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INFLUENCE OF SOLIDIFICATION ON SURFACE TENSION DRIVEN CONVECTION

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Abstract – The effect of solidification on the onset of surface tension driven convection in a reduced gravity environment is studied. Two simple but physically realistic configurations representing the solidification of a simple material are analyzed. The analysis shows that as a result of the solidification process the critical Marangoni number is shifted to lower values indicating that solidification has a destabilizing effect upon the liquid. From this result it is concluded that convection can be brought about in the liquid phase at lower Marangoni numbers when solidification is present than when it is not. The effects of other parameters introduced by the solidification process are analyzed and discussed.

NOMENCLATURE

A_i,	constants of integration;
A_m,	constant defined in equation (4.5);
a_0,	complex time constant, $= a_{0r} + ia_{0i}$;
B,	Biot modulus, $= qd/k_l$;
b,	$\partial\sigma/\partial T$;
C,	specific heat ratio, $= c_{p_l}/c_{p_s}$;
d,	liquid layer thickness;
D,	differential operator, $= \dfrac{\mathrm{d}}{\mathrm{d}z}$;
K,	thermal conductivity ratio, $= k_l/k_s$;
k,	thermal conductivity;
L,	latent heat of fusion;
M,	Marangoni number, $= (\partial\sigma/\partial T)(\partial T/\partial z)\dfrac{d^2}{\mu_l\kappa_l}$;
Pr,	Prandtl number, $= \nu_l/\kappa_l$;
Q,	$= q_0 d/k_l(T_2 - T_m)$;
q,	heat-transfer coefficient defined in equation (3.5b);
q_0,	heat flux at the lower surface of the solid for the second model;
R,	$= v_p d/\nu_l$;
S,	the position of the solid–liquid interface;
T,	temperature;
T_m,	temperature of solidification;
T_1,	temperature of the lower surface of the solid;
T_2,	temperature of the upper surface of the liquid;
t,	time;
$\mathbf{u}$,	velocity vector;
v,	perturbation velocity;
v_p,	speed of the solid–liquid interface;
w,	perturbation velocity component in the z-direction;
W,	basic flow velocity component in the z-direction.

Greek symbols

α,	perturbation wave number;
β,	non-dimensional temperature gradient of the basic flow for the first model;
Δ,	thermal diffusivity ratio κ_s/κ_l;
δ,	density ratio ρ_l/ρ_s;
Λ,	$= L/c_{p_s}(T_2 - T_1)$ for the first model; $= L/c_{p_s}(T_2 - T_m)$ for the second model;
λ_i,	characteristic values of the solution in equations (4.4) and (4.5);
λ_m,	defined in equation (4.5);
μ_l,	dynamic viscosity of melt;
ν_l,	kinematic viscosity of melt $= \mu_l/\rho_l$;
Σ,	$= (T_2 - T_m)(\partial\sigma/\partial T)/\mu_l v_p$;
σ,	coefficient of surface tension;
σ_0,	a constant value of surface tension;
θ,	perturbation temperature.

Superscripts

$'$,	perturbation quantity.

Subscripts

l,	liquid phase;
s,	solid phase.

1. INTRODUCTION

A GREAT deal of interest has recently been generated in the possibility of producing new materials in the reduced gravity environment that will be provided during the forthcoming missions of Spacelab [1, 2]. The range of possibilities extends from producing large crystals of uniform properties to manufacturing materials with unique properties. Most of these processes involve solidifying materials from the liquid state. Convective motion within the liquid during solidification can influence the local material composition and the shape of the solid–liquid interface and can result in solids with nonuniform properties and crystal defects. The microgravity environment of Spacelab is being viewed as one in which the buoyancy forces are eliminated so that convection driven by thermal gradients will not occur. It is hoped that this will lead to an improved solidification process. However, convection may occur for other reasons and whether convection is negligible or not during solidification constitutes a vital question bearing on the value of future material processing in a microgravity environment. Little information exists presently on convection during solidification under such circumstances.

Although buoyancy driven convection may be suppressed in the microgravity environment through the reduced Rayleigh number, Marangoni type convection may take place [3]. This latter type of convection arises when nonuniformities in the surface tension are present in a fluid which is subjected to a normal temperature gradient [4, 5]. Since most solidification processes take place in molten zones with strong temperature gradients, it is speculated that Marangoni flow will be of great importance in microgravity experiments. In the work reported here we will investigate the effect of solidification at reduced gravity on the onset of Marangoni convection in the liquid phase.

The analysis of the solidification problem in its entirety is quite complex. The formulation of the problem in its simplest general form leads to a nonlinear system of equations with moving boundaries, which makes it quite intractable [6]. Some of the recent advances towards the solution of this problem may be found in the books of Rubenstein [7], Ockendon and Hodgkins [8], and Wilson *et al.* [9]. Since interest in the present work is in the conditions for the onset of convection, the total solidification problem will not be considered but rather a stability analysis will be performed on a material system undergoing solidification.

One of the earliest investigations involving stability analysis for a solidifying system was that of Mullins and Sekerka [10] in which the stability of the solid–liquid interface was studied for a binary alloy. This work was followed by the study of Wollkind and Segel [11], in which the same problem was examined using a nonlinear stability analysis. The mean state of both of these analyses was one-dimensional with both the liquid and solid phases extending to infinity. In addition, these works were not concerned with the convective motion in either the mean or the perturbed states. However, the analyses were of considerable value in establishing the stability criteria for the interface in terms of the concentration and temperature gradients of the mean state.

Schubert *et al.* [12] applied linear stability analysis to a system of two liquids of infinite depth which were undergoing a phase change. The convective motion caused by buoyancy forces was examined for this configuration. However, little information is available on the effect of the coupling between phase change and surface tension driven convection upon the stability of the liquid phase.

These previous studies have examined a system of infinite depth. However, in some space processing applications the melt layer can be expected to be relatively shallow and a slightly different analysis is necessary. In such circumstances, and in the absence of gravity, it is anticipated that surface tension effects at the top of the melt can influence the convection in the melt, and this may in turn affect the stability and shape of the solid–liquid interface. It should be emphasized that in the absence of gravity the melt layer need not be extremely shallow for the above-mentioned coupling to take place. In real physical situations, however, more complicated interactions between the various dynamic and thermodynamic processes, such as chemical reactions, mass diffusion and time dependence of the basic state, can influence the shape of the interface. For an up-to-date review of and exposition of the various influences on the stability of interfaces, the reader is referred to the excellent article by Miller [13] and the references contained therein. In the present work the coupling between surface tension effects and the solidification process is studied as a first attempt at understanding some of the underlying problems to be encountered in space processing of materials.

