

Convective instability of a spherical fluid inclusion

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When a vertical temperature gradient is applied to a large solid containing a spherical fluid inclusion, the temperature in the fluid is a function only of height. The stability of this fluid against convection is investigated and it is found that the principle of exchange of stabilities applies. The linear differential system governing stability is then solved; the results show that the thermal conductivity of the surrounding solid is always stabilizing and that the most unstable mode is the first asymmetric mode, for which the critical Rayleigh number is given. The energy method can be applied, with due modifications to account for heat conduction in the surrounding solid. The same mathematical governing differential system would then be obtained, giving the same number for the upper bound of the Rayleigh numbers below which the fluid is stable. This number is then truly critical: The fluid is stable or unstable according to whether the Rayleigh number is below or above it, whatever the magnitude of the disturbance. The results are discussed in the context of the movement of the spherical inclusion in a soluble solid. The greater instability of the asymmetric mode indicates that when instability occurs, the fluid inclusion will have a sidewise component, which is greater for a greater supercritical Rayleigh number. The effect of double diffusion is also discussed.

I. INTRODUCTION

Fluid inclusions in a soluble solid and their movement when a temperature gradient is present constitute an interesting subject of geological studies. Take, for simplicity, the case of a spherical inclusion of a liquid of thermal conductivity k and radius a imbedded in a soluble solid of density ρ_s and thermal conductivity k_s . Let there be a vertical temperature gradient β_s in the solid, disturbed only by the presence of the liquid inclusion. If we assume that there is no flow in the liquid, the temperature \bar{T}_s in the solid and the temperature \bar{T} in the fluid must satisfy the Laplace equation, so that

$$\nabla^2 \bar{T}_s = 0 \quad \text{and} \quad \Delta^2 \bar{T} = 0, \quad (1)$$

in which ∇^2 is the Laplacian operator, which in spherical coordinates (r, θ, Φ) is

$$\nabla^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial}{\partial \theta} \right) + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2}{\partial \Phi^2}. \quad (2)$$

The polar axis of the spherical coordinates is directed vertically upward. The boundary conditions at the spherical surface are

$$k_s \frac{\partial \bar{T}_s}{\partial r} = k \frac{\partial \bar{T}}{\partial r} \quad \text{and} \quad \bar{T}_s = \bar{T} \quad \text{at} \quad r = a. \quad (3)$$

In addition, \bar{T} must be regular at $r = 0$ and \bar{T}_s must approach $\beta_s x_3$ far away from the sphere, x_3 being the vertical one of the Cartesian coordinates (x_1, x_2, x_3) , with origin at the center of the sphere, which is also the origin of the spherical coordinates.

It can be immediately verified that (with T_0 denoting a constant)

$$\bar{T}_s = \beta_s x_3 + \frac{\beta_1 a^3}{2} \frac{x_3}{r^3} + T_0 \quad \text{and} \quad \bar{T} = \beta x_3 + T_0 \quad (4)$$

satisfy (1), the regularity condition at the origin for \bar{T} , and the condition on \bar{T}_s at infinity. Since

$$x_3 = r \cos \theta,$$

the conditions (3) demand

$$k_s (\beta_s - \beta_1) = k\beta \quad \text{and} \quad \beta_s + \beta_1/2 = \beta, \quad (5)$$

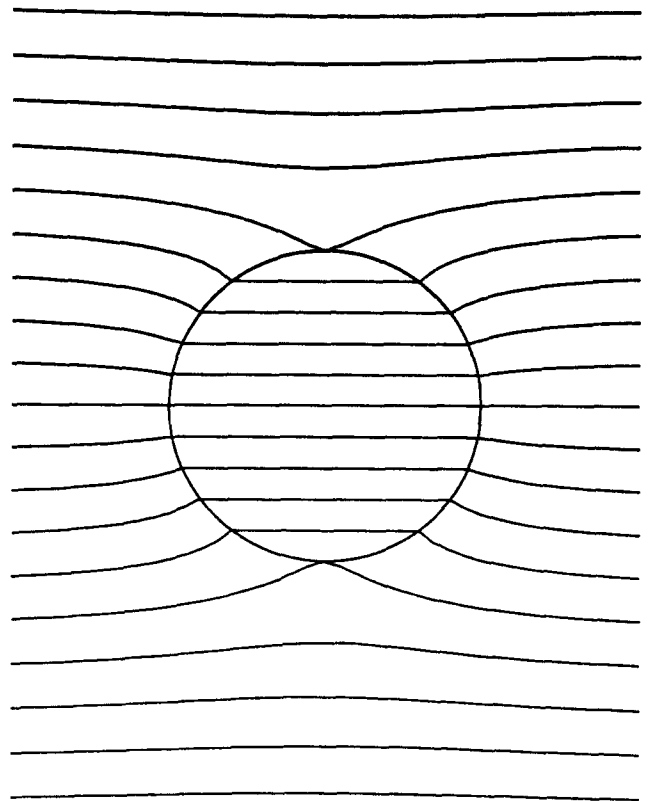


FIG. 1. Isotherms showing linear temperature distribution in the spherical fluid inclusion.

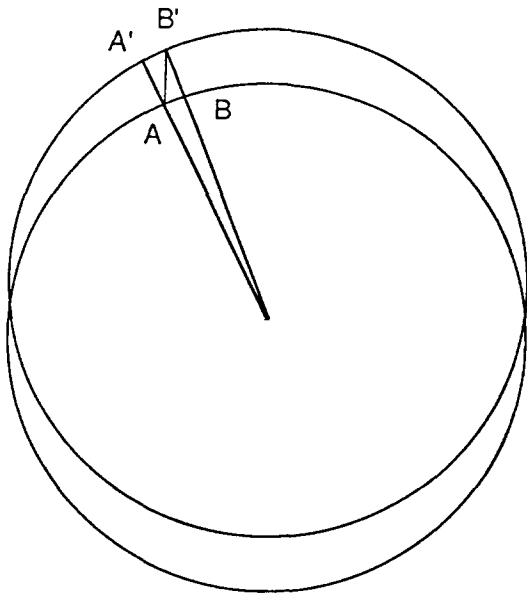


FIG. 2. Sketch showing that erosion causes point A to move to A' and B to B, resulting in the apparent vertical movement of A to A'.

which give

$$\beta_1 = [(2\lambda - 2)/(2\lambda + 1)]\beta_s, \quad \beta = [3\lambda / (2\lambda + 1)]\beta_s, \quad (6)$$

where

$$\lambda = k_s/k. \quad (7)$$

The interesting part of the solutions (4) is that (Fig. 1) the temperature \bar{T} in the fluid is a linear function of x_3 , so that ordinary thermal convection need not arise. But if the solid is soluble in the liquid, there must be mass transfer from the hotter hemisphere to the colder one and that will push the fluid up (if β_s is positive). We can calculate the velocity of the liquid by assuming the solution is saturated everywhere and that the saturation concentration (in mass per unit volume) c is related to \bar{T} , in the temperature range experienced by the fluid, by

$$c = \gamma(\bar{T} - T_0) + c_0. \quad (8)$$

If the mass diffusivity of the substance of the solute is κ_c , its mass is transported downward at a rate of $\kappa_c (\partial c / \partial x_3)$ (mass per unit area per unit time). The speed with which the spherical boundary moves upward is then ($\rho_s =$ density of the solid)

$$u_3 = \frac{\kappa_c}{\rho_s} \frac{\partial c}{\partial x_3} = \frac{\kappa_c \beta \gamma}{\rho_s}. \quad (9)$$

The reader may verify that this conclusion is reached whether the simple argument of vertical transport is used, as here, or whether the *radial* erosion or deposition of mass at the spherical boundary is calculated from $\kappa_c (\partial c / \partial r)$. See Fig. 2.

