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INSTABILITY OF A SOLIDIFYING BINARY MIXTURE

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## ABSTRACT

### INSTABILITY OF A SOLIDIFYING BINARY MIXTURE

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An analysis is performed on the stability of a solidifying binary mixture due to surface tension variation of the free liquid surface. The basic state solution is obtained numerically as a non-stationary function of time. Due to the time dependence of the basic state, the stability analysis is of the global type which utilizes a variational technique. Also due to the fact that the basic state is a complex function of both space and time, the stability analysis is performed through numerical means.

## INTRODUCTION

A great deal of interest has been generated recently in the possibility of producing new materials in the reduced gravity environment provided during the forthcoming missions of Spacelab. The range of possibilities extend from producing large crystals of uniform properties to manufacturing materials with unique properties. Most of these processes involve the solidification of materials from the liquid state. Convective motions within the liquid during solidification can influence the local material composition and the shape of the solid-liquid interface which may result in solids with non-uniform 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 materials processing in low-gravity environment. Little information exists presently on convection during solidification under such circumstances.

The work reported here is a continuation of an analytical investigation into the nature of convective motion in a binary liquid layer due to surface tension forces during its solidification. The onset of convection will be determined through a stability analysis which is described below.

## STATEMENT OF THE PROBLEM

The occurrence of convective motions in a fluid has been studied both theoretically and experimentally for approximately a century. The problem is very well documented in several books and numerous articles with all of its variations. It is obvious that in order to study analytically the detailed convective motions in a fluid in any configuration requires nothing less than the total solution of the Navier-Stokes equations and Energy Conservation equations. This is well known to be a formidable, although not an impossible, task requiring considerable amounts of time as well as financial resources.

In certain applications, it is sufficient to be able to know whether or not under certain conditions a fluid could sustain convective motions. The answer to such a question requires far less work; but, again, the information is essentially just of a binary form. Such information can be obtained through hydrodynamic stability analysis. Essentially one introduces perturbation into a well-known basic state and studies the evolutions of these perturbations in time.

If the perturbations decay with time, then the basic state is said to be stable and no convective motion will ensue. If, on the other hand, the perturbations are found to grow with time, then the basic state is said to be unstable and convective motion will take place. Fundamental to such a stability analysis is the existence of a stationary basic state. Unfortunately, the problem under consideration does not have such a basic. However, it has been shown that it is possible to carry out a meaningful stability analysis of such a basic state through several techniques. We have chosen the energy stability method as the best suited for the problem under consideration. This technique is elucidated in detail in the monographs of Joseph [1].

In this work, we consider the stability of a binary fluid layer which is being solidified from below and has an upper free surface. The fluid layer is assumed to be of infinite extent in the horizontal direction. Since it is assumed that the process is being carried out in the low gravity environment of Spacelab, it is anticipated that the driving force is the surface tension force at the free surface. The solution to the basic state has already been obtained in a previous report (Antar [2]). As is expected, the basic state is a function of both time and space. Thus, the stability analysis used must account for the variations of the basic state with time.

The stability analysis starts with the perturbation equations which may be written in the following form

$$\frac{\partial \underline{u}}{\partial t} + \underline{u} \cdot \nabla \underline{u} + \underline{U} \cdot \nabla \underline{u} + \underline{u} \cdot \nabla \underline{U} = -\frac{1}{\rho_0} \nabla p + \nu \nabla^2 \underline{u} + g[\beta_1 \theta + \beta_2 \gamma] \hat{k} \quad (1)$$

$$\frac{\partial \gamma}{\partial t} + \underline{u} \cdot \nabla \gamma + \underline{U} \cdot \nabla \gamma + \underline{u} \cdot \nabla C = D \nabla^2 \gamma \quad (2)$$

$$\frac{\partial \theta}{\partial t} + \underline{u} \cdot \nabla \theta + \underline{U} \cdot \nabla \theta + \underline{u} \cdot \nabla T = \kappa \nabla^2 \theta \quad (3)$$

where  $\underline{u}$  ( $u, v, w$ ) is the perturbation velocity vector,  $\underline{U}$  ( $U, V, W$ ) is the basic state velocity vector.  $\gamma$  and  $C$  are the perturbation and basic state concentration and  $\theta$  and  $T$  are the perturbation and basic state temperature, respectively.  $\nu$ ,  $D$  and  $\kappa$  are the kinematic viscosity, thermal diffusivity and the solutal diffusion coefficient, respectively. These equations are for a Boussinesque fluid and subject to the following boundary condition

$$\underline{u} = \theta = \gamma = 0$$

on the lower surface.