In this study the stability of two simple solidifying systems which reflect physically realistic configurations is analyzed at zero gravity. The systems considered are solid–liquid systems of finite depth for a pure material which is undergoing a phase transition. The possibility of the onset of convection in the liquid phase is considered. Two configurations are analyzed. In the stationary process the phase boundary is stationary while in the constant rate process it is propagating at a constant speed. The details of the two models together with the stability analysis are presented in the next two sections. This is followed by a discussion of the results in Section 4. Finally, in the last section, a set of conclusions is given together with some recommendations that might be of benefit for the design and analysis of low gravity materials processing experiments.

2. THE BASIC STATE

Solidification is basically a time-dependent process for a system of finite depth because of the movement of the solid–liquid interface. An analysis of the stability of such a time-dependent process is presently in the state of development. Therefore, it was necessary to apply the analysis to two special models which were stationary or could be made stationary by a coordinate transformation. In this section the stationary mean state, which will be perturbed, is presented for each model.

For the first model consider a layer of fluid on top of a layer of solid in which the solid–liquid interface is stationary (stationary solidification process). Both the solid and the liquid layers are infinite in the horizontal direction but are of finite depth. A sketch of this model is shown in Fig. 1. Since the solid–liquid interface is stationary in this case, the solidification process, through the latent heat, does not influence the mean state but will play a significant role in the perturbed state. Such a model may represent an extremely slow solidification process which might be encountered in actual crystal growth experiments.

The steady state temperature distribution in both the solid and the liquid phases, in the absence of any convection, can be shown to be a linear function of depth and is given by [14]:

$$\bar{T}_l = T_m - (T_m - T_1)/K + \frac{z}{S+d}\{T_2 - T_1/K - T_m(1 - 1/K)\}, \quad (2.1)$$

$$\bar{T}_s = \left\{T_m(K-1) + T_1 + \frac{z}{S+d}[KT_2 - T_1 - T_m(K-1)]\right\}/K. \quad (2.2)$$

This solution was obtained by assuming constant temperatures at both the upper surface of the liquid and the lower surface of the solid, T_2 and T_1, respectively. Also, the solid–liquid interface position is uniquely determined by the boundary conditions and is given by:

$$S = \frac{d(T_m - T_1)}{K(T_2 - T_m)}; \quad T_2 > T_m > T_1. \quad (2.3)$$

Note that for this model the thermodynamic and transport properties of both the solid and the liquid phases were taken to be constant.

The second model (constant rate solidification) also consists of two adjacent, finite depth layers of solid and melt which are infinite in the horizontal direction and separated by a planar interface. However, in this case the solid–liquid interface is taken to propagate at a constant speed, v_p, in the vertical direction while melt is being continuously fed to the system to keep the melt depth constant. A sketch of this problem is given in Fig. 2. The upper surface of the liquid is maintained at a constant temperature T_2 while the phase interface is at the liquid–solid equilibrium temperature T_m. This model may represent a continuous solidification process which is being cooled at a constant rate, q_0, from the lower surface of the solid. From a balance of mass and energy across the interface, the velocity in the liquid, which is in the direction of propagation of the interface, is found to be:

$$\bar{W}_l = v_p(1/\delta - 1). \quad (2.4)$$

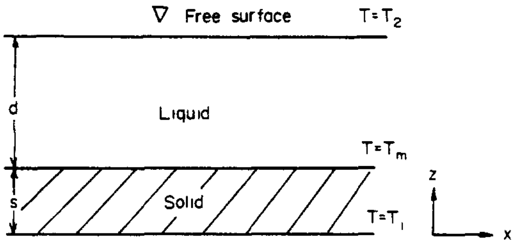

FIG. 1. Description sketch of the stationary solidification problem.

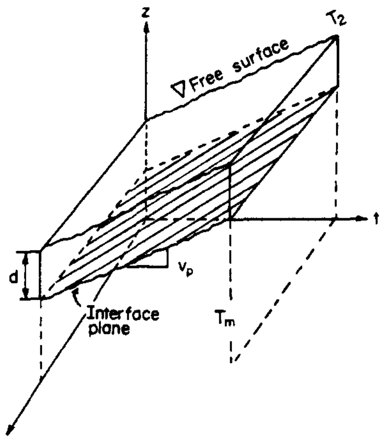

FIG. 2. Description sketch of the constant rate solidification problem.

In this problem the thermodynamic properties of both the solid and the liquid are taken to be different but constant throughout each layer.

The temperature distributions in both the solid and liquid phases, subject to a constant temperature at the upper surface of the melt and a constant rate of heat loss at the lower surface of the solid, are given by [14]:

$$\bar{T}_l = T_m + \frac{(T_m - T_2)\{\exp(-v_p z/\delta\kappa_l) - 1\}}{1 - \exp(-v_p d/\delta\kappa_l)} \quad (2.5)$$

$$\bar{T}_s = T_m + (\kappa_s q_0/v_p k_s) \times \exp(-v_p d/\kappa_s)\{1 - \exp(-v_p z/\kappa_s)\}. \quad (2.6)$$

The instantaneous position of the solid–liquid interface may be obtained from integrating the interface velocity, v_p, which is given by the following transcendental equation:

$$q_{0s}\exp(-v_p d/\kappa_s) + (k_l v_p/\delta\kappa_l)(T_2 - T_m) \times \{1 - \exp(-v_p d/\delta\kappa_l)\}^{-1} = \rho_s L v_p. \quad (2.7)$$

Note that in this problem the solidification process enters both the mean state and the perturbed state. The above solutions are obtained after transforming the problem to a moving coordinate system attached to the planar solid–liquid interface via the transformation $x = \tilde{x}$, $z = \tilde{z} - v_p t$ and $t = \tilde{t}$, where the tilde indicate the original stationary coordinate system.

