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Compositionally convective and morphological instabilities of a binary fluid layer under freezing with nonlinear analysis

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Abstract—The instabilities of a fluid layer of a binary alloy, cooled from above and consequently frozen at bottom, are considered. The released light material at the freezing interface is diffused by pressure and composition gradients. As a result of a small cooling rate and a large thermal diffusivity, the thermal effect is inefficient, compared with the compositional one, for driving a possible convection. Cellular convective modes of long and short wavelengths, requiring $1 < R < 1+S+qa^2/Q$, and morphological mode of short wavelength, requiring $R > 1+S+qa^2/Q$, are found. As Schmidt number $P_L \to \infty$, the instabilities set in stationarily at the marginal state. Nonlinear analysis of cellular convective modes of long wavelengths shows that finite amplitudes of disturbances just past the marginal state behave like $(R-R_c)^{1/2}$. Subcritical instabilities are possible for cellular convective modes other than rolls.

INTRODUCTION

Thermal convection of one component fluid, driven through thermal buoyancy, has been extensively studied [1–5]. Nonlinear analysis has also been investigated [6–8]. Double-diffusive convection of a two-component fluid can be induced through the mutual interaction of thermal and compositional gradients, even the density profile is statically stable [9, 10]. Either oscillatory or subcritical instability is possible [11]. Wollkind and Segel [12] treat the morphological instability of a freezing interface. Both cellular and dendritic structures of the interface are discovered. Sekerka *et al.* [13] study the onset of the coupled compositional convective and morphological instability with a stabilizing temperature gradient and find that the oscillatory instability is possible. In their study, Loper and Roberts [14] and Jou [15] include the material diffusion by the pressure gradient.

The main reason for studying the compositional convection with freezing lies with the concern of the efficiency of energy conversion and of the self-induced energy source. Due to the large thermal diffusivity, most of the available heat is conducted along the adiabat before the convection can set in. In contrast, it takes only a small flux of light material, due to the small material diffusivity, to reach a marginal state through the material diffusion [14–16]. The key factors of compositional convection are the cooling rate, the released light material and the material diffusion.

PHYSICAL FORMULATION

A fluid layer of binary alloy, composed of a heavy metallic component and a light non-metallic component, lies between two horizontal, infinite and rigid boundaries with a distance $(d-\eta)$ apart. The top boundary is subject to a small outward heat flux and the freezing, provided $\mathrm{d}T_L/\mathrm{d}p > \mathrm{d}T_A/\mathrm{d}p$, occurs at the lower boundary, where T_L is the liquidus temperature, T_A is the adiabatic temperature and p is the pressure. In considering the thermal and dynamical state of the earth's core, the solid inner core has been forming and growing from the liquid outer core of a binary alloy over a long period of time. The liquid outer core can be treated as being nearly adiabatic and homogeneous. For a liquid outer core to exist, the adiabat must intersect the liquidus at the freezing interface. Stevenson [17] has shown quantitatively that the ratio of $\mathrm{d}T_L/\mathrm{d}p$ to $\mathrm{d}T_A/\mathrm{d}p$ is about 1.67 and the condition, $\mathrm{d}T_L/\mathrm{d}p > \mathrm{d}T_A/\mathrm{d}p$, is well satisfied. The solidification thus proceeds from the bottom upward, despite the top being the coldest.

Since the composition of light component in the liquid alloy ξ and that in the solid alloy ξ_S are less

NOMENCLATURE

a	wavenumber
A	finite amplitude
C	specific heat capacity
C_p	specific heat capacity at constant pressure
D	material diffusivity
$\bar{D}$	d/dz
d	original position of the upper boundary
F	normalized solution of ξ' at marginal state
f	horizontal harmonic function
g	gravity
H	outward heat flux
k	thermal diffusivity
K	thermal conductivity
$L = l\hat{T}$	latent heat
$\hat{\mathbf{n}}$	normal unit vector
p	pressure
t	time
T	temperature
$T_0(0)$	temperature at the lower boundary
T_A	adiabatic temperature
T_C	conducting temperature
T_L	liquidus temperature
$\hat{T}$	liquidus temperature of one component
$\underline{u}$	velocity
u_z	vertical velocity
$\underline{v}$	advancing velocity of the lower boundary
V	advancing velocity of the planar lower boundary
W	normalized solution of u_z at marginal state
w	scalar variable
x, y, z	Cartesian coordinates
X, Y, Z	strained Cartesian coordinates
$\hat{\mathbf{z}}$	vertical unit vector.

Greek symbols

α	coefficient of thermal expansion
β	rate of change of temperature
γ	coefficient of surface tension
δ	volume change upon freezing
$\xi\bar{\delta}$	volume change due to compositional change
η	position of the lower boundary
μ_l	relative chemical potential of liquid alloy
$\bar{\mu}$	$\partial\mu_l/\partial\xi$
μ	dynamical viscosity
ν	kinematic viscosity
ξ	light composition of liquid alloy
$\xi_0(0)$	light composition at the lower boundary
$\rho(\rho_s)$	density of liquid (solid) alloy
$\sigma = \sigma_r + \sigma_i$	complex frequency.

Parameters

ε_1	dV/D
ε_2	$\rho\bar{\delta}gd/\bar{\mu}\xi_0(0)$
ε_k	dV/K
Q	$(\rho\bar{\delta}gd^2)^2/\nu D\bar{\mu}$
R	$\bar{\mu}V\,\xi_0(0)/\rho\bar{\delta}gD$
P_L	Schmidt number ν/D
P_T	$HC/VlT_0(0)$
S	$\delta/\xi_0(0)\bar{\delta}$
S_0	$Vl^2T_0(0)/\rho\delta\xi_0(0)g$
Ω	K/D
Ω_0	$l^2T_0(0)/C\bar{\mu}\xi_0^2(0)$
q	$\gamma/\rho^2\xi_0(0)\bar{\delta}g$.

Superscripts

$\bar{}$	basic state
$'$	perturbed state
$\bar{}$	horizontal average
$*$	complex conjugate.

Subscripts

c	critical state.

than that at the eutectic point ξ_e, where the eutectic point has the lowest temperature at which the liquid alloy can exist, the solid alloy contains less light component and a density jump across the freezing interface causes the releasing of some light material. Convective instabilities are possible, when a destabilizing compositional profile occurs, provided $d\xi/dp > 0$ inside the fluid layer.