The fluid, then, is constrained to move upward with the same velocity u_3 everywhere. The heat equations will then have to include the effects of this velocity and a second approximation for both the temperature and the velocity (for the fluid) fields has to be made. However, u_3 is very small, or

the relevant Reynolds and Péclet numbers are very small, so that (9) gives the velocity of the fluid in the same way that the Stokes law gives the fall velocity of a small sphere in a viscous fluid at small Reynolds numbers. Indeed, (9) is a kind of an inverse Stokes law, since it gives the velocity of a liquid sphere moving through a solid.

The erosion and deposition of solute give rise to heat sinks and sources, respectively, because of the absorption or release of latent heat. The effect of these heat sinks and sources can be taken into account quite simply. From the second equation in (4) one obtains that the normal gradient of \bar{T} is $\beta \cos \theta$. The solute dissolved (or deposited) per unit time and per unit area is $\kappa_c \beta \gamma \cos \theta$. The strength of the heat sink per unit area of the boundary is then $m\beta \cos \theta$, where

$$m = L\kappa_c \gamma,$$

with L the latent heat per unit mass. Still using (4), one replaces conditions (3) by

$$k_s \frac{\partial \bar{T}_s}{\partial r} = k \frac{\partial T}{\partial r} + m\beta \cos \theta, \quad \bar{T}_s = \bar{T} \text{ at } r = a.$$

This gives, instead of (5),

$$k_s (\beta_s - \beta_1) = k\beta + m\beta \text{ and } \beta_s + \beta_1/2 = \beta,$$

and instead of (6),

$$\beta_1 = \frac{2(\lambda - 1 - mk^{-1})}{2\lambda + 1 + mk^{-1}} \beta_s, \quad \beta = \frac{3\lambda}{2\lambda + 1 + mk^{-1}} \beta_s.$$

With the β so determined, (9) continues to give the speed of movement of the liquid sphere. It can be seen from (9) and the above equation giving β that the temperature gradient inside the sphere and its speed of movement decreases as the latent heat (and therefore m) increases, as is to be expected.

The question of the stability of the temperature stratification in the fluid then naturally arises, either when β is negative, or if the effect of double diffusion is considered, even if β is positive. As is well known, the problem of nonoscillatory instability when double diffusion is considered can be reduced to that of the simpler case of ordinary thermal-convective instability. When the fluid is unstable, the movement of the fluid inclusion in the solid may be much faster, depending on the Rayleigh number, and may not even be vertical. This rather intriguing situation motivates this study. We shall neglect the effect of the u_3 given by (9) on the stability calculations, since that effect is very small, and we shall concentrate on thermal instability (i.e., without considering double diffusion) first.

II. EQUATIONS GOVERNING THE STABILITY PROBLEM

The mean temperature fields in the solid and the fluid are given by (4), and the mean density ρ_m of the fluid is given by

$$\rho_m = \rho_0 [1 - \alpha(\bar{T} - T_0)], \quad (10)$$

where α is the coefficient of thermal expansion and ρ_0 the density of the fluid at $x_3 = 0$. The mean pressure gradient is in the vertical direction and is given by

$$\frac{\partial p_m}{\partial x_3} = -\rho_0 g [1 - \alpha(\bar{T} - T_0)], \quad (11)$$

where p_m is the mean pressure and g the gravitational accel-

eration. If T_s denotes the temperature in the solid and T that in the fluid, and if ρ and p denote, respectively, the density and pressure of the fluid, we have

$$T_s = \bar{T}_s + T'_s, \quad T = \bar{T} + T',$$

$$\rho = \rho_m + \rho', \quad p = p_m + p',$$

where the primes indicate perturbation quantities. Since

$$\rho = \rho_0[1 - \alpha(T - T_0)], \quad (12)$$

we have

$$\rho' = -\alpha T' \rho_0. \quad (13)$$

We shall consider the temperature differences $T_s - T_0$, $T - T_0$, $\bar{T}_s - T_0$, and $\bar{T} - T_0$, and we shall express these as well as T'_s and T' in units of βa . Furthermore, we shall measure time t in units of a^2/κ , and use a as the length scale for x_1, x_2 , and x_3 . Velocities will be measured in units of κ/a , and pressure in units of $\rho_0 \kappa \nu/a^2$, where ν is the kinematic viscosity. Then all the questions will be in dimensionless terms. For instance, the mean temperature distributions in the solid is now given in dimensionless terms by

$$\bar{T}_s - T_0 = \frac{3\lambda}{2\lambda + 1} x_3 + \frac{\lambda - 1}{3\lambda} \frac{x_3}{r^3}, \quad (14)$$

and that in the fluid by

$$\bar{T} - T_0 = x_3. \quad (15)$$

The velocity perturbation is denoted by u_i ($i = 1, 2, 3$) measured in units of κ/a . The usual linearization procedure then filters out the mean quantities in the governing equations except where they are multiplied by a perturbation quantity. If the Boussinesq approximation is adopted, the dimensionless linearized equations of motion are

$$\frac{1}{\text{Pr}} \frac{\partial u_i}{\partial t} = -\frac{\partial p'}{\partial x_i} - R T' \delta_{i3} + \nabla^2 u_i \quad (i = 1, 2, 3), \quad (16)$$

where δ_{i3} is a Kronecker delta, ∇^2 is the Laplacian operator in Cartesian coordinates, and

$$R = -g\alpha\beta a^4/\kappa\nu \quad \text{and} \quad \text{Pr} = \nu/\kappa \quad (17)$$

are, respectively, the Rayleigh number and the Prandtl number. (For unstable modes β is negative.) The equation of continuity is

$$\frac{\partial u_i}{\partial x_i} = 0. \quad (18)$$

The heat equation is, for the liquid,

$$\frac{\partial T'}{\partial t} + u_3 = \nabla^2 T' \quad (19)$$

and, for the solid, with $\hat{\lambda} = \kappa_s/\kappa$ ($\kappa_s =$ thermal diffusivity of the solid),

$$\frac{\partial T'_s}{\partial t} = \hat{\lambda} \nabla^2 T'_s. \quad (20)$$

From (16) and (18) one obtains

$$\nabla^2 p' = -R \frac{\partial T'}{\partial x_3}. \quad (21)$$

III. BOUNDARY CONDITIONS

The boundary conditions at the spherical surface are

$$u_i = 0 \quad \text{at} \quad r = 1, \quad (22)$$

$$T' = T'_s \quad \text{at} \quad r = 1, \quad (23)$$

$$\frac{\partial T'}{\partial r} = \lambda \frac{\partial T'_s}{\partial r} \quad \text{at} \quad r = 1. \quad (24)$$