3. LINEAR STABILITY ANALYSIS

A. *Stationary solidification process*

The conditions for the onset of convection in the liquid phase are determined through a linear stability analysis. Briefly, a perturbation is imposed on the original mean state and the fate of this perturbation with time is observed. If the perturbation decays with time then the original state is said to be stable, and if, on the other hand, it grows with time then the original state is unstable and convective motion will set in. To implement this analysis, the field variables are first decomposed into a mean and a perturbation component in the following form:

$$\begin{aligned} T_s &= \bar{T}_s + T'_s, \\ T_l &= \bar{T}_l + T'_l, \\ \mathbf{u} &= \bar{\mathbf{u}} + \mathbf{u}'. \end{aligned} \tag{3.1}$$

These variables are then substituted into the conservation equations for mass, momentum and energy. Upon subtracting out the equations for the mean state, equations governing the perturbation functions are obtained. For a linear stability analysis these equations are linearized resulting in the following set of equations governing the perturbation functions for the stationary solidification problem:

$$\left[\frac{\partial}{\partial t} - (\nu_l/\kappa_l)\nabla^2\right]\nabla^2 w' = 0, \tag{3.2}$$

$$\left(\frac{\partial}{\partial t} - \nabla^2\right)T'_l = -w'\frac{\mathrm{d}\bar{T}_l}{\mathrm{d}z}, \tag{3.3}$$

$$\left[\frac{\partial}{\partial t} - (\kappa_s/\kappa_l)\nabla^2\right]T'_s = 0, \tag{3.4}$$

where

$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}.$$

In equation (3.3) $\bar{T}_l$ is the mean state temperature distribution which is given by expression (2.1). Also, the above equations have been made dimensionless with the following length, time, velocity and temperature scales, respectively: d, κ_l/d^2, d/κ_l, $T_2 - T_1$.

The boundary conditions for the perturbation functions may be obtained in a similar manner and for the present problem the conditions at the free surface of the liquid are:

$$k_l\frac{\partial T'_l}{\partial z} = -qT'_l\,; \quad w' = 0, \tag{3.5a, b}$$

$$b\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)T'_l = \mu\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - \frac{\partial^2}{\partial z^2}\right)w'. \tag{3.5c}$$

Condition (3.5c) is derived from the balance of the tangential forces at the free surface of the liquid where it is assumed that the surface tension is a linear function of temperature, i.e.

$$\sigma = \sigma_0 + bT'_l.$$

Condition (3.5a) is the so-called "radiation condition" which expresses the perturbation of the balance of heat conducted across the liquid with that convected away from the surface of the liquid. The conditions at the solid–liquid interface are:

$$T'_l = T'_s = 0\,; \quad \frac{\partial w'}{\partial z} = 0, \tag{3.5d, e, f}$$

$$k_s\frac{\partial T'_s}{\partial z} - k_l\frac{\partial T'_l}{\partial z} = -\rho_l L w'. \tag{3.5g}$$

Condition (3.5g) is derived from a linearization of the energy balance at the interface with the added assumption that the interface geometry is planar to a first order approximation (see [14] for further details on the derivation of this condition). The final condition needed for the complete description of the problem must be applied at the lower surface of the solid and is given by:

$$T'_s = 0. \tag{3.5h}$$

The system of equations (3.2)–(3.4), together with the boundary conditions (3.5), constitutes a set of linear, homogeneous equations which admit a solution through separation of variables in the form:

$$[w', T'_x, T'_l] = [v(z), \theta_s(z), \theta_l(z)]\cos\alpha x\,\exp(a_0 t). \tag{3.6}$$

This form of the solution represents a wave which has an amplitude in the z-direction and a wave number, α, while a_0 is a constant. Such a form of the solution allows an arbitrary disturbance to be analyzed into a complete set of normal modes whereby the stability of each individual mode can be examined separately. For a general description it is a common practice to take a_0 to be a complex number, i.e.

$$a_0 = a_{0r} + ia_{0i}.$$

Upon substituting this form of the solution (3.6) into the governing equations and the boundary conditions, a system of linear, homogeneous, ordinary differential equations with homogeneous boundary conditions results. Such a system constitutes an eigenvalue problem for the eigenvalue a_0. The solution of such a problem is quite straightforward and the question of the stability or instability of the system is settled through the determination of the sign of a_{0r}. To do this the principle of "exchange of stability" was invoked by assuming that a_{0i} is zero whenever a_{0r} is zero. With this assumption the system of equations governing the neutral ($a_0 = 0$) perturbation function takes the following form:

$$(D^2 - \alpha^2)^3\theta_l = 0, \tag{3.7}$$

$$(D^2 - \alpha^2)\theta_s = 0. \tag{3.8}$$

Similarly, the boundary conditions at the liquid free surface take the following form:

$$D\theta_l + B\theta_l = 0\,; \quad (D^2 - \alpha^2)\theta_l = 0, \tag{3.9a, b}$$

$$(D^4 - \alpha^4)\theta_l - M\alpha^2\theta_l = 0. \tag{3.9c}$$

The conditions at the solid–liquid interface are given by:

$$\theta_l = \theta_s = 0\,; \quad D(D^2 - \alpha^2)\theta_l = 0, \tag{3.9d, e, f}$$

$$D\theta_s = KD\theta_l - \left(\frac{\delta\Lambda}{\Delta\beta}\right)(D^2 - \alpha^2)\theta_l = 0, \tag{3.9g}$$

and the condition at the lower solid surface is

$$\theta_s = 0. \qquad (3.9h)$$

The results of the solution of these equations are given in Section 4.

B. *Constant rate solidification*

The stability analysis and general formulation of the perturbation problem for this case follows along the same line as was developed for the previous case. For this problem the mean flow velocity is not zero, as it was for the first case, and has a component in the z-direction given by expression (2.4). Thus, again substituting (3.6) into the equations of mass, momentum and energy, subtracting the mean state, and linearizing the resulting equations yields the following system of non-dimensional equations for the perturbation functions:

$$\left[\frac{\partial}{\partial t} - (1/\delta)\frac{\partial}{\partial z} - (\nu_l/v_p d)\nabla^2\right]\nabla^2 w' = 0, \qquad (3.10)$$

$$\left[\frac{\partial}{\partial t} - (1/\delta)\frac{\partial}{\partial z} - (\kappa_l/v_p d)\nabla^2\right]T_l' = -w'\frac{\mathrm{d}\bar{T}_l}{\mathrm{d}z}, \qquad (3.11)$$

$$\left[\frac{\partial}{\partial t} - \frac{\partial}{\partial z} - (\kappa_s/v_p d)\nabla^2\right]T_s' = 0. \qquad (3.12)$$

In this case the equations are written in terms of a coordinate system which is fixed with the mean position of the solid–liquid interface. Also, all of the variables in the above equations have been made dimensionless using the following scales of length, time, velocity and temperature: d, v_p/d, v_p, and $T_2 - T_m$, respectively. Note that the temperature gradient of the mean state for this case, $\bar{T}_l$, can be obtained from equation (2.5).

The boundary conditions for the present case in the new reference frame are identical with conditions (3.5).