In a thin boundary layer near the freezing interface, convective motions are suppressed and heat is removed through conduction. The freezing interface, advancing upward with a small speed by accumulating the frozen binary alloy, is perturbed. If $dT_C/dp < dT_L/dp$, the liquid alloy is subcooled and is frozen directly onto the interface and any perturbation eventually dies out, where T_C is the conduction temperature, the convective flows exhibit cellular patterns. If $dT_C/dp > dT_L/dp$, the liquid alloy is constitutionally supercooled. The freezing is enhanced by the unstable growth of the dendrites. The convective flows may exhibit non-cellular patterns.

The governing equations and boundary conditions, assuming the Boussinesq's approximation and neglecting the thermal buoyancy, are [14, 15]

$$\nabla \cdot \underline{u} = 0 \tag{1}$$

$$\frac{\partial \xi}{\partial t} + (\underline{u} \cdot \nabla)\xi = D\left(\nabla^2 \xi + \frac{\bar{\delta}}{\bar{\mu}} \nabla^2 p\right) \tag{2}$$

$$\rho\left[\frac{\partial u}{\partial t} + (\underline{u} \cdot \nabla)\underline{u}\right] = -p + \rho g + \mu \nabla^2 \underline{u} \tag{3}$$

$$\rho C\left[\frac{\partial T}{\partial t} + (\underline{u} \cdot \nabla) T\right] = k \nabla^2 T \tag{4}$$

$$\rho^{-1} = \rho_s^{-1} + \xi\bar{\delta} + \delta \tag{5}$$

$$\underline{u} = 0, \quad \hat{\mathbf{z}} \cdot \nabla T = -\frac{H}{k}, \quad \hat{\mathbf{z}} \cdot \left(\nabla \xi + \frac{\bar{\delta}}{\bar{\mu}} \nabla p\right) = 0 \quad \text{at} \quad \mathbf{z} = d \tag{6}$$

$$\hat{\mathbf{n}} \cdot \underline{u} = 0, \quad \hat{\mathbf{n}} \times \underline{u} = 0 \tag{7}$$

$$(\underline{v} \cdot \hat{\mathbf{n}})\rho_s l T = -k \nabla T \cdot \hat{\mathbf{n}} \tag{8}$$

$$(\underline{v} \cdot \hat{\mathbf{n}})\rho_s \xi = -\rho D\left(\nabla \xi + \frac{\bar{\delta}}{\bar{\mu}} \nabla p\right) \cdot \hat{\mathbf{n}} \tag{9}$$

$$T = T_L = \hat{T} + \frac{\delta}{l} p - \frac{\bar{\mu}}{2l} \xi^2 + \frac{\gamma \nabla_H^2 \eta}{\rho l [1 + (\nabla_H \eta)^2]^{3/2}} \quad \text{at} \quad z = \eta(x, y, t) \tag{10}$$

where $\underline{u}$ is the velocity, T is the temperature, D is the material diffusion, μ is the viscosity, C is the heat capacity, k is the thermal conductivity, $\rho(\rho_s)$ is the liquid(solid) density, $\xi\bar{\delta}$ and δ are volume changes due to the compositional change and the freezing, H is the heat flux, $\hat{T}$ is the liquidus of one component, $L = l\hat{T}$ is the latent heat, μ_l is the relative chemical potential, $\bar{\mu} = \partial\mu_l/\partial\xi$ and γ is the coefficient of surface tension.

For the non-convective state, we assume that the dependent variables are functions of z and t only, the lower boundary is planar and advances upward at a small velocity (i.e. $\eta = \eta(t)$ and $v = V\hat{\mathbf{z}}$) and $\rho_s = \rho$. A set of non-dimensional equations, by setting $z \to dz$, $t \to d^2/Dt$, $T \to T_r T$ and $p \to (\rho g d)p$, is obtained.

$$\frac{\partial \xi}{\partial t} = \frac{\partial^2 \xi}{\partial z^2} + \varepsilon_2 \frac{\partial^2 p}{\partial z^2} \tag{11}$$

$$\frac{\partial p}{\partial z} = -1 \tag{12}$$

$$\frac{1}{\Omega} \frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial z^2} \tag{13}$$

$$\frac{\partial T}{\partial z} = -\varepsilon_k \left(\frac{H}{T_r V \rho C}\right), \quad \frac{\partial \xi}{\partial z} + \varepsilon_2 \frac{\partial p}{\partial z} = 0 \quad \text{at} \quad z = 1 \tag{14}$$

$$\frac{\partial T}{\partial z} = -\varepsilon_k \left(\frac{\rho l}{C}\right) T \quad \frac{\partial \xi}{\partial z} + \varepsilon_2 \frac{\partial p}{\partial z} = -\varepsilon_1 \xi \quad \text{at} \quad z = \eta = \varepsilon_1 t \tag{15}$$

where $\varepsilon_1 = dV/D$, $\varepsilon_2 = \rho\bar{\delta}gd/\bar{\mu}\xi_r$, $\varepsilon_k = dV/K$ and $\Omega = K/D$ and K is the thermal diffusivity and T_r and ξ_r are the reference temperature and composition. Since the compositional and thermal diffusive processes act quickly compared with the advancement of the boundary, we may assume $\varepsilon_1 \ll 1$ and $\varepsilon_k \ll 1$. Expanding about $z = 0$ and linearizing the boundary condition (15), we have

$$\frac{\partial T(0)}{\partial z} + \frac{\partial^2 T(0)}{\partial z^2} = -\varepsilon_1 \frac{1}{\Omega}\left(\frac{\rho l}{C}\right)\left[T(0) + \frac{\partial T(0)}{\partial z}\eta\right]$$

$$\times \left[\frac{\partial \xi(0)}{\partial z} + \varepsilon_2 \frac{\partial p(0)}{\partial z}\right] + \eta \left[\frac{\partial^2 \xi(0)}{\partial z^2} + \varepsilon_2 \frac{\partial^2 p(0)}{\partial z^2}\right]$$

$$= -\varepsilon_1 \left[\xi(0) + \frac{\partial \xi(0)}{\partial z}\eta\right] \quad \text{at} \quad z = 0. \tag{16}$$