These demand, respectively, no slip at the boundary, continuity of temperature, and continuity of heat flux. (Replace λ by $\lambda/(1 + mk^{-1})$ to include the effect of L .) Further,

$$T'_s = 0 \quad \text{at} \quad r = \infty \quad (25)$$

and

$$T', u'_i, \quad \text{and} \quad p' \quad \text{must be regular at} \quad r = 0. \quad (26)$$

IV. NONOSCILLATION OF CONVECTION

Since there are no time-dependent coefficients in all the linear equations involved, one can assume that all perturbation quantities contain the exponential factor $\exp(\sigma t)$, where

$$\sigma = \sigma_r + i\sigma_i.$$

The fluid is therefore stable, neutrally stable, or unstable if σ_r is negative, zero, or positive, respectively.

Multiplying (16) by u_i^* , where the asterisk indicates the complex conjugate, and integrating over the fluid domain V , we have

$$\sigma K = -\text{Pr} Q - \text{Pr} R \int T' u_3^* dV, \quad (27)$$

where

$$K = \int u_i u_i^* dV, \quad Q = \int \frac{\partial u_i}{\partial x_j} \frac{\partial u_i^*}{\partial x_j} dV. \quad (28)$$

Multiplying (19) by T'^* and (20) by $(\lambda/\hat{\lambda}) T'_s^*$, integrating over V and the solid domain V_s , respectively, and adding the results, using (23) and (24) when necessary, one has

$$\sigma(H + H_s) = -S - S_s - \int T'^* u_3 dV, \quad (29)$$

where

$$H = \int |T'|^2 dV, \quad H_s = \frac{\lambda}{\hat{\lambda}} \int |T'_s|^2 dV_s, \quad (30)$$

$$S = \int |\text{grad } T'|^2 dV, \quad (31)$$

$$S_s = \lambda \int |\text{grad } T'_s|^2 dV_s.$$

Multiplying (29) by $-R \text{Pr}$, taking the complex conjugate of the result, and adding (27) to it, one has

$$\sigma K = -\text{Pr} R \sigma^* (H + H_s) = -\text{Pr} Q + \text{Pr} R (S + S_s). \quad (32)$$

The real and imaginary parts of (32) are

$$\sigma_r [K - \text{Pr} R (H + H_s)] = -\text{Pr} Q + \text{Pr} R (S + S_s), \quad (33)$$

$$\sigma_i [K + \text{Pr} R (H + H_s)] = 0, \quad (34)$$

in which, one recalls that all the quantities or numbers represented by capital letters are real and positive. If $\sigma_i \neq 0$, (34) shows that R is negative and then (33) shows that σ_r is

negative. Hence for instability or neutral stability, σ_i must be zero.

V. ANALYSIS OF THE PROBLEM OF LINEAR STABILITY

For neutral stability, then, we shall take σ to be zero. This greatly simplifies the calculations. First, the thermal boundary condition for the fluid alone at the spherical surface can now be formulated.

Equation (20) is now

$$\nabla^2 T'_s = 0, \quad (35)$$

where ∇^2 is given by (2). The elemental solution of (35), nonsingular at infinity, is

$$T'_s = r^{-n-1} P_n^m(\cos \theta) \cos m\phi, \quad (36)$$

where P_n^m is Legendre's associated functions of the first kind. (If $m = 0$, they are just the Legendre polynomials.) For this solution,

$$\frac{\partial T'_s}{\partial r} = -(n+1)T'_s \text{ at } r = 1. \quad (37)$$

Then (23) and (24) give

$$\frac{\partial T'}{\partial r} = -(n+1)\lambda T' \text{ at } r = 1, \quad (38)$$

which allows the separation of the problem for the fluid from the heat equation for the solid.

We shall, for simplicity, write

$$h = RT' \text{ and } h_s = RT'_s. \quad (39)$$

Then, for neutral stability, (16) becomes

$$\nabla^2 u_i = \frac{\partial p'}{\partial x_i} + h\delta_{i3} \quad (40)$$

and (19), (21), and (20) assume the forms, respectively,

$$\nabla^2 h = Ru_3, \quad (41)$$

$$\nabla^2 p' = -\frac{\partial h}{\partial x_3}, \quad (42)$$

$$\nabla^2 h_s = 0. \quad (43)$$

The thermal boundary condition (38) now has the form

$$\frac{\partial h}{\partial r} = -(n+1)\lambda h \text{ at } r = 1. \quad (44)$$

Since the boundary is spherical, it is natural to use spherical coordinates (r, θ, Φ) and the corresponding velocity components (u, v, w) . Then, for neutral stability, the non-oscillation of the disturbance shown in Sec. IV allows us to write the Navier-Stokes equations

$$0 = -\frac{\partial p'}{\partial r} - h \cos \theta + \left(\nabla^2 u - \frac{2u}{r^2} - \frac{2}{r^2} \frac{\partial v}{\partial \theta} - \frac{2v \cot \theta}{r^2} - \frac{2}{r^2 \sin \theta} \frac{\partial w}{\partial \phi} \right), \quad (45a)$$

$$0 = -\frac{1}{r} \frac{\partial p'}{\partial \theta} + h \sin \theta + \left(\nabla^2 v + \frac{2}{r^2} \frac{\partial u}{\partial \theta} - \frac{v}{r^2 \sin^2 \theta} - \frac{2 \cot \theta}{r^2 \sin \theta} \frac{\partial w}{\partial \phi} \right), \quad (45b)$$

$$0 = -\frac{1}{r \sin \theta} \frac{\partial p'}{\partial \phi} + \left(\nabla^2 w - \frac{w}{r^2 \sin^2 \theta} + \frac{2}{r^2 \sin \theta} \frac{\partial u}{\partial \phi} + \frac{2 \cot \theta}{r^2 \sin \theta} \frac{\partial v}{\partial \phi} \right), \quad (45c)$$

where ∇^2 is given by (2). The equation of continuity is

$$\frac{1}{r^2} \frac{\partial}{\partial r}(r^2 u) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta}(v \sin \theta) + \frac{1}{r \sin \theta} \frac{\partial w}{\partial \phi} = 0. \quad (46)$$