Following the same reasoning which led to the system (3.7)–(3.9), the following system of equations for the neutral perturbation amplitude functions for the second model are obtained:

$$[(1/\delta)D + (1/R)(D^2 - \alpha^2)](D^2 - \alpha^2)v = 0, \qquad (3.13)$$

$$[(1/\delta)D + (1/PrR)(D^2 - \alpha^2)]\theta_l = v\frac{\mathrm{d}\bar{T}_l}{\mathrm{d}z}, \qquad (3.14)$$

$$[D + (\Delta/PrR)(D^2 - \alpha^2)]\theta_s = 0, \qquad (3.15)$$

with the boundary conditions given by

$$Dv = 0; \quad \theta_l = \theta_s = 0, \qquad (3.16\text{a, b, c})$$

$$D\theta_l - (\Lambda\delta PrC)v = 0, \qquad (3.16\text{d})$$

at the solid–liquid interface, and

$$D\theta_l + B\theta_l = 0; \quad v = 0, \qquad (3.16\text{e, f})$$

$$(D^2 + \alpha^2)v - \Sigma\alpha^2\theta_l = 0, \qquad (3.16\text{g})$$

as the liquid-free surface, and

$$\theta_s = 0, \qquad (3.16\text{h})$$

at the solid lower surface.

4. RESULTS AND DISCUSSION

A. *Stationary process*

The solution to equations (3.7) and (3.8) can be immediately written in the form:

$$\theta_l = \sum_{i=0}^{2} z^i[A_i \sinh(\alpha z) + A_{i+3}\cosh(\alpha z)], \qquad (4.1)$$

$$\theta_s = A_6 \sinh(\alpha z) + A_7 \cosh(\alpha z), \qquad (4.2)$$

where the eight constants of integration A_i can be evaluated from the eight boundary conditions (3.9). However, the coefficients need not be determined explicitly since in the present study it is only desired to determine whether the original state is stable with respect to small perturbations or not. Instead, the common practice followed in stability analysis is adopted here, and the regions of stability and instability in terms of the parameters involved in each problem are identified. This is implemented by solving the algebraic equation arising from setting the determinant of the coefficients of the constants of integration to zero, which results in an equation for the neutral stability curve in terms of the parameters involved. Figure 3 shows several of the neutral stability curves for this problem, where the stable region lies to the left of each curve and the unstable region to the right.

For the stationary process the dispersion relation for the neutral stability curve can be written in terms of the Marangoni number, M, in the form:

$$M = f(\alpha, \beta, B, \Lambda, \Delta, K), \qquad (4.3)$$

where α is the perturbation wave number, B is the Biot modulus, β is the non-dimensional temperature gradient across the liquid, and Λ is the non-dimensional form of the latent heat of fusion. Δ and K are the ratios of the thermal diffusivity and thermal conductivity of the liquid and solid phases, respectively. Δ, K, and Λ are usually fixed by the choice of the material used and

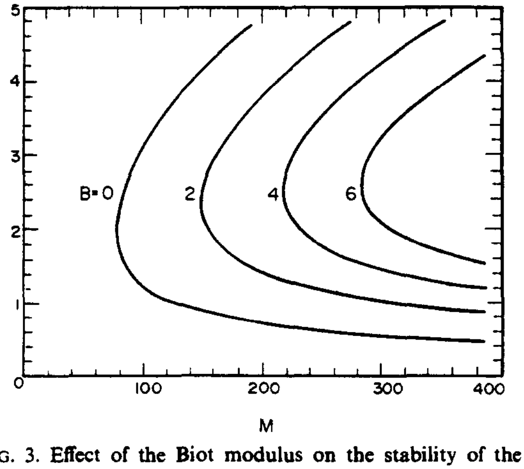

FIG. 3. Effect of the Biot modulus on the stability of the stationary process. $\Lambda = 20$, $\beta = 1.2$, $K = 0.7$ and $\Delta = 1.5$.

are assumed to be constants. Thus, for any given material the only variable parameters are β and B. The exact functional form of (4.3) is quite lengthy and is given in the Appendix.

In order to gain insight into the influence of the various parameters on the onset of convection, the variation of the neutral stability curve with respect to changes in the parameter values was studied. The variation of M with α for various values of B is shown in Fig. 3 for a value of Λ of 20. The qualitative and quantitative form of these curves is almost identical to those of Pearson [4]. It is shown in Fig. 3 that increasing the heat transfer rate through the upper surface (increasing B) stabilizes the liquid for a given value of Λ. Also, the wave number of the most unstable wave increases slightly with increasing B.

Of major interest is the effect of Λ, the non-dimensional latent heat of fusion, on the extent of the stability regions. As Λ is decreased the value of the critical Marangoni number (smallest value of M for instability) for all of these curves shifts toward lower values of M indicating a smaller stable region. This variation of the critical Marangoni number with Λ is shown in Fig. 4. However, as illustrated in that figure, M_{crit} deviates from its large Λ asymptotic value only at reasonably small values of Λ. For small temperature gradients the value of Λ for sodium, for instance, is of the order 100 (see Table 1) and would be small only for large temperature gradients. Therefore, the influence of solidification upon the destabilization process for the stationary process, i.e. the deviation from the calculations of Pearson [4], is small for moderate temperature gradients but is a destabilizing effect as the temperature gradient is increased. Note that the present boundary conditions correspond to the conducting case of Pearson. Also, as Λ becomes large the liquid velocity becomes small [see equation (3.5g)] and the results are expected to approach those for a non-solidifying liquid, as can be seen in Fig. 4.

Since Λ is the most important parameter for this study, the effect of the remaining parameters on the neutral stability curves for the stationary process will not be shown here. Their influence can be easily calculated for any given material.

B. *Constant rate process*

The solution to equations (3.13)–(3.16) for the constant rate solidification problem can also be written in a straightforward manner. First, the perturbation velocity is given by:

$$v = \sum_{i=1}^{4} A_i \exp(\lambda_i z), \tag{4.4}$$

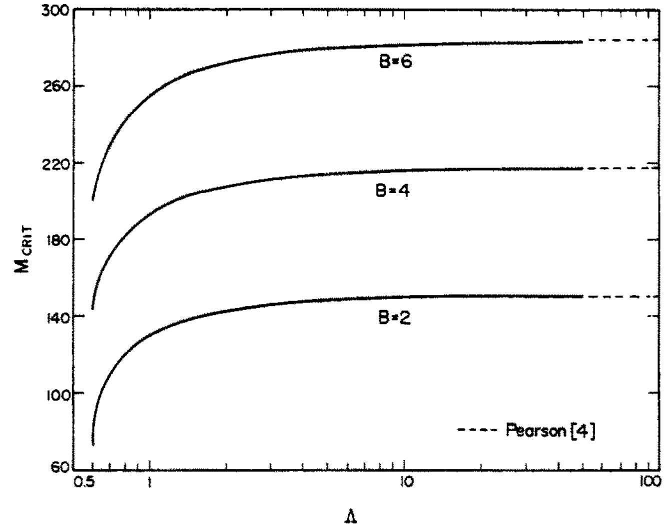

FIG. 4. Effect of the latent heat on the stability of the stationary process. $\beta = 1.2$, $K = 0.7$ and $\Delta = 1.5$.