While at the upper surface, we must have the following boundary conditions:

$$k \frac{\partial \theta}{\partial z} = -\tau \theta \quad (4a)$$

$$\frac{\partial \gamma}{\partial z} = 0 \quad (4b)$$

$$\mu \left( \frac{\partial w}{\partial x} + \frac{\partial u}{\partial z} \right) = \frac{\partial \sigma}{\partial x} \quad (4c)$$

$$\mu \left( \frac{\partial w}{\partial y} + \frac{\partial v}{\partial z} \right) = \frac{\partial \sigma}{\partial y} \quad (4d)$$

where  $\sigma$  is the surface tension. On all the sidewalls we must have the perturbation function vanishing.

The governing equations are first non-dimensionalized using the fluid depth  $d$  for a length scale,  $d/\kappa$  for a time scale,  $\kappa/d$  for a velocity scale,  $T_1 - T_0$  and  $C_1 - C_0$  for temperature and concentration scales respectively. Upon introducing these scales into equations (1) - (4) we get the following equations for the perturbations functions

$$\frac{\partial u}{\partial t} + u \cdot \nabla u + v \cdot \nabla u + w \cdot \nabla u = -\nabla p + \nabla^2 u + P(R\theta + LR_c \gamma) \quad (5)$$

$$\frac{\partial \theta}{\partial t} + u \cdot \nabla \theta + v \cdot \nabla \theta + w \cdot \nabla \theta = \nabla^2 \theta \quad (6)$$

$$\frac{\partial \gamma}{\partial t} + u \cdot \nabla \gamma + v \cdot \nabla \gamma + w \cdot \nabla \gamma = L \nabla^2 \gamma \quad (7)$$

with the following boundary conditions

$$\theta = \psi = \gamma = 0 \quad (8)$$

on all side except the upper surface and

$$\frac{\partial \theta}{\partial z} + \beta \theta = 0 \quad (9a)$$

$$\frac{\partial \gamma}{\partial z} = 0 \quad (9b)$$

$$\frac{\partial \psi}{\partial z} + M \frac{\partial \theta}{\partial x} + M_c L \frac{\partial \gamma}{\partial x} = 0 \quad (9c)$$

$$\frac{\partial \psi}{\partial z} + M \frac{\partial \theta}{\partial y} + M_c L \frac{\partial \gamma}{\partial y} = 0 \quad (9d)$$

at the upper surface. In these equations we have the following non-dimensional numbers.

$$P = \text{Prandtl No.} = \nu / \kappa$$

$$R = \text{Rayleigh No.} = \beta \cdot g \Delta T d^3 / \nu \kappa$$

$$R_c = \text{Solutal Rayleigh No.} = g \beta_c \Delta C d^3 / \nu D$$

$$L = \text{Lewis No.} = D / \kappa$$

$$B = \text{Biot modulus} = \tau d / \kappa$$

$$M = \text{Marangoni No.} = (-\partial \sigma / \partial T) \Delta T d / \mu \kappa$$

$$M_c = \text{Solutal Marangoni No.} = (-\partial \sigma / \partial C) \Delta C d / \mu D$$

where  $\partial \sigma / \partial T$  and  $\partial \sigma / \partial C$  are the constants from the surface tension variations with both temperature and concentrations.