Manipulating the perturbation expansions for ξ and T about ε_1

$$\xi(z, t) = \xi_0(z) + \varepsilon_1 \xi_1(z, t) + O(\varepsilon_1^2)$$

$$T(z, t) = T_0(z) + \varepsilon_1 T_1(z, t) + O(\varepsilon_1^2)$$

and applying to equations (11)–(14) and (16), the non-convective solutions in dimensional forms are

$$\xi = \xi_0(0)\left\{1 + \varepsilon_2\left[\frac{z}{d} - R\left(\frac{z}{d} - \frac{z^2}{2d^2}\right)\right] + R\frac{D}{d^2}t\right\} \tag{17}$$

$$T = T_0(0) + \varepsilon_1 \left\{\left[-\left(\frac{H}{V\rho}\right) + \left(\frac{l}{C}\right) T_0(0)\right] \times \left[\frac{1}{\Omega}\frac{z^2}{2d^2} + \frac{D}{d^2}t\right] - \frac{1}{\Omega}\left(\frac{l}{C}\right) T_0(0) \frac{z}{d}\right\} \tag{18}$$

where we choose $T_r = T(0)$ and $\xi_r = \xi_0(0)$ and define $R = \bar{\mu}V\xi_0(0)/\rho\bar{\delta}gD$. To $O(1)$, the heat flux is absent and no freezing is possible. To $O(\varepsilon_1)$, the heat flux is imposed on the top and the freezing does occur at the bottom. Rescaling equation (17), by setting $\xi \to v/\rho\bar{\delta}gd^3\xi$ and $z \to dz$, to give

$$\frac{\partial \xi}{\partial z} = Q[1 - R(1 - z)] \tag{19}$$

where $Q = (\rho\bar{\delta}gd^2)^2/vD\bar{\mu}$.

Although the light composition is increasing with time, its profile is assumed unchanged. The minimum ξ, occurring at $z = 1 - 1/R$, is equal to $Q(R-1)/2R + \xi_0(0)$. For $R > 1$, a destabilizing compositional profile (i.e. $\partial\xi/\partial z < 0$) appears near the freezing interface. In the earth's liquid outer core, the physical parameter R, neglecting thermal effects, has been shown to be about 660, which satisfies the condition $R > 1$ very well, and the compositional convection is dynamically vigorous [18].

We separate each dependent variable into a non-convective part and a convective part (i.e. $\underline{u} = \underline{\bar{u}}(z, t) + \underline{u}'(x, y, z, t), \ldots,$ etc.). We also let $\eta = \bar{\eta}$

$(t)+\eta'(x,y,t)$ and $\underline{v} = V\hat{z}+v'(x,y,t)$. Assumption $|\eta/d| \ll 1$ allows us to replace $\hat{\mathbf{n}}$ by $\hat{\mathbf{z}}$ and $\partial/\partial n$ by $\partial/\partial z$. Non-dimensional governing equations of the convective state may be obtained by letting

$$\xi' \to \frac{vD}{\rho\bar{\delta}g\,d^3}\xi' \quad p' \to \frac{\rho vD}{d}p' \quad T' \to \frac{DVdlT_0(0)}{K^2C}T'$$

$$\underline{u}' \to \frac{D}{d}\underline{u}' \quad \eta' \to d\eta' \quad \underline{x} \to d\underline{x} \quad \text{and} \quad t \to \frac{d}{v}t$$

we expand the perturbed boundary conditions at $z = \eta$ about $z = \bar{\eta}$ and, further, about $z = 0$ and keep the first-order terms only. The non-dimensional governing equations for convective motion are

$$\frac{1}{P_L}\left[\varepsilon_1\frac{\partial \underline{u}'}{\partial t}+(\underline{u}'\cdot\nabla)\underline{u}'\right] = -\nabla p'+\xi'\hat{\mathbf{z}}+\nabla^2\underline{u}' \quad (20)$$

$$\varepsilon_1\frac{\partial \xi'}{\partial t}+(\underline{u}'\cdot\nabla)\xi'+Q[1-R(1-z)]\underline{u}_z' = \nabla^2\xi' \quad (21)$$

$$\frac{1}{\Omega}\left[\varepsilon_1\frac{\partial T'}{\partial t}+(\underline{u}'\cdot\nabla)T'\right]+[(1-P_T)z-1]u_z' = \nabla^2 T' \quad (22)$$

$$\nabla\cdot\underline{u}' = 0 \quad (23)$$

$$\underline{u}' = \frac{\partial\xi'}{\partial z} = \frac{\partial T'}{\partial z} = 0 \quad \text{at} \quad z = 1 \quad (24)$$

$$\underline{u}' = 0 \quad (25a)$$

$$\frac{\partial\xi'}{\partial z}+QR\eta' = 0 \quad (25b)$$

$$\frac{\partial T'}{\partial z}+\Omega(1-P_T)\eta' = 0 \quad (25c)$$

$$\Omega_0\frac{QR}{\Omega^2}T'+\xi+Q[-S_0+S+(1-R)]\eta' + q\nabla_H^2\eta' = 0 \quad \text{at} \quad z = 0 \quad (25d)$$

where $P_L = v/D$, $P_T = H/VlT_0(0)$, $\Omega_0 = l^2T_0(0)/C\bar{\mu}\xi_0^2(0)$, $S_0 = Vl^2T_0(0)/\rho\delta\xi_0(0)g$, $S = \delta/\xi_0(0)\bar{\delta}$ and $q = \gamma/\rho^2\xi_0(0)\bar{\delta}g$. The perturbed pressure terms have been dropped by assuming $(\rho\bar{\delta})^2gd/\bar{\mu} \ll 1$ and $\rho\bar{\delta}\delta gd/\bar{\mu}\xi_0(0) \ll 1$. Although S_0 can be incorporated into S, however we have assumed a small heat flux on the top such that $S_0 \ll S$. Elimination of η' among equations (25b)–(25d) yields

$$\left(\frac{QR}{\Omega}\right)\frac{\partial T'}{\partial z}-(1-P_T)\frac{\partial\xi'}{\partial z} = 0 \quad (26)$$

$$\left(\frac{QR}{\Omega}\right)^2T'+QR\xi'-Q[S+(1-R)]\frac{\partial\xi'}{\partial z}+q\nabla_H^2\frac{\partial\xi'}{\partial z} = 0. \quad (27)$$