Table 1. Values of non-dimensional parameters

Material	$Pr(@T)$†	Λ	K‡	Δ‡	P‡	C‡
Sodium	0.0107 (100)	$83.1/\Delta T$†	0.730	1.344	0.975	1.012
Potassium	0.0085 (100)	$73.5/\Delta T$	0.511	3.050	0.975	1.011
Bismuth	0.0171 (300)	$400/\Delta T$	1.74	0.715	1.034	1.20
Lead	0.0206 (400)	$174/\Delta T$	0.437	2.20	0.965	1.038
Tin	0.016 (240)	$93/\Delta T$	0.476	2.32	0.975	1.13
Zinc	0.0275 (450)	$262/\Delta T$	0.502	2.41	0.935	1.29
Water	13.6 (0)	$159/\Delta T$	0.255	9.41	1.090	2.008

†Temperatures measured in °C.

‡Properties determined at T_m^* for sodium and potassium and at 25°C for the solid phase for other materials.

where

$$\lambda_{1.2} = \pm\alpha,$$

$$\lambda_{3.4} = -\tfrac{1}{2}(R/\delta) \pm \tfrac{1}{2}\{(R/\delta)^2 + 4\alpha^2\}^{1/2}.$$

Consequently, the perturbation temperature takes the form:

$$\theta_l = \sum_{i=1}^{4} A_i C_i \exp[(\lambda_m + \lambda_i)z] + \sum_{i=5}^{6} A_i \exp(\lambda_i z), \tag{4.5}$$

where

$$\lambda_{5.6} = -(PrR/2\delta) \pm \tfrac{1}{2}\{(PrR2\delta)^2 + 4\alpha^2\}^{1/2},$$

and

$$C_i = A_m\{(\lambda_m + \lambda_i)/\delta + (1/PrR) \times [(\lambda_m + \lambda_i)^2 - \alpha^2]\}^{-1}; \quad i = 1, \ldots, 4,$$

$$A_m = \lambda_m\{1 - \exp(-\lambda_m)\}^{-1},$$

$$\lambda_m = PrR/\delta.$$

The perturbation temperature in the solid, on the other hand takes the form:

$$\theta_s = A_7 \exp(\alpha z) + A_8 \exp(-\alpha z). \tag{4.6}$$

Again the various coefficients, A_i, can be evaluated through the boundary conditions (3.16). The neutral stability curves were obtained in the manner outlined earlier. For this problem the functional relationship between the various parameters for the neutral stability curve takes the following form:

$$\Sigma = f(\alpha, \delta, B, R, Pr, Q, \Lambda, \Delta, C), \tag{4.7}$$

where Σ is a surface tension parameter which is different from the Marangoni number, because the mean state temperature distribution in the liquid for this model is not linear. However, if a mean temperature gradient defined by

$$\frac{\mathrm{d}\bar{T}_l}{\mathrm{d}z} = (T_2 - T_m)/d,$$

is introduced and used to define a Marangoni number, M, then the relationship between Σ and M is given by:

$$\Sigma = M/PrR. \tag{4.8}$$

However, since the parameter Σ appears naturally in this problem, it will be used in the discussion of the results.

The extra parameters that are involved in the neutral stability curve for the constant rate process and which were not involved in the stationary process are: R, the non-dimensional form of the solid–liquid interface speed; Pr, the Prandtl number of the liquid; Q, a non-dimensional form of the heat withdrawal rate from the lower solid surface, and δ and C which are the ratios of the densities and the specific heats of the liquid and solid phases, respectively. Since the speed of the interface is a function of the heat withdrawal rate from the solid, then Q and R are not independent and are related by the nonlinear matching condition, equation (2.7). However, the relationship between them can be made simpler if it is assumed that the interface speed is slow, i.e. $R \approx O(1)$. With the further assumptions that $Pr \approx O(10^{-2})$ and $\Delta \approx O(1)$, which are true for liquid metals, the relationship may be linearized, taking the form:

$$R = (\delta C/Pr)(Q + 1)(QK + \Lambda)^{-1}. \tag{4.9}$$

All of the results that are discussed through the rest of this section have been obtained using this simplified form. Again, the specific form of equation (4.7) may be found in the Appendix.

The effect of the various parameters upon the stability of the constant rate process will now be examined. First consider the influence of the rate of heat transfer through the lower surface of the solid, which is specified by the parameter Q. Increasing Q through q_0, which increases the solidification rate, leads to a lower critical value of Σ. Typical neutral stability curves for this case are shown in Fig. 5. The values of Q used correspond to very small values of q_0,

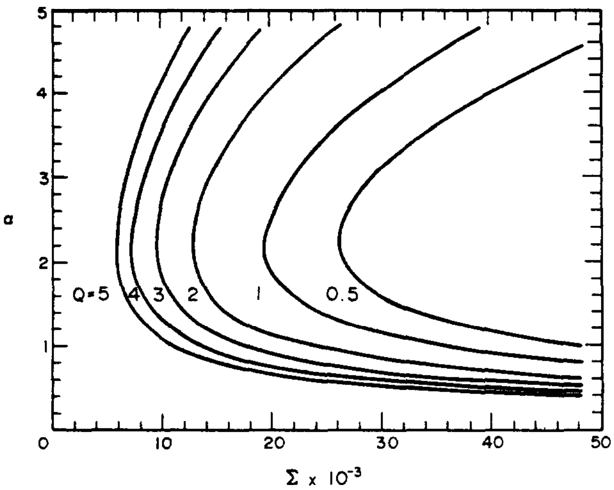

FIG. 5. Effect of the rate of heat transfer through the solid on the stability of the constant rate process. $Pr = 0.02$, $K = 0.5$, $\delta = 0.965$, $C = 1.04$, $\Lambda = 350$ and $B = 1.0$.

typically of the order 0.1–0.5 J/s cm^2. It is interesting to note that the wave numbers at the critical values of Σ are approximately constant and equal to 2.0–2.2. This corresponds to a most critical wave length of about πd.

On the other hand, increasing the rate of heat transfer through the upper liquid surface, i.e. increasing B, stabilizes the flow (Fig. 6). This result is identical to that found in the first model and also obtained by Pearson [3] for a non-solidifying liquid. Note that values of B can be varied by changing either the depth of the liquid or the heat-transfer coefficient q and its variation has no effect on the solidification rate R.