Now to obtain the energy equations we first take the dot product equation (5) with  $u$  and integrate about the volume under consideration to get

$$\begin{aligned} \frac{d}{dt} \int_V \rho^{-1} \rho'^2/2 dV = & - \int_V \nabla \underline{u} : \nabla \underline{u} dV + R \int_V \omega \theta dV + L R_c \int_V \omega \gamma dV \\ & - M \int_{z=1} \theta \frac{\partial \omega}{\partial z} dx dy - M_c L \int_{z=1} \gamma \frac{\partial \omega}{\partial z} dx dy \end{aligned} \quad (10)$$

Similarly it is possible to obtain equations for  $\theta'^2/2$  and  $\gamma'^2/2$  in the following integral form

$$\frac{d}{dt} \int_V \theta'^2/2 dV = - \int_V \omega \theta \frac{\partial T}{\partial z} dV - B \int_{z=1} \theta^2 dx dy - \int_V \nabla \theta \cdot \nabla \theta dV \quad (11)$$

$$\frac{d}{dt} \int_V \gamma'^2/2 dV = - \int_V \omega \gamma \frac{\partial C}{\partial z} dV - L \int_V \nabla \gamma \cdot \nabla \gamma dV \quad (12)$$

Now multiplying equation (11) by  $\lambda_\theta$  and equation (12) by  $\lambda_\gamma$  and adding equations (10) - (12) we get

$$\begin{aligned} \frac{d}{dt} K = & - \Delta + \int_V [(R - \lambda_\theta \frac{\partial T}{\partial z}) \omega \theta + (R_c L - \lambda_\gamma \frac{\partial C}{\partial z}) \omega \gamma] dV \\ & - \int_{z=1} [M \theta + M_c L \gamma] \frac{\partial \omega}{\partial z} dx dy \end{aligned} \quad (13)$$

where

$$K = \frac{1}{2} \int_V (P^{-1} \dot{q}^2 + \lambda_\theta \theta^2 + \lambda_r r^2) dV$$

$$D = \int_V [\nabla \underline{u} : \nabla \underline{u} + \lambda_\theta \nabla \theta \cdot \nabla \theta + \lambda_r \nabla r \cdot \nabla r] dV + B \int_{z=1} \lambda_\theta \theta^2 dx dy$$

Equations (13) can be cast in a symmetric form with the following change of variables

$$\theta' = \sqrt{\lambda_\theta} \theta \quad ; \quad r' = \sqrt{\lambda_r} r$$

$$\lambda_\theta = M \mu_\theta \quad ; \quad \lambda_r = M_c \mu_r$$

$$R = M N \quad ; \quad R_c = M_c N_c$$

which after dropping the primes takes the following forms:

$$\frac{dK}{dt} = -D + \sqrt{M} \Pi \quad (14)$$

where

$$K = \frac{1}{2} \int_V (P^{-1} \dot{q}^2 + \theta^2 + r^2) dV$$

$$D = \int_V (\nabla \underline{u} : \nabla \underline{u} + \nabla \theta \cdot \nabla \theta + \nabla r \cdot \nabla r) dV + B \int_{z=1} \theta^2 dx dy$$

$$\begin{aligned} \Pi = & \alpha_\theta \mu_\theta^{-1/2} \left( \int_0^r c_\theta \omega \theta dr - \int_{z=1} \theta \frac{\partial \omega}{\partial z} dx dy \right) \\ & + \alpha_f \mu_f^{-1/2} \left( \int_0^r c_f \omega_f dr - \int_{z=1} r \frac{\partial \omega}{\partial z} dx dy \right), \end{aligned}$$

and

$$\alpha_\theta = \sqrt{\frac{M}{M}}$$

$$\alpha_f = \sqrt{\frac{M_c}{M}}$$

$$c_\theta = N - \mu_\theta \frac{\partial T}{\partial z}$$

$$c_f = L N_c - \mu_f \frac{\partial C}{\partial z}$$

$$M = M + M_c$$

Now let us focus our attention on Eq. (14). Since then, the energy of the perturbation will increase or decrease with time depending on whether  $\sqrt{M} \Pi / D \geq 0$ . Of course the direct and only way of gaining this information is by solving the integro-differential system explicitly. However Eq. (14) can be written as an inequality in the following form

$$\frac{1}{D} \frac{dK}{dt} = -1 + \sqrt{M} \frac{\Pi}{D} \leq -1 + \frac{\sqrt{M}}{R} \quad (15)$$

where

$$e^{-1} = \max_{\pi} \{ \pi \} \quad (16a)$$

with

$$D = 1 \quad (16b)$$

where now the problem is cast as a variational problem defined by (15) and (16).