In view of (46), the last three terms of (45a) can be replaced by

$$\frac{2}{r^3} \frac{\partial}{\partial r}(r^2 u) = \frac{4u}{r^2} + \frac{2}{r} \frac{\partial u}{\partial r}, \quad (47)$$

and a simple calculation shows that (45a) reduces to the form

$$\frac{\partial p'}{\partial r} + h \cos \theta = \frac{1}{r} \nabla^2(ru). \quad (48)$$

The velocity components u and v are related to u_3 by

$$u_3 = u \cos \theta - v \sin \theta \quad (49)$$

and in (42) one has

$$\frac{\partial}{\partial x_3} = \cos \theta \frac{\partial}{\partial r} - \frac{\sin \theta}{r} \frac{\partial}{\partial \theta}. \quad (50)$$

One has to solve (41), (42), (48), (45b), and (45c), with (49) and (50) used in (41) and (42). Apart from regularity for all quantities at $r = 0$, the boundary conditions are (44) and

$$u = \frac{\partial u}{\partial r} = 0 \text{ at } r = 1, \quad (51)$$

$$v = 0 = w \text{ at } r = 1. \quad (52)$$

The condition on $\partial u / \partial r$ in (51) arises from (46). Once (46) is satisfied on $r = 1$, and (51) and (52) guarantee that satisfaction, it is satisfied everywhere in the fluid, since (42) has been derived on the basis of (18). Indeed, (40) gives

$$\nabla^2 \frac{\partial u_i}{\partial x_i} = \nabla^2 p' + \frac{\partial h}{\partial x_3}, \quad (53)$$

so that if (42) is satisfied one has

$$\nabla^2 \frac{\partial u_i}{\partial x_i} = 0. \quad (54)$$

If $\partial u_i / \partial x_i$ is nonsingular within $r = 1$, and is zero on $r = 1$, it is zero everywhere inside the spherical surface, as is well known in potential theory. Alternatively, one can use (46) to replace (45b) or (45c) in the differential system to be solved.

It has been necessary to present the governing equations in both Cartesian and spherical coordinates because the simplest approach involves the use of both coordinate systems. This approach is as follows.

(a) Expand h in some suitable basis functions satisfying (44).

(b) Solve (42) for p' , retaining the undetermined complementary solution.

(c) Solve (48) for u , using the complementary solution available from step (b) to help satisfy (51).

(d) Solve the third equation of (40) for u_3 .

(e) Go to (41) to get the determinant whose vanishing gives R .

(f) Solve (49) for v and (46) for w , if v and w are desired.

We shall now use this procedure to solve the problem.

First, we set forth a few well-known formulas regarding associated Legendre functions of the first kind (including Legendre polynomials), and some functions involving Bessel functions of half-orders. The solution of the Laplace equation in spherical coordinates by separation of variables is of the form

$$f(r)P_n^m(\mu)\cos m(\phi + \epsilon), \quad (55)$$

where

$$f(r) = r^n \text{ or } r^{-n-1}, \quad \mu = \cos \theta, \quad (56)$$

ϵ is a phase angle which can be set to zero, $P_n^m(\mu)$ is an associated Legendre function of the first kind, and m and n are integers with $m \leq n$. (When $n < m$ the function P_n^m is zero.) The following formulas will be useful:

$$(2n+1)\mu P_n^m = (n-m+1)P_{n+1}^m + (n+m)P_{n-1}^m, \quad (57)$$

$$(\mu^2 - 1)\frac{dP_n^m}{d\mu} = \frac{1}{2n+1}(n(n-m+1)P_{n+1}^m - (n+1)(n+m)P_{n-1}^m). \quad (58)$$

We shall use the basis functions $F_n(\alpha_n r)$ defined by

$$\left(\frac{d^2}{dr^2} + \frac{2}{r}\frac{dr}{dr} + \alpha_n^2 - \frac{n(n+1)}{r^2}\right)F_n(\alpha_n r) = 0, \quad (59)$$

the regularity condition at $r = 0$, and, on account of (44),

$$\frac{dF_n(\alpha_n r)}{dr} = -(n+1)\lambda F_n(\alpha_n r) \text{ at } r = 1. \quad (60)$$

For each n , there are infinitely many eigenvalues for α_n , which we denote by α_{nj} ($j = 1, 2, 3, \dots$). It is well known that the solutions of (56) are of the form

$$F_n(\alpha_{nj}r) = J_{n+0.5}(\alpha_{nj}r)/\sqrt{\alpha_{nj}r}, \quad (61)$$

where the J stands for the Bessel function. Near $r = 0$, then, $F_n(\alpha_{nj}r)$ behaves as r^n . The first several functions F_n are given explicitly in the Appendix. An F_n has a finite number of terms for a finite n . It has been used before by Chandrasekhar.¹

From corresponding formulas for Bessel functions, one has

$$\frac{dF_n}{dr} = \frac{\alpha_{nj}}{2n+1}[-(n+1)F_{n+1} + nF_{n-1}], \quad (62)$$

$$\frac{dF_n}{dr} = \frac{n}{r}F_n - \alpha_{nj}F_{n+1} = -\frac{n+1}{r}F_n + \alpha_{nj}F_{n-1}, \quad (63)$$

$$\frac{2n+1}{r}F_n = \alpha_{nj}(F_{n+1} + F_{n-1}). \quad (64)$$

The argument of the functions in (62)–(64) is $\alpha_{nj}r$.

Two other useful formulas, upon the use of (57), (58), (63), and (64) are

$$\begin{aligned} & \frac{\partial}{\partial x_3} P_n^m(\mu)F_n(\alpha_{nj}r) \\ &= \mu P_n^m \frac{dF_n}{dr} + (1-\mu^2) \frac{dP_n^m}{d\mu} \frac{F_n}{r} \\ &= \frac{\alpha_{nj}}{2n+1} [-(n-m+1)P_{n+1}^m F_{n+1} \\ & \quad + (n+m)P_{n-1}^m F_{n-1}], \end{aligned} \quad (65)$$

$$\frac{\partial}{\partial x_3} r^n P_n^m(\mu) = (n+m)r^{n-1}P_{n-1}^m. \quad (66)$$

Equation (65) is gratifying because each term on the right-hand side has the same indices below. That is, P_{n+1}^m is multiplied by F_{n+1} and P_{n-1}^m is multiplied by F_{n-1} . This is a most fortunate situation, as we shall see.