Since the thermal diffusivity is much larger than the material diffusivity (i.e. $\Omega \gg 1$), equation (22) is compositionally and diffusively dominated such that $T' = O(\Omega)$ and equation (27) gives rise to

$$R\xi'+(R-1-S)\frac{\partial\xi'}{\partial z}+\frac{q}{Q}\nabla_H^2\left(\frac{\partial\xi'}{\partial z}\right) = 0 \quad \text{at} \quad z = 0. \quad (28)$$

As mentioned previously, cellular convective modes require $dT_C/dp < dT_L/dp$, corresponding to $R < 1+S+qa^2/Q$, while morphological modes require $dT_C/dp > dT_L/dp$, corresponding to $R > 1+S+qa^2/Q$, here the effect of surface tension is taken into account. In considering the freezing interface, lying between the earth's solid inner and liquid outer cores, the ratio of volume change upon freezing to that due to the compositional change S is approximated to be 0.29 such that the condition $1+S < R$ is well satisfied [18]. In general, we may conclude that morphological instability becomes dominant inside the earth's core.

LINEAR ANALYSIS

The governing equations and boundary conditions for the compositional convection, after neglecting thermal effects, are

$$\frac{1}{P_L}\left[\varepsilon_1\frac{\partial \underline{u}'}{\partial t}+(\underline{u}'\cdot\nabla)\underline{u}'\right] = -\nabla p'+\xi'\hat{\mathbf{z}}+\nabla^2\underline{u}' \quad (29a)$$

$$\varepsilon_1\frac{\partial \xi'}{\partial t}+(\underline{u}'\cdot\nabla)\xi'+Q[1-R(1-z)]\underline{u}_z' = \nabla^2\xi' \quad (29b)$$

$$\nabla\cdot\underline{u}' = 0 \quad (29c)$$

$$\underline{u}' = \frac{\partial\xi'}{\partial z} = 0 \quad \text{at} \quad z = 1 \quad (30)$$

$$\underline{u}' = R\xi'+(R-1-S)\frac{\partial\xi'}{\partial z}+\frac{q}{Q}\nabla_H^2\left(\frac{\partial\xi'}{\partial z}\right) = 0 \quad \text{at} \quad z = 0. \quad (31)$$

For linear analysis, we neglect the nonlinear terms in equation (29). After applying the operator $\hat{\mathbf{z}}\cdot(\nabla\times\nabla\times)$ to equation (29a), we have

$$\frac{\varepsilon_1}{P_L}\frac{\partial}{\partial t}\nabla_H^2 u_z' = \nabla_H^2\xi'+\nabla_H^4 u_z'. \quad (32)$$

Let us analyze the problem in terms of normal modes by assuming

$$u_z' = e^{\sigma t}a^2f(x,y)W(z)$$

$$\underline{u}_H' = e^{\sigma t}\nabla_H f(x,y)\bar{D}W(z)$$

$$\xi' = e^{\sigma t}f(x,y)F(z)$$

where $\nabla_H = \partial/\partial x\hat{i}+\partial/\partial y\hat{j}$, $\nabla_H^2 f = -a^2 f$ and $\bar{D} = d/dz$.

Now equations (29)–(32) have the following forms

$$(\bar{D}^2-a^2)\left(\bar{D}^2-a^2-\frac{\varepsilon_1}{P_L}\sigma\right)W = F \qquad (33a)$$

$$(\bar{D}^2-a^2-\varepsilon_1\sigma)F = -a^2Q[R(1-z)-1]W \qquad (33b)$$

$$W = \bar{D}W = \bar{D}F = 0 \quad \text{at} \quad z=1 \qquad (34)$$

$$W = \bar{D}W = RF+\left(R-1-S-\frac{q}{Q}a^2\right)\bar{D}F = 0 \quad \text{at} \quad z=0. \qquad (35)$$

The principle of exchange of stability is valid and the convection sets in stationarily at the marginal state, provided $\sigma_r = \sigma_i = 0$, where $\sigma_r = \mathrm{Re}(\sigma)$ and $\sigma_i = \mathrm{Im}(\sigma)$. Otherwise, overstability is possible, provided $\sigma_r = 0$ and $\sigma_i \neq 0$. Multiplying equation (33b) by the complex conjugate function of F (i.e. F^*), integrating over z from 0 to 1 and using equations (33a), (34) and (35), then we have, by setting $\sigma_r = 0$,

$$-\int_0^1 \{[|\bar{D}F|^2+a^2|F|^2] + a^2Q[1-R(1-z)]|(\bar{D}^2-a^2)W|^2\}\,dz + \frac{R}{R-1-S-qa^2/Q}|\bar{D}F^*(0)|^2 = a^2QR\frac{\varepsilon_1\sigma_i}{P_L}G_i$$

$$\varepsilon_1\sigma_i\int_0^1\left\{|F|^2-a^2Q\frac{1}{P_L}[1-R(1-z)][|\bar{D}W|^2 + a^2|W|^2]\right\}dz = 2a^2QR(H_i-a^2G_i) \qquad (36)$$

where

$$G_i = \mathrm{Im}\left[\int_0^1 \bar{D}W\cdot W^*\,dz\right]$$

and

$$H_i = \mathrm{Im}\left[\int_0^1 \bar{D}W\cdot \bar{D}^2W^*\,dz\right].$$

It should be remembered that $1-R(1-z) < 0$ for a possible convection.

