutral stability curve (dashed curve) plotted in accordance with Eq. (29) and the self-similar solution (solid curve) plotted in accordance with expressions (14) and (15), \( \frac{\partial D}{\partial T} = 1.72 \cdot 10^{-5} \text{cm}^2/\text{K} \cdot \text{s} \) and \( D(T_{\mu}) = 1.87 \cdot 10^{-4} \text{cm}^2/\text{s} \),
\[ \lambda_\alpha \approx 3.2 \cdot 10^{-4} \text{cm}/\sqrt{s}, \quad T_{\text{wa}} \approx 474.72 \text{K}. \]

Fig. 3: The neutral stability curve (dashed curve) plotted in accordance with Eq. (29) and the self-similar solution (solid curve) plotted in accordance with expressions (14) and (15), \( \frac{\partial D}{\partial T} = 1.72 \cdot 10^{-5} \text{cm}^2/\text{K} \cdot \text{s} \) and \( D(T_{\mu}) = 2.07 \cdot 10^{-4} \text{cm}^2/\text{s} \),
\[ \lambda_\alpha \approx 2.79 \cdot 10^{-4} \text{cm}/\sqrt{s}, \quad T_{\text{wa}} \approx 474.88 \text{K}. \]

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