Λ, the latent heat parameter, may be changed by changing L/c_{p_s} for constant $(T_2 - T_m)$ or vice versa. Different materials will have different values of L/c_{p_s}, as shown in Table 1, and this will result in different values of Λ for a given $(T_2 - T_m)$. The effect of the change of Λ through change of L/c_{p_s} upon $\Sigma_{\rm crit}$ is shown in Fig. 7. Materials with higher values of L/c_{p_s} are more stable. Note that R, the non-dimensional rate of solidification, decreases as Λ is increased or Q is decreased [equation (4.9)].

Changing Λ by changing $(T_2 - T_m)$ results also in a change in Q. In general, the effect is similar to that shown in Fig. 7. On the whole, the effect of changing Λ for this model is in the same direction as that in the first model, namely, lower values of Λ lead to smaller regions of stability, as shown in Fig. 8. The value of the critical Marangoni number approaches that for no solidification for large values of Λ, even though the comparison is not easily inferred since the present problem involves extra parameters. The effect of solidification is much greater for the constant rate process than for the stationary process and indicates that solidification has a strong destabilizing influence in this case.

The parameters Pr, K, δ and C are constant for a given material. However, the effect of these parameters on the stability of this configuration was examined so that comparison could be made of the relative stability of different materials. The Prandtl number can vary over many orders of magnitude depending upon the material (Table 1). Because R is inversely proportional to Pr, calculations were performed at a constant $R = 1.0$ by varying Λ simultaneously with Pr. The variation of the critical value of Σ with Pr for this case is shown in Fig. 9. However, the variation of the critical Marangoni number with Pr is quite small, as summarized in Table 2.

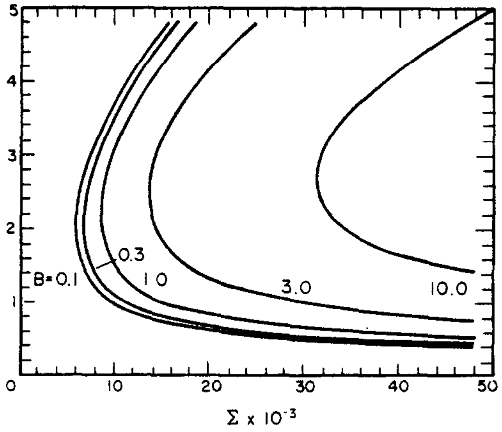

FIG. 6. Effect of the Biot modulus on the stability of the constant rate process. $Pr = 0.01$, $K = 0.5$, $\delta = 0.975$, $C = 1.01$, $\Lambda = 175$ and $Q = 1.0$.

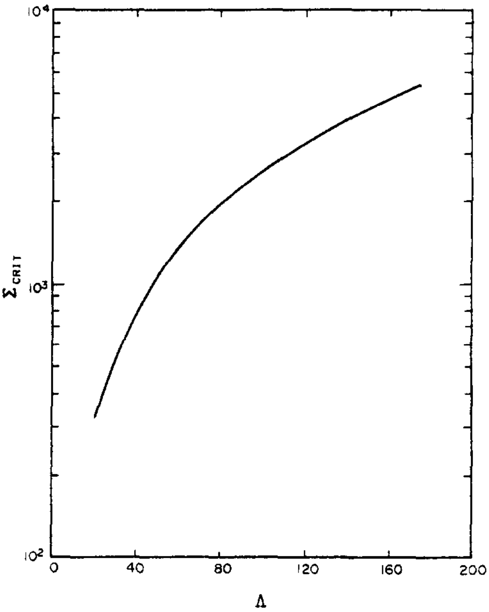

FIG. 7. Effect of the latent heat parameter on the stability of the constant rate process ($T_2 - T_m$ constant). $Pr = 0.01$, $Q = 2.0$, $K = 0.7$, $\delta = 1.0$, $C = 1.0$ and $B = 1.0$.

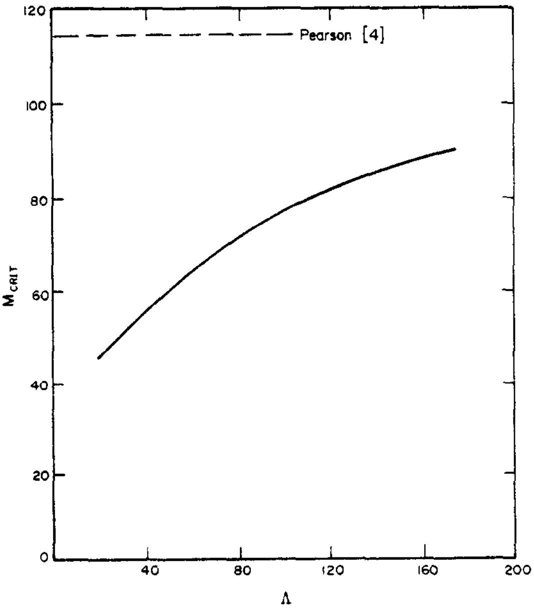

FIG. 8. Effect of latent heat parameters on the critical Marangoni number for the constant rate process. $Pr = 0.01$, $K = 0.7$, $Q = 2.0$, $\delta = 1.0$, $C = 1.0$ and $B = 1.0$.

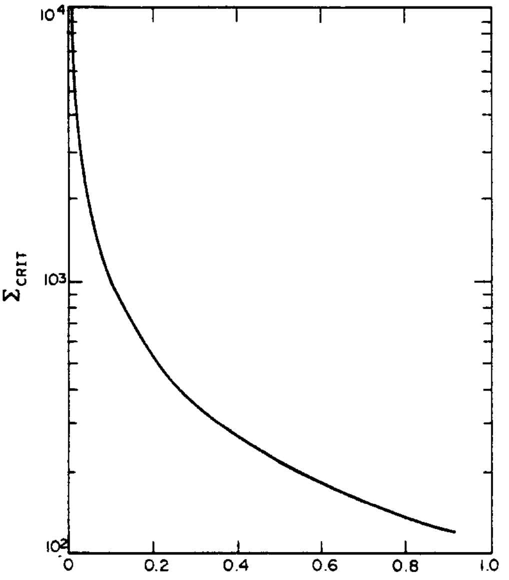

FIG. 9. Effect of the Prandtl number on the stability of the constant rate process. $R = 1.0$, $Q = 1.0$, $K = 0.7$, $\delta = 1.0$, $C = 1.0$ and $B = 1.0$.