Numerically solving equations (33)–(35) shows that R is minimized when $\varepsilon_1\sigma_i = 0$. The principle of exchange of stabilities is valid and compositional convection sets in stationarily. The eigenvalue R, in the limits $S\to\infty$ and $q/Q\to 0$, can be expressed as [15],

$$R = \frac{\int_0^1 [|\bar{D}F|^2+a^2|F|^2]\,dz + a^2Q\int_0^1 |(\bar{D}^2-a^2)W|^2\,dz}{a^2Q\int_0^1 (1-z)|(\bar{D}^2-a^2)W|^2\,dz}. \qquad (37)$$

Applying the operator $(\hat{z}\cdot\nabla\times)$ to equation (29a), we have

$$\nabla^2(\hat{z}\cdot\nabla\times\underline{u}') = 0. \qquad (38)$$

Non-slip boundary conditions together with equation (38) imply that

$$\hat{z}\cdot(\nabla\times\underline{u}') = 0 \qquad (39)$$

which allows us to express $\underline{u}'$ in terms of a scalar variable w;

$$\underline{u}' = \nabla\left(\frac{\partial w}{\partial z}\right)-(\nabla^2 w)\hat{z}.$$

The steady convective state with $P_L\to\infty$ are

$$P = \nabla^2\left(\frac{\partial w}{\partial z}\right)$$

$$\nabla^4 w = \xi$$

$$\nabla^2\xi = Q[R(1-z)-1]\nabla_H^2 w + \nabla\xi\cdot\nabla\left(\frac{\partial w}{\partial z}\right)-(\nabla^2 w)\left(\frac{\partial w}{\partial z}\right) \qquad (40)$$

$$w = \frac{\partial w}{\partial z} = \frac{\partial \xi}{\partial z} = 0 \quad \text{at} \quad z=1 \qquad (41)$$

$$w = \frac{\partial w}{\partial z} = R\xi+(R-1-S)\frac{\partial\xi}{\partial z} = 0 \quad \text{at} \quad z=0. \qquad (42)$$

The critical value of Q with R vanishing has been shown to be equal to -720 at the critical wavenumber $a_c^2 = 0$ [4]. We assume the minimum value of R in terms of Q also occurs for $a^2 \ll 1$ and $S \gg 1$ and expand ξ, w and R asymptotically in terms of a^2 with $1/S = O(a^4)$

$$\xi(x,y,z) = \xi_0(X,Y,z)+a^2\xi_1(X,Y,z)+O(a^4)$$

$$w(x,y,z) = w_0(X,Y,z)+a^2w_1(X,Y,z)+O(a^4)$$

$$R = R_0+a^2R_1+O(a^4) \qquad (43)$$

where $X = ax$ and $Y = ay$.

After substituting equation (43) into equations (40)–(42), a set of asymptotic solutions is found

$$(a^0): w_0 = \xi_0\frac{z^2(1-z)^2}{24}$$

$$\xi_0 = \xi_0(X,Y)$$

$$R_0 = \left(2+\frac{1440}{Q}\right)\left(1+\frac{1440}{QSa^2}\right) \qquad (44)$$

$$O(a^2): w_1 = z^2(1-z^2)[g_1(z)+\frac{Qg_2(z)}{288}\nabla_H^2\xi_0 + \frac{g_4(z)}{144}(\nabla_H\xi_0)^2]/55440$$

$$\xi_1 = \left[\frac{g_4(z)}{14}+\frac{Qg_5(z)}{10080}\right]\nabla_H^2\xi_0+\frac{g_6(z)}{720}(\nabla_H\xi_0)^2$$

$$R_1 = \frac{1}{Q\nabla_H^2\xi_0}\left\{-\left[\frac{4080}{77} - \frac{(720+Q)^2}{216216}\right]\nabla_H^4\xi_0 + \frac{(720+Q)}{16632}\nabla_H^2(\nabla_H\xi_0)^2 + \frac{5(720+Q)}{16632}\nabla_H\cdot[(\nabla_H^2\xi_0)\nabla_H\xi_0] + \frac{1}{252}\nabla_H\cdot[(\nabla_H\xi_0)^2\nabla_H\xi_0]\right\} \quad (45)$$

where

$$g_1(z) = 63+161z-126z^2+49z^3 - 7z^4-63z^5+46z^6-10z^7$$

$$g_2(z) = 23+16z+9z^2+2z^3-5z^4-12z^5+14z^6-4z^7$$

$$g_3(z) = 11\cdot(15+10z+5z^2-5z^4+2z^5)$$

$$g_4(z) = -7z^2+70z^4-126z^5+84z^6-20z^7$$

$$g_5(z) = 35z^4-84z^5+70z^6-20z^7$$

$$g_6(z) = 10z^3-15z^4+6z^5$$

and $\nabla_H^2 = \partial^2/\partial X^2+\partial^2/\partial Y^2$.

For the linear analysis, we may neglect the nonlinear terms in equation (45) and assume

$$\nabla_H^2\xi_0 = -\xi_0.$$

To the second order approximation, we find, for cellular convective modes of long wavelengths with $a^2 \ll 1$, $S \gg 1$ and $q = O(1)$, that

$$R = \left(2+\frac{1440}{Q}\right)\left(1+\frac{1440}{QSa^2}\right) + \left[\frac{4080}{77} - \frac{(720+Q)^2}{216216}\right]\frac{a^2}{Q} + O\left(\frac{a^4}{Q}\right) \quad (46)$$

which gives the critical wavenumber as

$$a_c^2 = \left\{1440\left(2+\frac{1440}{Q}\right)\Big/S\left[\frac{4080}{77} - \frac{(720+Q)^2}{216216}\right]\right\}^{1/2}$$

provided $Q < Q^* = 2664.77$. The minimum value of a_c^2 with respect to Q occurs at $Q = Q^{**} = 1013.26$ and equals to $11.226/S^{1/2}$. In the limit $S \to \infty$, the critical values are reduced to the following [14],

$$R_c = \left(2+\frac{1440}{Q}\right) \quad \text{and} \quad a_c^2 = 0.$$

For $Q^* \ll Q$, we assume a new scalar $Z = z/\varepsilon$. A possible balance, from equation (33), is achieved by choosing

$$\varepsilon = O(Q^{-1/5}), a = O(Q^{1/5}), F = O(1), W = O(Q^{-4/5}) \text{ and } R = 1+O(Q^{-1/5}). \quad (47)$$

The linearized equations (33) and boundary conditions (34) and (35) are numerically solved. The cellular convective modes of short wavelength with $a^2 \gg 1$, $Q \gg Q^*$ and $q = O(1)$ are found. By quadratically approximating R_c and a_c^2 at $Q = 10^5$, 10^6 and 10^7, we have

$$R_c - 1 = 3.0304Q^{-1/5}+23.705Q^{-2/5}-30.187Q^{-3/5}$$

$$a_c^2 = 14.847-2.2877Q^{1/5}+0.2803Q^{2/5}$$

provided $Q(R-1)/a^4 = O(1)$.