We now expand h as follows:

$$h + \sum_n \sum_{j=1}^{\infty} A_{nj} P_n^m(\mu) F_n(\alpha_{nj}r) \cos m\phi, \quad (67)$$

where for each m , n runs through either odd or even integers not less than m . (For instance, if $m = 0$, n can either be 0, 2, 4, ... or 1, 3, 5, If $m = 1$, n runs through 1, 3, 5, ... , or through 2, 4, 6, That the increment of n is 2 in either series will be clear from the subsequent development.) Then we solve (42) and obtain

$$\begin{aligned} p &= \sum_n \sum_j \frac{A_{nj}}{\alpha_{nj}^2} \left(\frac{\partial}{\partial x_3} P_n^m F_n + B'_{nj} r^{n+1} P_{n+1}^m \right. \\ & \quad \left. + C'_{nj} r^{n-1} P_{n-1}^m \right) \cos m\phi \\ &= \sum_n \sum_j \frac{A_{nj}}{(2n+1)\alpha_{nj}} [-(n-m+1)P_{n+1}^m F_{n+1} \\ & \quad + (n+m)P_{n-1}^m F_{n-1} + B_{nj} r^{n+1} P_{n+1}^m \\ & \quad + C_{nj} r^{n-1} P_{n-1}^m] \cos m\phi, \end{aligned} \quad (68)$$

where

$$(B_{nj}, C_{nj}) = [(2n+1)/\alpha_{nj}] (B'_{nj}, C'_{nj}). \quad (69)$$

The argument for the P functions is μ and that for the F functions is $\alpha_{nj}r$. From (68) we obtain, after some cancellations,

$$\begin{aligned} \frac{\partial p'}{\partial r} + \mu h &= \sum_n \sum_j \frac{A_{nj}}{(2n+1)\alpha_{nj}} \\ & \quad \times \left(\frac{(n-m+1)(n+2)}{r} P_{n+1}^m F_{n+1} \right. \\ & \quad \left. + \frac{(n+m)(n-1)}{r} P_{n-1}^m F_{n-1} \right. \\ & \quad \left. + (n+1)B_{nj} r^n P_{n+1}^m \right. \\ & \quad \left. + (n-1)C_{nj} r^{n-2} P_{n-1}^m \right) \cos m\phi, \end{aligned} \quad (70)$$

where, as in (68), P_{n-1}^m is zero if $n-1 < m$. We now solve (48) and obtain

$$u = \sum_n \sum_j \frac{A_{nj}}{(2n+1)\alpha_{nj}} \left[-\frac{1}{\alpha_{nj}^2} \left(\frac{(n-m+1)(n+2)}{r} \right. \right. \\ \times P_{n+1}^m F_{n+1} + \frac{(n-1)(n+m)}{r} P_{n-1}^m F_{n-1}) \\ \left. \left. + \frac{n+1}{4n+10} B_{nj} r^{n+2} P_{n+1}^m + \frac{n-1}{4n+2} C_{nj} r^n P_{n-1}^m \right. \right. \\ \left. \left. + D_{nj} r^n P_{n+1}^m + E_{nj} r^{n-2} P_{n-1}^m \right] \cos m\phi. \quad (71)$$

The boundary conditions on u in (51) then determine the coefficients:

$$B_{nj} = \frac{(n-m+1)(n+2)(2n+5)}{(n+1)\alpha_{nj}^2} \\ \times [\alpha_{nj} F_n(\alpha_{nj}) - (2n+3)F_{n+1}(\alpha_{nj})], \quad (72)$$

$$D_{nj} = \frac{(n-m+1)(n+2)}{2\alpha_{nj}^2} \\ \times [(2n+5)F_{n+1}(\alpha_{nj}) - \alpha_{nj} F_n(\alpha_{nj})], \quad (73)$$

$$C_{nj} = -\frac{(2n+1)(n+m)}{\alpha_{nj}} F_n(\alpha_{nj}), \quad (74)$$

$$E_{nj} = \frac{(n-1)(n+m)}{2\alpha_{nj}^2} [2F_{n-1}(\alpha_{nj}) + \alpha_{nj} F_n(\alpha_{nj})]. \quad (75)$$

For $n < m+1$ C_{nj} and E_{nj} will not be needed. From (71), it is clear that C_{1j} is not needed even for $m=0$.

We now turn to the third equation in (40), or

$$\nabla^2 u_3 = \frac{\partial p'}{\partial x_3} + h. \quad (76)$$

First,

$$\frac{\partial p'}{\partial x_3} = \sum_n \sum_j \frac{A_{nj}}{(2n+1)\alpha_{nj}} \left(-\frac{n-m+1}{2n+3} \alpha_{nj} [-(n-m+2)P_{n+2}^m F_{n+2} + (n+m+1)P_n^m F_n] + \frac{n+m}{2n-1} \alpha_{nj} \right. \\ \times [-(n-m)P_n^m F_n + (n+m-1)P_{n-2}^m F_{n-2}] + (n+m+1)B_{nj} r^n P_n^m \\ \left. + (n+m-1)C_{nj} r^{n-2} P_{n-2}^m \right) \cos m\phi, \quad (77)$$

in which, as elsewhere, a P function is zero if its subscript is less than its superscript. The argument of the F functions is $\alpha_{nj}r$. Substituting (67) and (77) into (76), and solving it, we obtain

$$u_3 = -\sum_n \sum_j \frac{A_{nj}}{(2n+1)\alpha_{nj}^2} \left(\frac{(n-m+1)(n-m+2)}{2n+3} P_{n+2}^m F_{n+2} + \frac{2(2n+1)(n^2+n-1+m^2)}{(2n-1)(2n+3)} P_n^m F_n \right. \\ \left. + \frac{(n+m)(n+m-1)}{2n-1} P_{n-2}^m F_{n-2} - \frac{(n+m+1)}{4n+6} \alpha_{nj} B_{nj} r^{n+2} P_n^m - \frac{n+m-1}{4n-2} \alpha_{nj} C_{nj} r^n P_{n-2}^m \right. \\ \left. + G_{nj} r^{n+2} P_{n+2}^m + H_{nj} r^n P_n^m + K_{nj} r^{n-2} P_{n-2}^m \right) \cos m\phi. \quad (78)$$

TABLE I. Values of eigenvalues of α_{nj} .

