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# Morphological stability of a solid–liquid interface growing in a cylindrical mold

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ABSTRACT

The morphological stability of the planar interface of dilute alloys solidifying in a cylindrical mold is analyzed based on the perturbation model presented by Mullins and Sekerka under the assumption that the interface crosses the mold wall at right angles, to examine the effect of the inside diameter of the mold. When the interface grows in a mold of a larger inside diameter, the stability-instability criterion of the planar interface is coincident with the MS criterion. On the other hand, in a mold of a smaller diameter, the rippled interface is permitted to take a frequency of discrete values (the permitted frequency), and the planar interface grows stably under thermal conditions slightly exceeding the MS criterion. Also, there exists a minimum permitted frequency $\omega_{\min}$, and the critical inside diameter $d_c$ is derived from $\omega_{\min}$. When the alloy solidifies in a mold of an inside diameter less than $d_c$, the interface grows stably under thermal conditions in which the MS model predicts unstable growth of the interface. Moreover, there is a lower limit $d_G$ in $d_c$, and when the alloy solidifies in a mold of an inside diameter less than $d_G$, the interface grows stably even at a zero temperature gradient in the liquid.



## 1. Introduction

Regarding the morphological stability of the solid–liquid interface during the solidification of alloys, Tiller et al. [1] presented a model in which the planar interface becomes unstable when a constitutionally supercooled zone appears in the liquid in front of the interface. Their model (the CS model) explained the fact that the planar interface becomes unstable when the ratio of the temperature gradient to the growth rate reached a critical value proportional to the solute concentration. A decade later, Mullins and Sekerka [2–5] presented a model in which instability of the planar interface occurs when the amplitude of the rippled interface begins to increase with time. Their model (the MS model) not only presented the critical condition at the onset of the instability of the planar interface (the MS criterion), but also clarified the existence of the absolute stability of the planar interface. Until now, most discussions on the morphological stability of the interface have been conducted based on the MS model, under the tacit assumption that the relevant field is so large that the influence of the mold on the stability of the interface is negligible.

In recent years, microelectromechanical systems (MEMS) and microsensors have been developed with the accompanying progress in micromanufacturing technology, which includes the microcasting process [6–9]. In this process, the solid–liquid interface grows in small parts of a micrometric size under the restrictions of the mold walls.

Discussions have been carried out on the morphological stability of solids growing with a fixed shape, and when a solid of spherical shape [10–12] or cylindrical shape [13] grows in molten alloy, a rippled interface does not take a frequency of an arbitrary value but of discrete values dominated by the size of the solid. The value of the frequency, however, changes with increasing solid size, and the planar interface grows stably until the frequency becomes a critical value determined by the solid size. In this case, the critical conditions are dependent on the critical solid size, but are independent of the size of the medium surrounding the solid. In the case of solidification in the mold, on the other hand, the stability of the interface is largely affected by the shape and size of the mold. For instance, when a thin plate of the alloy solidifies [14–16], the critical conditions for stable growth of the planar interface are dependent on the plate thickness. Also, when the alloy directionally solidifies in a cylindrical mold of a small inside diameter, the stability-instability criterion of the interface is expected to be dependent on the inside diameter.

The present work examines the effect of the inside diameter of the mold on the stability of the planar interface growing in a cylindrical mold of a smaller inside diameter, such as in the microcasting at a high aspect ratio [6,9].

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## 2. Assumptions

We analyze the morphological stability of the planar interface of dilute binary alloys growing in a mold of a smaller inside diameter, based on the perturbation model (the MS model) [2–4].

When the alloy solidifies in the mold, the shape of the interface is dependent on the temperature distribution in the mold [17] and is also constrained by the mold wall. The contact angle between the solid–liquid interface and the mold wall is therefore often measured as a function of the mold size, as shown in Fig. 1 [18]. When the alloy solidifies in a mold of a smaller inside diameter, the temperature becomes uniform in the cross section of the alloy, and the interface is strained by the interfacial energy between the solid and the liquid $\gamma_{LS}$, which balances with the interfacial energies of the liquid–mold wall interface $\gamma_{LM}$ and the solid–mold wall interface $\gamma_{SM}$ through Young's equation [19]:

$$\gamma_{LM} = \gamma_{LS}\cos\theta_C + \gamma_{SM} \tag{1}$$

where $\theta_C$ is the contact angle between the solid–liquid interface and the mold wall. Although the contact angle in Fig. 1 was measured for a stationary interface and not for a growing interface, similar results are expected for slowly growing interface. Consequently, postulating that the interface grows slowly in a mold of a smaller inside diameter of less than 0.1 mm, we assume that the solid–liquid interface crosses the mold wall at right angles.

During solidification, convection driven by the temperature distribution occurs in molten alloy and affects the shape of the interface [17]. When the interface grows in a cylindrical mold of a smaller inside diameter, however, the Reynolds number Re in the mold becomes smaller. For example, when molten alloy flows axially in a mold of an inside diameter of 0.1 mm with a velocity of 10 mm/s, Re is calculated to be less than 5 by using the data on the kinematic viscosity of typical molten alloys [20], and the molten alloy behaves as a viscous fluid. Since the temperature is uniform in the cross section of a mold of an inside diameter of less than 0.1 mm, the molten alloy does not flow in the radial direction, but a laminar flow driven by the shrinkage during solidification occurs in the axial direction with a velocity of $\mu V$, where $\mu$ is the solidification shrinkage and $V$ is the growth rate of the interface. The effect of the solidification shrinkage should therefore be considered in the analysis of the temperature and solute concentration during solidification. The solidification shrinkage of the alloy is, however, generally only a few percent [21], and the increment of the growth rate is small. Consequently, we do not consider the effect of the solidification shrinkage in the analysis.