For morphological modes of short wavelength with $Q(R-1)/a^4 = o(1)$, the perturbation expansions of the variables are expressed in terms of the wavenumber a^2, then, the first order approximation of the eigenvalue becomes,

$$R = \frac{(1+S+a^2q/Q)a(1-e^{2a})}{[(a+1)-e^{2a}(a-1)]} \quad (48)$$

which gives rise to the critical values [15],

$$R_c = (1+S)\left\{1+\left[\frac{q}{2Q(1+S)}\right]^{1/3}\right\}$$

$$a_c^2 = \left[\frac{Q(1+S)}{2q}\right]^{2/3}.$$

In the limit $q \to 0$, at which case the effect of surface tension becomes neglected, the critical values reduce to

$$R_c = 1+S \quad \text{and} \quad a_c^2 \to \infty.$$

R_c and a_c^2 as functions of S for various values of Q and q/Q are shown in Figs. 1 and 2. Two major modes of instabilities, cellular convective and morphological, are discovered and would compete with each other for the occurrence at the marginal state. The mode, having a smaller eigenvalue R as the critical one R_c, becomes dominant. R_c of morphological modes is plotted, for $q/Q = 0$, as a straight line and is deviated, for $q/Q \neq 0$, away from that line slightly upward at small values of S and negligibly at large values of S. While R_c of cellular convective modes are plotted below those of morphological modes and, as the value of q/Q increases from zero and up, would extend the

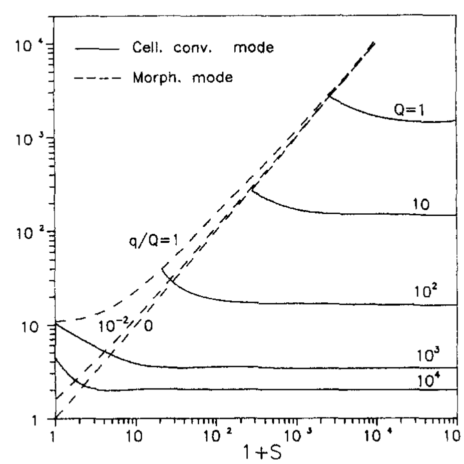

Fig. 1. R_c as a function of S for various values of Q and q/Q.

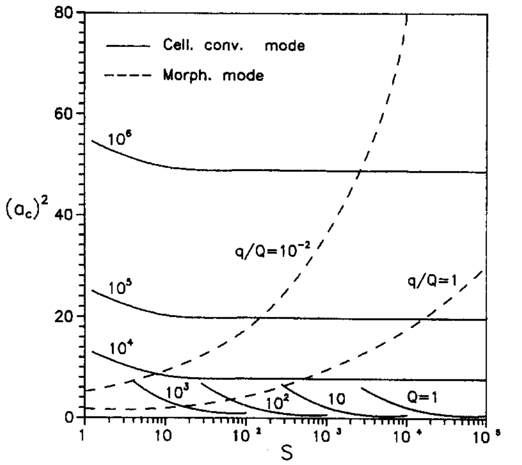

Fig. 2. a_c^2 as a function of S for various values of Q and q/Q.

curves leftward and upward. The increasing value of q/Q would suppress both cellular convective and morphological modes, especially $Q^* \ll Q$ and S small.

It is shown obviously that, for values of S large and fixed, the critical values R_c of cellular convective modes of both long and short wavelengths are insignificantly affected by the effect of surface tension, associated with the parameter q, but strongly dependent on the diffusive effect of the light material, exerted by the pressure gradient and associated with the parameter Q. This decreasing trend of R_c with Q is quite rapid at small values of Q (i.e. $Q < Q^*$) for cellular convective modes of long wavelengths and become relatively slow at large values of Q (i.e. $Q^* \ll Q$) for those of short wavelengths, which cases are even irrespective of the values of S.

R_c of morphological modes strongly depends on the effect of surface tension. This increasing trend of R_c with q/Q is observed for $Q^* \ll Q$ and S small and becomes negligible for Q small and S large. a_c^2 of morphological modes are infinite for any value of Q and S, provided $q/Q = 0$, and become finite and increase with S, provided $q/Q \neq 0$, and decrease with q/Q slowly for S small and rapidly for S large. a_c^2 of cellular convective modes of long wave lengths decrease with S and become vanishing as $S \to \infty$, and those of short wave lengths decrease with S slowly and approach fixed values. While a_c^2 of cellular convective modes decrease with Q in the region of long wavelengths and, then, increase with Q in the region of short wavelengths, there exists a value of Q, say Q^{**}, such that a_c^2 with respect to Q is minimum. It is found that, for S very large, minimum a_c^2 occurs at $Q^{**} = 1013.26$ and is approximated by $11.226/S^{1/2}$.

For a fixed value of q/Q or Q, R_c of the prevailing morphological modes increases with S upto a frontier value S_m and, as S goes from the frontier value S_m to infinity, R_c of cellular convective modes, now becoming dominant, decreases with S. There always exists, across the frontier value S_m, a jump from the morphological mode to the cellular convective mode. Table 1 shows, for various values of Q and $q = 0$, the critical values R_c and a_c^2 at the frontier value S_m and its related prevailing modes. In the range $Q < Q'$, where $Q' = 351.6$ with a corresponding frontier value $S'_m = 11.499$, cellular convective modes have smaller eigenvalues than those of morphological ones at the frontier value S_m and become solely dominant, while, in the range $Q' \leqslant Q$, both cellular convective and morphological modes, having the same eigenvalue at the frontier value S_m, coexist simultaneously and the flow, as a result of appearance of both wavenumbers, would show a mixed structure. For $q/Q \neq 0$, the frontier points (Q, S_m) from Fig. 1, for Q fixed, are raised slightly upward, especially $Q^* \ll Q$ and S small.