Let us introduce the lagrange multipliers  $M_u(t)$  and  $p(x, y, z, t)$  into the variational problem (15) - (16) with the following optimization procedure resulting

$$\delta \left\{ \pi + \int \frac{\partial p}{\partial M_u} (\nabla \cdot u) dV - \frac{1}{M_u} D \right\} = 0 \quad (17)$$

From variational calculus we know that the solution to (17) is the same as the solution of the Euler-Lagrange equation resulting from (17). For the present case, the Euler-Lagrange system of equations are the following

$$\frac{\partial p}{\partial x} - \nabla^2 u = 0 \quad (18)$$

$$\frac{\partial p}{\partial y} - \nabla^2 v = 0 \quad (19)$$

$$\frac{\alpha_B}{\sqrt{M_B}} \frac{M_u}{z} C_B \theta + \frac{\alpha_A}{\sqrt{M_A}} \frac{M_u}{z} C_A \Gamma - \frac{\partial p}{\partial z} + \nabla^2 w = 0 \quad (20)$$

$$\frac{M_u}{z} \frac{\alpha_B}{\sqrt{M_B}} C_B w + \nabla^2 \theta = 0 \quad (21)$$

$$\frac{M_A}{z} \frac{\alpha_F}{\sqrt{M_F}} C_T w + \nabla^2 \gamma = 0 \quad (22)$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0 \quad (23)$$

with the following boundary conditions

$$\frac{\partial u}{\partial z} + \frac{1}{z} M_A \frac{\alpha_\theta}{\sqrt{M_\theta}} \frac{\partial \theta}{\partial x} + \frac{1}{z} M_A \frac{\alpha_\delta}{\sqrt{M_\delta}} \frac{\partial \delta}{\partial x} = 0 \quad (24a)$$

$$\frac{\partial v}{\partial z} + \frac{1}{z} M_A \frac{\alpha_\theta}{\sqrt{M_\theta}} \frac{\partial \theta}{\partial y} + \frac{1}{z} M_A \frac{\alpha_\delta}{\sqrt{M_\delta}} \frac{\partial \delta}{\partial y} = 0 \quad (24b)$$

$$B\theta + \frac{\partial \theta}{\partial z} + \frac{1}{z} M_A \frac{\alpha_\theta}{\sqrt{M_\theta}} \frac{\partial w}{\partial z} = 0 \quad (24c)$$

$$\frac{\partial \delta}{\partial z} + \frac{1}{z} M_A \frac{\alpha_\delta}{\sqrt{M_\delta}} \frac{\partial w}{\partial z} = 0 \quad (24d)$$

at  $z = 1$  and

$$u = v = w = \theta = \delta = 0 \quad (24e)$$

at  $z = 0$ .

Now as discussed earlier since the basic state itself a function of time then the term II in the energy equation (14) is a function of time. Also, the lagrange multipliers and the term in (16a) are also function of time. Thus, the variational problems (15) - (16) must be solved at each instant of time which requires the solution of the Euler-Lagrange equations at each point in time. Thus, the procedure is to solve the system (18)-(24) at each instant of time and obtaining the optimization parameter  $M \mu (\tau)$  as a function of time.

## REFERENCES

1. Joseph, D. D., Stability of Fluid Motions I and II. Springer Tracts in Natural Philosophy, Vol. 27 and 28, Springer Verlag, Berlin Heidelberg New York, 1976.
2. Antar, B. N., "Solidification of a Binary Mixture" in Research Reports - 1981 NASA/ASEE Summer Faculty Fellowship Program. Edited by A. Karr, et al., NASA CR - 161855, 1982.