| $n, j \backslash \lambda$ | 0.5 | 1 | 5 | 10.72 | ∞ |
|---------------------------|---------------|---------------|---------------|---------------|-----------|
| 0,1 | 1.165 561 19 | 1.570 796 33 | 2.570 431 56 | 2.855 825 15 | π |
| 0,2 | 4.604 216 78 | 4.712 388 98 | 5.354 031 84 | 5.749 063 38 | 2π |
| 0,3 | 7.789 883 75 | 7.853 981 63 | 8.302 929 18 | 8.694 984 46 | 9.424 76 |
| 1,1 | 2.743 707 27 | π | 4.066 202 10 | 4.286 387 37 | 4.493 40 |
| 1,2 | 6.116 764 26 | 2π | 7.056 819 40 | 7.378 345 88 | 7.725 24 |
| 1,3 | 9.316 615 63 | 9.424 777 96 | 10.059 893 23 | 10.431 622 41 | 10.904 11 |
| 2,1 | 4.067 315 91 | 4.493 409 46 | 5.392 525 84 | 5.585 732 25 | 5.763 45 |
| 2,2 | 7.517 465 12 | 7.725 251 84 | 8.542 952 84 | 8.818 685 90 | 9.095 00 |
| 2,3 | 10.761 984 24 | 10.904 121 66 | 11.625 651 18 | 11.955 908 18 | 12.322 93 |
| 3,1 | 5.311 410 66 | 5.763 459 20 | 6.648 002 74 | 6.826 005 71 | 6.987 93 |
| 3,2 | 8.854 793 54 | 9.095 011 33 | 9.931 192 69 | 10.178 195 06 | 10.417 11 |
| 3,3 | 12.152 991 68 | 12.322 940 97 | 13.090 033 82 | 13.387 971 99 | 13.698 01 |
| 4,1 | 6.513 445 84 | 6.987 932 00 | 7.862 622 03 | 8.030 700 50 | 8.182 55 |
| 4,2 | 10.150 100 30 | 10.417 118 55 | 11.261 903 15 | 11.489 360 84 | 11.704 90 |
| 4,3 | 13.504 450 97 | 13.698 023 15 | 14.491 662 20 | 14.765 382 91 | 15.039 66 |
| 5,1 | 7.688 605 78 | 8.182 561 45 | 9.049 992 85 | 9.211 005 21 | 9.355 81 |
| 5,2 | 11.414 998 45 | 11.704 907 15 | 12.553 804 24 | 12.767 091 90 | 12.966 52 |
| 5,3 | 14.825 518 09 | 15.039 664 71 | 15.849 782 40 | 16.105 060 48 | 16.354 70 |
| 6,1 | 8.844 763 42 | 9.355 812 11 | 10.217 604 84 | 10.373 288 84 | 10.512 83 |
| 6,2 | 12.656 620 94 | 12.966 530 17 | 13.817 276 09 | 14.019 791 29 | 14.207 39 |
| 6,3 | 16.122 315 18 | 16.354 709 64 | 17.175 538 74 | 17.416 383 74 | 17.647 97 |

TABLE II. The critical Rayleigh number R_{cr} for various modes.

| Mode | N, J | λ | | | | |
|-------|--------|-----------|---------|---------|---------|----------|
| | | 0.5 | 1 | 5 | 10.72 | ∞ |
| (i) | 0,3 | 1161 | 1160 | 1158 | 1158 | 1156 |
| | 2,2 | 967 | 1111 | 907 | 1181 | 978 |
| | 2,3 | 938.738 | 959.579 | 994.294 | 1000.92 | 1008.25 |
| | 4,3 | 938.366 | 959.206 | 993.937 | 1000.57 | 1008.25 |
| (ii) | 3,1 | 7855 | 5642 | 3530 | 2618 | 2056 |
| | 5,1 | 7706 | 7281 | 5450 | 4984 | 4584 |
| | 3,2 | 3077 | 3063 | 5957 | 3277 | 2867 |
| | 3,3 | 3026.92 | 3112.36 | 3264.98 | 3299.17 | 3337.55 |
| | 5,3 | 3017.64 | 3102.63 | 3254.72 | 3288.85 | 3327.11 |
| (iii) | 1,1 | 407 | 490 | 660 | 703 | 752 |
| | 3,1 | 407 | 489 | 653 | 685 | 714 |
| | 5,1 | 407 | 491 | 659 | 702 | 749 |
| | 1,2 | 385 | 464 | 648 | 697 | 749 |
| | 3,2 | 384 | 463 | 645 | 697 | 748 |
| | 3,3 | 383.394 | 462.653 | 645.021 | 694.611 | 744.903 |
| | 5,3 | 383.344 | 462.276 | 645.146 | 693.014 | 747.015 |

The coefficients $G, H,$ and K are determined by the condition

$$u_3 = 0 \text{ at } r = 1$$

and are

$$\begin{aligned}
 G_{nj} &= -\frac{(n-m+1)(n-m+2)}{2n+3} F_{n+2}(\alpha_{nj}), \\
 H_{nj} &= -\frac{2(2n+1)(n^2+n-1+m^2)}{(2n-1)(2n+3)} F_n(\alpha_{nj}) \\
 &+ \frac{n+m+1}{4n+6} \alpha_{nj} B_{nj},
 \end{aligned} \tag{79}$$

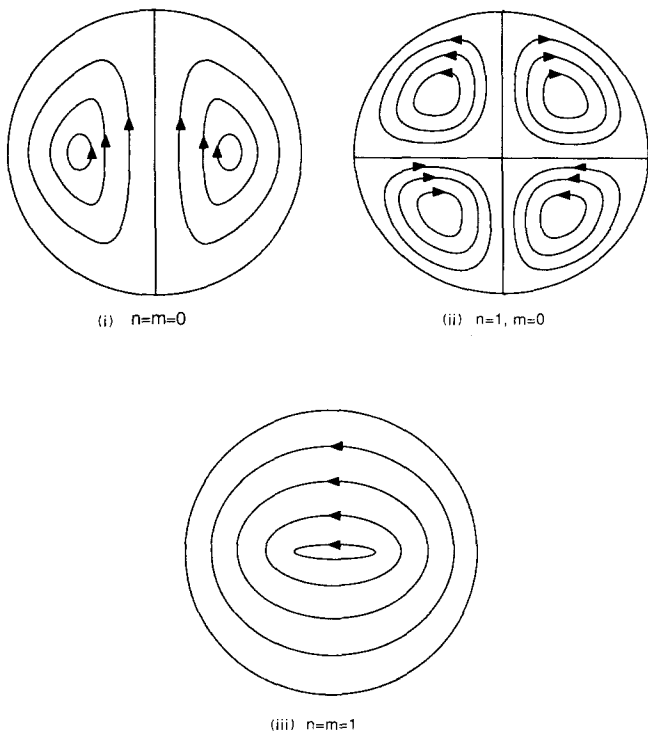


FIG. 3. Flow patterns for the three modes. (i) $m = 0$ even, starting from $n = 0$; (ii) $m = 0, n$ odd, starting from $n = 1$; and (iii) $m = 1, n$ odd, starting from $n = 1$.

$$\begin{aligned}
 K_{nj} &= -\frac{(n+m)(n+m-1)}{2n-1} F_{n-2}(\alpha_{nj}) \\
 &+ \frac{n+m-1}{4n-2} \alpha_{nj} C_{nj}.
 \end{aligned}$$

Now (67) and (78) are substituted into (41) and one seeks to equate the coefficients of $P_n^m(\mu)F_n(\alpha_{nj}r)$ on the two sides of (41). To do this, one needs only to multiply (41) by $r^2 F_n(\alpha_{nj}r)$ and integrate from $r = 0$ to $r = 1$. The associated Legendre functions, appearing as they do in (67) and (78), need only to be sorted out, and no integration with respect to μ is necessary. In this way one obtains a doubly infinite number of linear homogeneous equations in the coefficients A_{nj} . Taking a finite number of them, and requiring that the A 's are not all zero, one obtains a determinant the vanishing of which determines R .