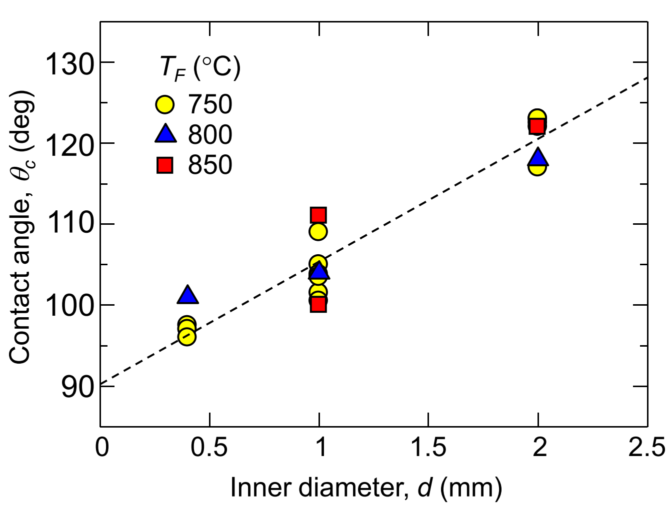


**Fig. 1.** Change in the contact angle $\theta_c$ between the stationary solid–liquid interface of Al–4 mass% Cu alloy and the mold wall with an inside diameter $d$ of the mold, at different furnace temperatures $T_F$.

To analyze the morphological stability of the interface, the shape of the rippled interface should be fixed. In terms of cylindrical coordinates, the shape is uniquely expanded in a series of Bessel functions and a Fourier series in the radial and circumferential directions, respectively. Also, the fluctuation of the interface strained by interfacial tension is analogous to the vibration of the membrane strained by surface tension, and it is known that a circular membrane with the diameter $d$ vibrates with natural frequencies [22] given by

$$z = \sum_{m=0}^{\infty}\sum_{n=1}^{\infty} a_{mn} J_m(\omega_{mn} r)\left[\cos(m\theta) + b_{mn}\sin(m\theta)\right]\left[\cos(\omega_{mn} t) + c_{mn}\sin(\omega_{mn} t)\right] \tag{2}$$

where $z$, $r$ and $\theta$ are the coordinates in the surface normal, the radial and the circumferential directions, respectively, and $t$ is the time. Also $a_{mn}$, $b_{mn}$, $c_{mn}$ and $\omega_{mn}$ are the constants determined by the initial and boundary conditions, $m$ and $n$ are integers determined by the boundary conditions, and $J_m(\omega_{mn} r)$ is the Bessel function of the first kind of order $m$. When the end of the membrane is fixed perpendicular to the surrounding wall, the constant $\omega_{mn}$ takes discrete values satisfying the following equation:

$$mJ_m\left(\frac{\omega_{mn} d}{2}\right) - \frac{\omega_{mn} d}{2} J_{m+1}\left(\frac{\omega_{mn} d}{2}\right) = 0 \tag{3}$$

Substituting $t=0$ into Eq. (2), we obtain the initial shape of the circular membrane.

$$z = \sum_{m=0}^{\infty}\sum_{n=1}^{\infty} a_{mn} J_m(\omega_{mn} r)\left[\cos(m\theta) + b_{mn}\sin(m\theta)\right] \tag{4}$$

When there is no heat flow through the mold wall, the temperature in the mold is uniform in the cross section, and the shape of the interface is formulated as Eq. (4). We then assume that the shape of the rippled interface with an infinitesimal amplitude of $\delta$ is simply given by the expression:

$$\phi = \delta J_m(\omega_{mn} r)\cos(m\theta)\,(m = 0, 1, 2, \ldots, \text{and } n = 1, 2, \cdots) \tag{5}$$

where the constant $\omega_{mn}$ takes discrete values determined by Eq. (3). Eq. (3) is satisfied by $\omega_{mn}=0$ for any vales of $m$, and when $\omega_{mn}=0$, $\phi$ becomes zero or takes a constant value to cause the flat interface, which is conflict with the condition of the rippled interface. We then eliminate $\omega_{mn}=0$ at $n=0$ from the solutions of Eq. (3), and take $\omega_{mn}$ of $m \geq 0$ and $n \geq 1$ as the positive solutions of Eq. (3).

Additionally, even when $\cos(m\theta)$ is replaced by $\sin(m\theta)$ in Eq. (5), the same results are obtained by subsequent calculations. Also, when the interface makes angles differing from the right angles with the mold wall, the analysis is conducted similarly to the present work along with replacement of the right side of Eq. (3) with $\tan(\psi)$, where $\psi$ is the angle that the interface makes with the normal of the mold wall.

## 3. Analysis

### 3.1. Amplitude of the rippled interface

We postulate that the solid–liquid interface grows steadily in a cylindrical mold of an inside diameter $d$ with a constant velocity $V$ in the $z$ direction, as shown in Fig. 2. Also, it is assumed that the heat is transferred by conduction but there is no heat flow through the mold wall, and the solute is transferred in the liquid by diffusion but there is no diffusion in the solid. Accordingly, the