Table 2. Effect of Pr on stability for $R = 1.0$; $Q = 1.0$; $B = 1.0$; $K = 1.0$; $P = 1.0$; $C = 1.0$

Pr	Λ	Σ_{crit}	M_{crit}	$\alpha(\Sigma_{crit})$
0.002	1000	4.96×10^4	99.2	2.2
0.01	200	9.96×10^3	99.6	2.2
0.1	20	1.05×10^3	101.8	2.2
0.3	6.0	3.59×10^2	107.7	2.2
0.6	2.7	2.00×10^2	120.0	2.1
0.9	1.5	1.45×10^2	130.5	1.9

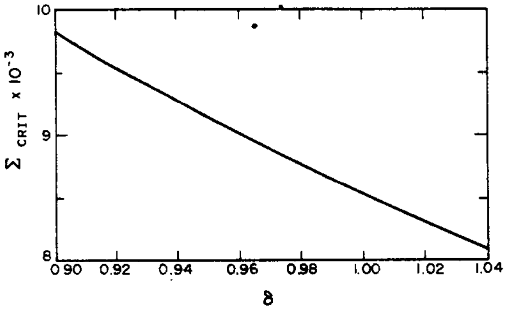

FIG. 10. Effect of the density change on the stability of the constant rate process. $Pr = 0.01$, $\Lambda = 175$, $Q = 1.0$, $K = 0.7$, $B = 1.0$ and $R = 1.1$.

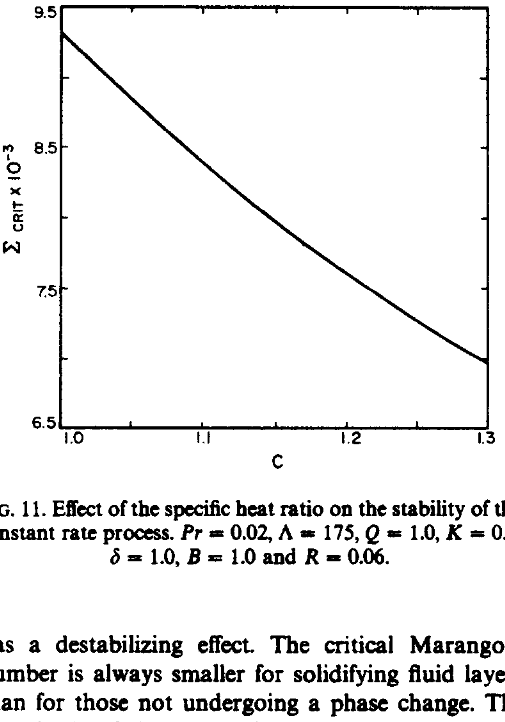

FIG. 11. Effect of the specific heat ratio on the stability of the constant rate process. $Pr = 0.02$, $\Lambda = 175$, $Q = 1.0$, $K = 0.5$, $\delta = 1.0$, $B = 1.0$ and $R = 0.06$.

The effect of density and specific heat ratios, δ and C, upon the critical value of Σ is shown in Figs. 10 and 11. The greater the increase in the density of the material upon solidification, i.e. the lower the value of δ, the more stable is the flow. Materials such as bismuth and water with values of $\delta > 1$ are somewhat less stable than materials with $\delta < 1$. The same conclusion can be drawn for increasing the specific heat ratio, C. K, the ratio of the coefficients of thermal conductivity, can be shown to have no effect on the stability of the system.

It should be noted that the Marangoni number for the most single component systems known to the authors will be negative for the problems discussed. This apparent discrepancy with previous work has been discussed by Seki *et al.* [15].

5. CONCLUDING REMARKS

Some general conclusions concerning the possibility for onset of convective motion in a planar solidifying system with a vertical temperature gradient at zero gravity can be drawn from the analysis of the two models discussed above. The most important conclusion is that solidification, through the parameter Λ, has a destabilizing effect. The critical Marangoni number is always smaller for solidifying fluid layers than for those not undergoing a phase change. The magnitude of the change in the critical Marangoni number depends upon the configuration of the solidifying system. This conclusion indicates the importance of carefully designing reduced gravity experiments to provide adequate isothermal conditions so that convection driven by surface tension will not occur.

In addition, it can be concluded that increasing the solidification rate by increasing the heat-transfer rate through the lower solid surface (increasing q_0) destabilizes the flow. On the other hand, increasing the perturbation heat-transfer rate through the upper surface of the liquid (increasing B) stabilizes the flow. In almost all of the cases which were investigated, it was found that the wave number of the most unstable disturbance is about 2.0–2.2, and this wave number is only a function of B.

Materials with larger values of density and specific heat ratios, δ and C, are less stable than those with lower ratios. The Prandtl number, however, has a small effect on the critical Marangoni number for a constant R. On the other hand, the thermal conductivity ratio K has no effect on the stability of the flow.

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REFERENCES

1. H. Massey, E. G. C. Burt, J. A. Champion, J. W. Christian and R. F. Rissone, Materials behavior in low gravity conditions, *Proc. R. Soc.* **A361**, 129 (1978).
2. Committee on Scientific and Technological Aspects of Materials Processing in Space, *Materials Processing in Space*, National Academy of Sciences, Washington, D.C. (1978).
3. T. C. Bannister, P. G. Grodzka, L. W. Spradley, S. V. Bourgeois, R. O. Hedden and B. R. Facemire, Apollo 17 heat flow and convection experiments final data analyses results, NASA TM-X-64772 (1973).
4. J. R. A. Pearson, On convection cells induced by surface tension, *J. Fluid Mech.* **4**, 489 (1958).
5. L. E. Scriven and C. V. Sterling, On cellular convection driven by surface tension gradients: effects of mean surface tension and surface viscosity, *J. Fluid Mech.* **19**, 321 (1964).
6. H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids*. Clarendon Press, Oxford (1959).
7. L. I. Rubenstein, *The Stefan Problem, Trans. Math. Monographs*. **27**, Am. Math. Soc., (1971).
8. J. R. Ockendon and W. R. Hodgkins, *Moving Boundary Problems in Heat Flow and Diffusion*, Oxford University Press (1975).
9. D. G. Wilson, A. D. Solomon and P. T. Bogs, *Moving Boundary Problems*. Academic Press, New York (1978).
10. W. W. Mullins and R. F. Sekerka, Stability of planar interface during solidification of a dilute binary alloy, *J. Appl. Phys.* **35**, 444 (1964).
11. D. J. Wollkind and L. A. Segel, A nonlinear stability analysis of the freezing of a dilute binary alloy, *Phil. Trans. R. Soc.* **268A**, 351 (1970).
12. G. Schubert, D. A. Yuen and D. L. Turcotte, Role of phase transition in a dynamic mantle, *Geophys. Jl R. Astronaut. Soc.* **42**, 705 (1975).
13. C. A. Miller, Stability of interfaces, *Surface Colloid Sci.* **10**, 227–293 (1978).
14. B. N. Antar and F. G. Collins, Studies of convection in a solidifying system with surface tension at reduced gravity, NASA CR-3051 (1978).
15. N. Seki, S. Fukusako and M. Sugawara, A criterion of onset of free convection in a horizontal melted water layer with free surface, *J. Heat Trans.* **99**, 92 (1977).