The integration procedure requires the evaluation of certain definite integrals; these are shown in the Appendix.

VI. RESULTS

First, the eigenvalues α_{nj} are determined from (60) and (61). The first two eigenvalues for each n are given in Table I, where $\lambda = 10.72$ is for NaCl and water.

Calculations for the Rayleigh number R were carried out for three modes:

- (i) $m = 0, n = 0, 2, 4, \dots$;
- (ii) $m = 0, n = 1, 3, 5, \dots$;
- (iii) $m = 1, n = 1, 3, 5, \dots$.

The results for the critical R are given in Table II, in which N is the maximum of the values of n taken and J is the maximum of j in a_{nj} . The number of terms with coefficients A_{nj} in (67) taken for calculation is $J(N+2)/2$ if N is even and $J(N+1)/2$ if N is odd. Thus for each mode, the last two lines in Table II give the critical R calculated from a 6×6 and a 9×9 determinant, respectively. The convergence seems quite satisfactory in general, with a discrepancy of less than one part in 300 between the results given by the last successive approximations, even in the worst cases. The flow patterns for the three modes are sketched in Fig. 3.

From Table II it is clear that mode (iii) is the most unstable. This is in agreement with the findings of Hale² and Yih,³ who treated the stability of a thermally stratified fluid in a vertical circular tube, that overturning from one half of the tube to the other half is the most unstable mode. It is also evident from Table II that the effect of the conductivity ratio on the critical R is much greater for mode (iii) than for modes (i) and (ii) on the percentage basis. In general, J has a greater influence on the accuracy of the results than N . The value 5957 for mode (ii) and $(N, J) = 3, 2$ indicates a large truncation error for $\lambda = 5$. Otherwise the R_{cr} values seem systematic. It is fortunate that for mode (iii), which is the most unstable mode, the R_{cr} values are very consistent and rapidly convergent from $J = 2$ onward for all values of λ . Even for $J = 1$ the values of R_{cr} are not very different from their values for greater J , except for $N = 3$ and $\lambda = \infty$ [for which the value of 714 given for R_{cr} seems to have a rather high error because of truncation (in N or J)].

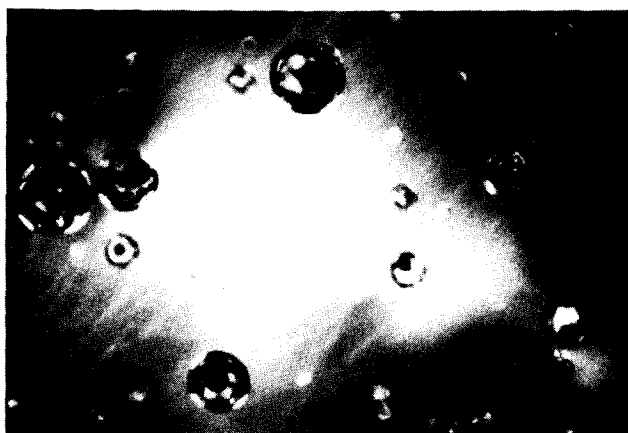


FIG. 4. Oil inclusions in fluorite (CaF_2). (Courtesy of Professor W. Kelly.)

VII. DISCUSSION

It is clear from the results that the critical R increases with λ . To reach this conclusion for modes (i) and (ii) it was necessary to take N and J sufficiently large. (Taking $N = 0$ and $J = 1$ gives the wrong trend.) For mode (iii) even taking $N = 1 = J$ gives the right trend and nearly the correct critical R . The increase of R with λ can also be demonstrated by a rather involved parameter differentiation, and agrees with Hurl *et al.*,⁴ who treated the Bénard problem.

When the Rayleigh number is above 693 in the case of salt and water, including the third-order terms in the governing equations will, in the usual way (although the calculation is now necessarily more complicated), show that the magnitude of the perturbation quantities is of the order of $(R - 693)^{1/2}$. Since the asymmetric mode is most unstable, and since it contains the factor $\sin \phi$ or $\cos \phi$, the deposition or erosion of the solid by the fluid, when the solid is soluble, will have a sidewise component. The greater $R - 693$ is, the more the movement will be sidewise. When instability occurs, then, larger fluid inclusions will move in a cone of a larger vertex angle, if the surrounding solid is soluble. In that case, keeping in mind that the flux is saturated with the solute, and thus the mass concentration of the solute is propor-

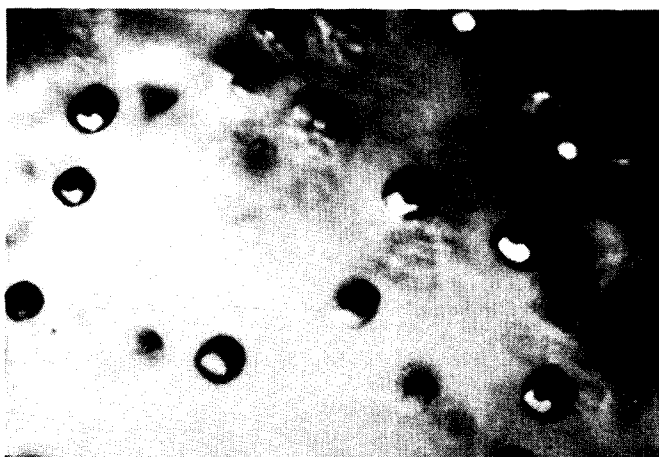


FIG. 5. Aqueous inclusions in synthetic KNO_3 . (Courtesy of Professor W. Kelly.)

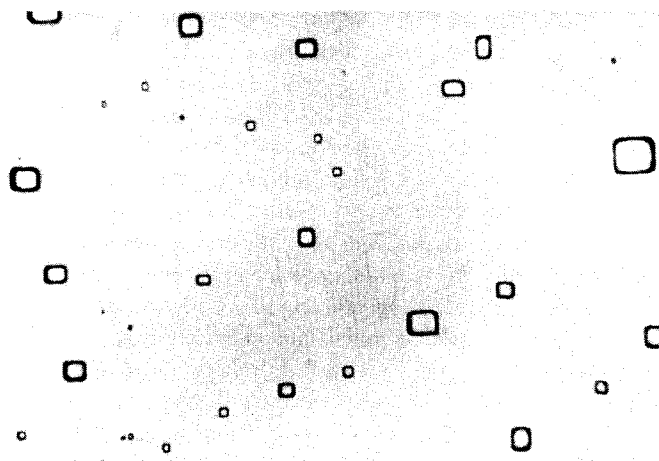


FIG. 6. Aqueous inclusions in NaCl , Windsor, Ontario, Canada. The inclusions imitate salt crystals and are called negative crystals. (Courtesy of Professor W. Kelly.)

tional to h , one obtains the erosion and deposition rates at the boundary from the solution (67). The dominant term in (67) contains the factor (for the most unstable case $m = 1$)

$$P_1^1(\mu) \cos m\phi \text{ or } \sin \theta \cos m\phi.$$

If only this dominant term were present, the spherical inclusion would move in a slanted direction without change of shape. However, the other (less important) terms would cause it slowly to vary its shape.