Domains on Q–S plane occupied by cellular convective modes of long and short wavelengths and morphological modes of short wavelengths are shown in Fig. 3. B_1 is the locus joint by points (S_m, Q). Along the locus B_1 upto (S'_m, Q'), abrupt jumps of both R_c and a_c^2 occur across B_1. While, along the locus B_1 from (S'_m, Q') and on, R_c is continuous and a_c^2 is discontinuous across B_1. B_1, B_2, D_1 and D_2 are artificial contours. Strictly speaking, cellular convective modes of long wavelengths are confined within the region between the contours D_1 and B_2 and of short wavelengths are confined within the region beyond the contour D_2. While morphological modes of short wavelengths are confined below the contour B_1. The effect of surface tension (i.e. $q/Q \neq 0$) would slightly lower the contour B_1 down and reduce the region of morphological modes.

NONLINEAR ANALYSIS

The onset of convective instability implies that the kinetic energy dissipated by viscosity is balanced by the potential energy of the light material released at

Table 1. Critical values R_c and a_c^2 at the frontier value S_m for $q = 0$

Q	S_m	a_c^2 (cell.)	a_c^2 (morph.)	R_c	Modes
10^4	1.2372	13.061	∞	2.2372	Both
10^3	3.8586	7.3597	∞	4.8586	Both
$Q' = 351.6$	$S'_m = 11.499$	7.4256	∞	12.499	Both
10^2	28.622	6.8317		29.056	Cellular
10^1	275.78	6.7180		268.41	Cellular
10^0	2747.2	6.7081		2661.8	Cellular

the freezing interface and the infinitesimal amplitude begins to amplify. As R goes beyond R_c, the disturbance will grow exponentially, but eventually it reaches a finite amplitude and becomes appreciable and the nonlinear effects on the fluid flows are no longer negligible.

Let us suppose that, in the nonlinear steady convection,

$$\langle u\rangle = 0, \quad \langle p\rangle = \bar{p} \quad \text{and} \quad \langle \xi\rangle = \bar{\xi} \tag{49}$$

where $\langle\ \rangle$ is the averaged quantity over the entire horizontal plane. We also assume that

$$\underline{u} = \underline{u}', \quad p = \bar{p}+p' \quad \text{and} \quad \xi = \bar{\xi}+\xi' \tag{50}$$

where $\underline{u}$, p' and ξ' are the dynamically perturbed variables and $\langle p'\rangle = \langle \xi'\rangle = 0$ by definitions of equation (50). Averaging the nonlinear steady governing equations over the entire horizontal plane, we have

$$\frac{1}{P_L}\langle u_z'^2\rangle = -\frac{\partial p}{\partial z}+\bar{\xi}$$

$$\frac{\partial}{\partial z}\langle u_z'\xi'\rangle = \frac{\partial^2\bar{\xi}}{\partial z^2}. \tag{51}$$

Equation (51b) can be integrated once to give

$$\langle u_z'\xi'\rangle = \frac{\partial\bar{\xi}}{\partial z}. \tag{52}$$

It should be remembered that $\bar{\xi} = 0$ for the linear analysis. Multiplying equation (29a) by $\underline{u}'$ and equation (29b) by ξ', averaging over the horizontal plane and then integrating on z from 0 to 1, we have

$$\int_0^1 \langle u_z'\xi'\rangle\,\mathrm{d}z+\int_0^1 \langle \underline{u}'\cdot\nabla^2\underline{u}'\rangle\,\mathrm{d}z = 0 \tag{53}$$

$$\int_0^1 (\langle \xi' u_z'\rangle)^2\,\mathrm{d}z+\int_0^1 [1-R(1-z)]\langle \xi' u_z'\rangle\,\mathrm{d}z = \int_0^1 \langle \xi'\nabla^2\xi'\rangle\,\mathrm{d}z. \tag{54}$$

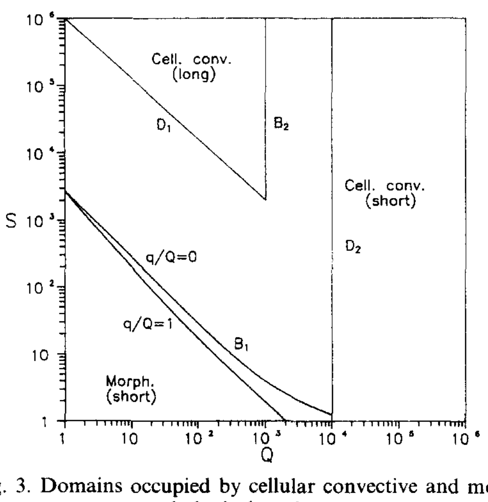

Fig. 3. Domains occupied by cellular convective and morphological modes.

Empirically, the 'shape assumption' is valid [19], if the flow pattern, first appearing at the marginal state, continues to manifest itself long past the marginal state. Let us assume that, just past the marginal state, ξ' and $\underline{u}'$ can be expressed as

$$\xi' = Af(x,y)F(z)$$

$$\underline{u}' = A[\tilde{D}W(z)\nabla_H f(x,y)+a^2 f(x,y)W(z)\hat{z}] \tag{55}$$

where $W(z)$ and $F(z)$ are the normalized solutions at the marginal state, A is the amplitude small but finite and $f(x,y)$ is a two-dimensional (2D) periodic wave function with wavenumber a. Without loss of generality we also assume

$$\langle f^2\rangle = 1 \quad \text{and} \quad \nabla_H^2 f = -a^2 f. \tag{56}$$

Substituting equation (55) into equation (54) and employing equation (56), the finite amplitude A just past the marginal state is expressed as

$$A^2 = \frac{Q(R-R_c)}{a^2}\cdot\frac{\int_0^1 (1-z)WF\,\mathrm{d}z}{\int_0^1 W^2F^2\,\mathrm{d}z} \tag{57}$$

where a^2 is assumed to be the critical wavenumber by the 'shape-assumption'. The amplitude of the nonlinear disturbance behaves like $(R-R_c)^{1/2}$.