APPENDIX

Exact Form of the Dispersion Relations

The exact functional form of the dispersion relation for the stationary process stability calculations, equation (4.3), is:

$$M = 8\left\{\frac{D_3 \cosh\alpha - \alpha D_2/3}{D_2\,(\coth\alpha - \alpha/3) - D_1}\right\} \tag{A1}$$

where D_1, D_2 and D_3 are the following determinants:

$$D_1 = \begin{vmatrix} \begin{array}{l}(1 + B - \alpha^2/3)C + \\ (\alpha/3 - \alpha B/3)S\end{array} & S & 2SC - \alpha \\ 4\alpha(S - \alpha C)/3 & 2\alpha C & 2C + 4\alpha S^2 \\ K & \dfrac{2\delta\Lambda\alpha}{\beta\Delta} - K\alpha & \dfrac{2\delta\Lambda}{\beta\Delta}S - K\alpha C \end{vmatrix} \tag{A2}$$

$$D_2 = \begin{vmatrix} \alpha C + BS & S & 2SC - \alpha \\ 0 & 2\alpha C & 2SC + 4\alpha S^2 \\ \alpha K & \dfrac{2\delta\Lambda}{\beta\Delta}\alpha - K\alpha & \dfrac{2\delta\Lambda}{\beta\Delta}S - K\alpha C \end{vmatrix} \tag{A3}$$

$$D_3 = \begin{vmatrix} \alpha C + BS & \begin{array}{l}(1 + B - \alpha^2/3)C + \\ (\alpha/3 - \alpha B/3)S\end{array} & S \\ 0 & 4\alpha(S - \alpha C)/3 & 2\alpha C \\ K\alpha & K & \dfrac{2\delta\Lambda}{\beta\Delta} - K\alpha \end{vmatrix} \tag{A4}$$

where

$$C = \cosh\alpha; \quad S = \sinh\alpha.$$

The functional form of the dispersion relation for the constant rate process, equation (4.7), is given here in a slightly different form from that of equation (4.7). The exact form of this relation can be written as

$$\det[A] = 0$$

where $[A]$ is a 6×6 matrix, whose individual coefficients are the following:

$$\left.\begin{aligned}
a_{1i} &= C_i \\
a_{2i} &= \lambda_i \\
a_{3i} &= \exp(\lambda_i) \\
a_{4i} &= (\lambda_m + \lambda_i)C_i \\
a_{5i} &= (\lambda_m + \lambda_i + B)C_i \exp(\lambda_m + \lambda_i) \\
a_{6i} &= \{\lambda_i^2 + \alpha^2 - \Sigma\alpha^2 C_i \exp(\lambda_m)\}\lambda_i
\end{aligned}\right\} \quad (i = 1, \ldots, 4)$$

$$a_{15} = a_{16} = 1$$

$$a_{25} = a_{26} = a_{35} = a_{36} = 0$$

$$a_{45} = \lambda_5$$

$$a_{46} = \lambda_6$$

$$a_{55} = \lambda_5 + B$$

$$a_{56} = \lambda_6 + B$$

$$a_{65} = -\Sigma\alpha^2 \exp(\lambda_5)$$

$$a_{66} = -\Sigma\alpha^2 \exp(\lambda_6)$$

where C_i, λ_i and λ_m are defined in Section 4(B).

INFLUENCE DE LA SOLIDIFICATION SUR LA TENSION INTERFACIALE GOUVERNEE PAR LA CONVECTION

Résumé — On étudie l'effet de la solidification sur la tension interfaciale gouvernée par un environnement à gravité réduite. On analyse deux configurations simples mais réalistes représentant la solidification d'un matériau simple. L'analyse montre qu'en conséquence du mécanisme de solidification, le nombre de Marangoni critique atteint des valeurs faibles, ce qui indique que la solidification a un effet déstabilisant sur le liquide. On en conclut que la convection peut apparaitre dans la phase liquide à des nombres de Marangoni plus faibles quand la solidification est présente qu'en son absence. On discute et on analyse les effets des autres paramètres introduits dans le mécanisme de la solification.

DER EINFLUß DES ERSTARRUNGSVORGANGS AUF DURCH OBERFLÄCHENSPANNUNG BEDINGTE KONVEKTION

Zusammenfassung — Der Einfluß des Erstarrungsvorgangs auf das Einsetzen von durch Oberflächenspannung verursachter Konvektion wird in einer Umgebung mit verminderter Schwerkraft untersucht. Zwei einfache, jedoch physikalisch wirklichkeitsnahe Anordnungen, die das Erstarren eines einfachen Materials wiedergeben, werden behandelt. Die Untersuchung zeigt, daß die kritische Marangoni-Zahl als Folge des Erstarrungsvorgangs zu niedrigeren Werten hin verschoben wird und daß das Erstarren eine destabilisierende Wirkung auf die Flüssigkeit hat. Aus diesem Ergebnis wird geschlossen, daß in der flüssigen Phase die Konvektion, wenn Erstarrung stattfindet, bei niedrigeren Marangoni-Zahlen ausgelöst wird, als wenn dieses nicht der Fall ist. Die Einflüsse anderer Parameter des Erstarrungsvorgangs werden untersucht und besprochen.

ВЛИЯНИЕ ЗАТВЕРДЕВАНИЯ НА КОНВЕКЦИЮ, ВЫЗЫВАЕМУЮ ПОВЕРХНОСТНЫМ НАТЯЖЕНИЕМ

Аннотация — Исследуется влияние затвердевания на возникновение конвекции, обусловленной поверхностным натяжением, в условиях ослабленной гравитации. Анализируются две простые, но физически реалистические конфигурации, представляющие затвердевание простых материалов. Анализ показывает, что в результате процесса затвердевания критическое значение числа Марангони смещается в сторону меньших значений, свидетельствуя о дестабилизирующем влиянии затвердевания на жидкость. Из этого следует, что при наличии затвердевания конвекция в жидкой фазе может возникать при более меньших значениях числа Марангони, чем при его отсутствии. Анализируется и обсуждается влияние других параметров, появляющихся в процессе затвердевания.