We have ignored the effect of double diffusion. When that effect is taken into account, the fluid can be unstable even when the temperature gradient β is positive. For neutral stability,

$$\frac{ga^4}{\rho_0\nu} \left(\frac{\beta'}{\kappa'} - \rho_0 \frac{\alpha\beta}{\kappa} \right) = 693,$$

where the first term on the left-hand side is the Rayleigh number for mass diffusion, β' is the salinity gradient, and κ' is the diffusivity of the solute. If the solid is salt (NaCl), κ' is much smaller than κ , and instability can occur at a positive β' much smaller than the value of $-\alpha\beta$ (when β is negative as in the main body of this paper) necessary to bring about thermal instability. Again, when instability occurs with double-diffusive effects taken into account, the fluid will as a whole (slowly) move sidewise as well as vertically.

It may be noted that if the energy method for hydrodynamic stability is employed, the usual calculation, with due modification to take thermal conduction in the solid into account, will produce, upon the application of the calculus of variation of Euler and Lagrange, the *same mathematical* differential system governing the upper bound of the Rayleigh number R below which the fluid is definitely stable, whatever the magnitude of the disturbance. That bound is therefore 693, which is therefore the true critical Rayleigh number: When R is above it the fluid is unstable, and when R is below it the fluid is stable, whatever the magnitude of the disturbance.

Finally, we wish to show a few photographs of fluid inclusions in minerals. Figure 4 shows perfectly spherical inclusions. Unfortunately, they are oil inclusions, in which

the mineral is not soluble. Figure 5 shows nearly spherical shapes, but the crystal planes of the mineral already have an influence on them, while Fig. 6 shows negative crystals, or aqueous inclusions imitating the shape of salt crystals. All these shapes occur in isothermal conditions. We have not been able to find shapes of fluid inclusions in minerals when a thermal gradient had been present for a long time.

In a previous paper, Yih⁵ showed that general ellipsoidal liquid inclusions can move with a constant velocity in a soluble solid provided gravity acts in a direction parallel to any one axis of the ellipsoid. Thus infinitely many permanent shapes are possible if surface energy associated with crystals is ignored. The photographs shown here seem to indicate, for isothermal conditions at any rate, a preferred asymptotic (with time) shape. For isothermal conditions Cline and Anthony⁶ showed a rounded-cubic shape of an aqueous inclusion in KCL after seven years in equilibrium. Thus there seems to be a preferred shape. Cline and Anthony⁶ also calculated the shapes (each of which is presumably unique under the specified conditions) of liquid inclusions, which depend, among other things, on the volume of the liquid. But Cline and Anthony⁶ ignored the temperature distribution in both the solid and the liquid, and this distribution is, as shown by Yih,⁵ important for determining the speed of migration of liquid inclusions and relevant to the permanence of their shapes. Yih,⁵ on the other hand, ignored the effect of surface energy in crystals and did not touch the question of the uniqueness and determination of the permanent shape of a liquid inclusion in a soluble solid with a temperature gradient. This question remains open.

ACKNOWLEDGMENTS

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APPENDIX: $F_n(x)$ AND DEFINITE INTEGRALS

The first several functions $F_n(x)$ are

$$F_0(x) = (2/\pi)^{1/2}(\sin x/x),$$

$$F_1(x) = \left(\frac{2}{\pi}\right)^{1/2} \frac{1}{x} \left(\frac{\sin x}{x} - \cos x\right),$$

$$F_2(x) = -\left(\frac{2}{\pi}\right)^{1/2} \frac{1}{x} \left[\frac{3 \cos x}{x} + \left(1 - \frac{3}{x^2}\right) \sin x\right],$$

$$F_3(x) = \left(\frac{2}{\pi}\right)^{1/2} \frac{1}{x} \left[\left(1 - \frac{15}{x^2}\right) \cos x - \left(\frac{6}{x} - \frac{15}{x^3}\right) \sin x\right],$$

$$F_n(x) = \frac{n-1}{x} F_{n-1}(x) - \frac{dF_{n-1}(x)}{dx}.$$

The following definite integrals have been needed in the calculations:

$$I_0(\alpha) = \int_0^1 r^4 F_0(\alpha r) dr = \frac{1}{\alpha^2} \left[2F_0(\alpha) - \left(\frac{6}{\alpha} - \alpha\right) F_1(\alpha) \right],$$

$$I_n(\alpha) = \int_0^1 r^5 F_n(\alpha r) dr \\ = (1/\alpha) [(2n+3)I_{n-1}(\alpha) - F_{n-1}(\alpha)];$$

$$L_0(\alpha) = \int_0^1 r^2 F_0(\alpha r) dr = \frac{1}{\alpha} F_1(\alpha),$$

$$L_n(\alpha) = \int_0^1 r^{n+2} F_n(\alpha r) dr \\ = (1/2) [(2n+1)L_{n-1}(\alpha) - F_{n-1}(\alpha)];$$

$$I_0(\alpha, \beta) \\ = \int_0^1 r^2 F_0(\alpha r) F_0(\beta r) dr \\ = [1/\alpha\beta(2\pi)^{1/2}] [F_0(\alpha - \beta) - F_0(\alpha + \beta)],$$

$$I_n(\alpha, \beta) \\ = \int_0^1 r^2 F_n(\alpha r) F_n(\beta r) dr \\ = \alpha^{-1} [\beta I_{n-1}(\alpha, \beta) - F_{n-1}(\alpha) F_n(\beta)];$$

$$\int_0^1 r^2 F_n^2(\alpha r) dr = I_n(\alpha, \alpha);$$

$$\int_0^1 r^2 F_0^2(\alpha r) dr = \frac{1}{2\alpha^3\pi} (2\alpha - \sin 2\alpha);$$

$$\int_0^1 r^2 F_1^2(\alpha r) dr = \frac{1}{2\alpha\pi^3} \left(\sin^2 \alpha + 2\alpha - \frac{4 \sin^2 \alpha}{\alpha} \right);$$

$$\int_0^1 r^2 F_2^2(\alpha r) dr \\ = (1/2\pi\alpha^3) [2\alpha - \sin 2\alpha - 6\alpha^{-3}(\alpha^2 - 2\alpha \sin 2\alpha \\ + \alpha^2 \cos 2\alpha + 1 - \cos 2\alpha)];$$

$$\int_0^1 r^2 F_3^2(\alpha r) dr \\ = (1/2\pi\alpha^3) (2\alpha + \sin 2\alpha - 6\alpha^{-5}(2\alpha^4 + 5\alpha^2 + 15) \\ + 6\alpha^{-5}[(2\alpha^4 - 25\alpha^2 + 15)\cos 2\alpha \\ - (10\alpha^3 - 30\alpha)\sin 2\alpha]).$$

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