We may assume $\xi_0 = Af(X, Y)$ such that, as $S\to\infty$ and $a^2 \ll 1$,

$$\langle \xi_0^2\rangle = A^2 \quad \text{and} \quad \langle f^2\rangle = 1. \tag{58}$$

It can be shown that

$$\langle fw_0\rangle = A\frac{z^2(1-z)^2}{24} \tag{59}$$

$$\langle fw_1\rangle = A\frac{z^2(1-z)^2}{55440}\left[-\frac{Q}{288}g_2(z)+\frac{GA}{144}g_3(z)\right] \tag{60}$$

$$\langle f\xi_1\rangle = A\left\{-\left[\tfrac{1}{14}g_4(z)+\frac{Q}{10080}g_5(z)\right]+\frac{GA}{720}g_6(z)\right\} \tag{61}$$

$$R_1 = R_{c1}-\frac{(720+Q)}{4158}AG+\frac{A^2E}{252} \tag{62}$$

$$R_{c1} = \frac{4080}{77}-\frac{(7720+Q)^2}{216216} \tag{63}$$

$$E = \langle(\nabla f)^4\rangle \tag{64}$$

$$G = \langle f(\nabla f)^2\rangle \tag{65}$$

where G is equal to zero for rolls and 1/6 for hexagons and E is equal to one for rolls. The eigenvalue R just past the marginal state assumes the form:

$$R = R_{c0} + R_{c1}\frac{a^2}{Q} + O\left(\frac{a^4}{Q}\right). \quad (66)$$

For cellular convection of the long wavelength rolls, $G = 0$ and $E = 1$. Equation (54) gives the finite amplitude as a function of eigenvalue:

$$A^2 = 252(R_1 - R_{c1}). \quad (67)$$

The finite amplitude behaves like $(R_1 - R_{c1})^{1/2}$ instead of $(R - R_c)^{1/2}$. For cellular convection of the long wavelength other than rolls,

$$A = \frac{126}{E}\left\{GB \pm \left[G^2B^2 + (R_1 - R_{c1})\frac{E}{63}\right]^{1/2}\right\} \quad (68)$$

where $B = (720+Q)/4158$. Subcritical instability is possible, provided $R_1 - R_{c1} < 0$ or

$$-AGB + \frac{A^2E}{252} < 0. \quad (69)$$

Since $G = 0$ for rolls, subcritical instability does not exist. For cellular patterns with G being nonzero, subcritical instability may occur, provided $0 < A < (252/E)GB$.

The perturbed fields for ξ' and u'_z are

$$\xi' = Af(x,y)\left\{1 - a^2\left[\tfrac{1}{14}g_4(z) + \frac{Q}{10080}g_5(z)\right] + O(a^4)\right\} \quad (70)$$

$$u'_z = Af(x,y)\{a^2[\tfrac{1}{24}z^2(1-z^2)] + O(a^4)\}. \quad (71)$$

If we substitute equations (62) and (63) into equation (52), the gradient of the horizontally averaged light composition, due to the nonlinear effects, becomes

$$\frac{\partial\bar{\xi}}{\partial z} = A^2a^2\frac{z^2(1-z)^2}{24} + O(a^4). \quad (72)$$

For cellular convection of rolls, we have

$$\frac{\partial\bar{\xi}}{\partial z} = \tfrac{21}{2}(R_1 - R_{c1})a^2z^2(1-z)^2 + O(a^4). \quad (73)$$

The net result of light compositional gradient in non-dimensional form, due to both non-convective state and nonlinear convective state, is derived as

$$\frac{\partial\xi}{\partial z} = Q[1 - R(1-z)] + A^2a^2\frac{z^2(1-z)^2}{24} + O(a^4). \quad (74)$$

The first term is associated with the diffusion of the light material released at the lower boundary, its effect alone will result in a destabilizing compositional effect. The second term is associated with the compositional convection, its effect alone will result in a stabilizing compositional effect.

CONCLUSIONS

The main purpose of the present study is to analyze the linear and nonlinear behaviors of cellular convective and morphological instabilities of a fluid layer of a binary alloy, cooled from above and consequently frozen at bottom. The released light material, resulting from the freezing effect, is diffused by pressure and compositional gradient. As a result of a small cooling rate and a large thermal diffusivity, the effect of thermal buoyancy is insignificant, compared with that of compositional buoyancy. The necessary condition for a possible compositional convection requires $R > 1$. The strength of compositional convection depends on three factors: the cooling rate, the released light material and the material diffusion. The principle of exchange of stabilities is valid and convective instability sets in stationarily. To the second-order approximation, critical eigenvalues and wavenumbers for cellular convective modes of long wavelengths with $a^2 \ll 1$, $S \gg 1$ and $q = O(1)$ are

$$R = \left(2 + \frac{1440}{Q}\right)\left(1 + \frac{1440}{QSa^2}\right) + \left[\frac{4080}{77} - \frac{(720+Q)^2}{216216}\right]\frac{a^2}{Q} + O\left(\frac{a^4}{Q}\right)$$

which gives the critical wavenumber as

$$a_c^2 = \left\{1440\left(2 + \frac{1440}{Q}\right)\bigg/ S\left[\frac{4080}{77} - \frac{(720+Q)^2}{216216}\right]\right\}^{1/2}.$$

In the limit $S \to \infty$, the critical values reduce to

$$R_c = \left(2 + \frac{1440}{Q}\right)$$

$$a_c^2 = 0$$

and those of short wavelengths with $a^2 \gg 1$, $Q \gg Q^*$ and $q = O(1)$ are

$$R_c - 1 = 3.0304Q^{-1/5} + 23.705Q^{-2/5} - 30.187Q^{-3/5}$$

$$a_c^2 = 14.847 - 2.2877Q^{1/5} + 0.2803Q^{2/5}$$

provided $Q(R-1)/a^4 = O(1)$.

For morphological modes of short wavelengths with $Q(R-1)/a^4 = o(1)$, the critical values are

$$R_c = (1+S)\left\{1 + \left[\frac{q}{2Q(1+s)}\right]^{1/3}\right\} a_c^2 = \left[\frac{Q(1+S)}{2q}\right]^{2/3}.$$

In the limit $q \to 0$, at which case the effect of surface tension becomes neglected, the critical values reduce to

$$R_c = 1 + S \quad \text{at} \quad a_c^2 \to \infty.$$

Nonlinear analysis reveals that disturbances of the finite amplitude are, just past the marginal state, behave like $(R - R_c)^{1/2}$. For cellular convection of long wavelength, subcritical instability is possible for cellular modes other than rolls, provided $0 < A <$

$252GB/E$. The net result of light compositional gradient, due to both non-convective state and nonlinear convective state is

$$\frac{\partial \xi}{\partial z} = Q[1-R(1-z)] + A^2 a^2 \frac{z^2(1-z)^2}{24} + O(a^4).$$

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