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STEADY THERMOCAPILLARY CONVECTION CELLS IN LIQUID DROPS

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ABSTRACT

A nominally spherical drop is used as a model for a theoretical analysis of thermocapillary convection and for estimates of convective flow rates in "levitated" melts at zero-g. Since in practice temperature fields and the resulting convective flow can be more complicated than the simple vertical temperature gradient and the single vortex ring, respectively, the convective flow arising from a general steady-state temperature field is analyzed. Expressions for the components of a steady velocity field are obtained by adapting the analytical method of Miller and Scriven. The vortex rings are illustrated by means of typical streamlines for the simpler, more symmetric, temperature fields. The circulation time is introduced as a measure of the rate of circulation in a convection cell and typical values are given for several materials.

INTRODUCTION

When buoyant forces are negligible, such as in a space laboratory, convective flows may still occur in a liquid as the result of gradients in the surface--or interfacial tension. These convective flows are commonly called the Marangoni effect although the Marangoni effect includes both this phenomenon of convection and the phenomenon of the deformation of a free liquid surface (Ref. 1). Among the causes of gradients in the surface tension are gradients in the concentration and in the temperature along the surface of a liquid.

Concentration and temperature gradients may not be completely eliminated in many processes, and in some instances, their presence may be necessary to produce the convection which is desired. The growth of crystals from a melt is a process in which the elimination of convection in the melt is desirable because convection produces non-uniform growth conditions, and, thereby, an increase in the number of dislocations in the crystal. The evaporative purification of a levitated melt is a process in which the opposite result is desired: rapid convection is important here because it increases the rate of purification by replenishing the impurity concentration at the surface and because it tends to maintain an uniform composition throughout the melt.

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The question of how much surface tension driven flows might enhance the rate of purification of a levitated melt at zero-g is the motivation for the work reported here. As a model which will begin to supply part of the answer to this question, we will consider thermocapillary convection--that is, convection resulting from a temperature gradient along the surface (Ref. 2)--in nearly spherical drops. The temperature field at the surface of the drop will be treated in a general way by writing it as an expansion in spherical harmonics. The model will examine one of the convective modes corresponding to one of the terms in the expansion of the surface temperature field. The equations of motion--the Navier-Stokes equations--will be solved within the assumptions of a steady-state and of creeping flow--terms which are nonlinear in the velocity will be ignored. The mathematical analysis will proceed along the lines of the method which Miller and Scriven (3) applied to the oscillations of a fluid droplet although here we will not retain the time-dependence of their problem. The physical boundary conditions will account for both the convective flow and for the deformation of the surface so that both aspects of the Marangoni effect will appear in the problem. The velocity field components which are obtained from the Miller-Scriven analysis will be used to derive the circulation time τ whose inverse characterizes the rate of circulation within a convection cell. An expression for τ will be worked out in detail for convection cells having axial symmetry. Estimates of τ will be given for a variety of materials when the convection pattern is a single, axially symmetric vortex ring. Also, relative circulation times will be calculated for several higher order, axially symmetric convective modes. Illustrations of these convective modes will be given.

THE TEMPERATURE FIELD

The temperature field responsible for the convection is considered to be a general, non-uniform, but steady, field which can be written as:

$$T(r, \theta, \phi) = T_0 + \sum_{n=1}^{\infty} \sum_{m=0}^n \sum_{\sigma} T_{mn}^{\sigma} r^n Y_{mn}^{\sigma}(\theta, \phi) \quad (\sigma=e,o), \quad (1)$$

where,

$$Y_{mn}^e = P_n^m(\cos\theta) \cos m \phi \quad (2a)$$

$$Y_{mn}^o = P_n^m(\cos\theta) \sin m \phi \quad (2b)$$

are spherical harmonics as defined by Morse and Feshbach (4). The function $P_n^m(\cos\theta)$ is an associated Legendre polynomial. The simple case of a constant vertical temperature gradient through the drop-- $Y_{10}^e = P_1(\cos\theta) = \cos\theta$ -- is a special case of the solution of Young, Goldstein and Block (5) who derived the velocity field in a bubble rising in a vertical temperature gradient.

SOLUTION OF THE NAVIER-STOKES EQUATIONS

To obtain the velocity field $u(r, \theta, \phi)$ and the hydrodynamic pressure field $p(r, \theta, \phi)$ within a drop of radius R , we solve the linearized Navier-Stokes equations

$$\mu \nabla^2 \vec{u} = \nabla p \quad (3a)$$

$$\nabla \cdot \vec{u} = 0, \quad (3b)$$

where μ is the coefficient of viscosity. We impose the requirements

- 1) of a finite solution at $r = 0$;
- 2) of the kinematic condition $u_r(R) = 0$, where u_r is the radial component of the velocity; and
- 3) of the physical boundary conditions which will be examined in the next section.

Since Eqns. (3a,b) are linear, a general solution can be written as a superposition of all the modes. Thus, it is sufficient to find a solution for one of the modes (n, m, σ) .

Eqns. (3a,b) can be integrated according to the method of Miller and Scriven. This method integrates (3a,b) in terms of u_r and the radial component of the vorticity, where the vorticity is defined by

$$\vec{\zeta} = \nabla \times \vec{u}. \quad (4)$$

The results of the Miller-Scriven method which satisfy requirements "1" and "2" are

$$u_r(r, \theta, \phi) = A_{mn}^\sigma r^{n-1} (R^2 - r^2) Y_{mn}^\sigma(\theta, \phi) \quad (5a)$$

$$\zeta_r(r, \theta, \phi) = B_{mn}^\sigma r^{n-1} Y_{mn}^\sigma(\theta, \phi). \quad (5b)$$

The remaining integration constants A_{mn}^σ and B_{mn}^σ will be obtained from the physical boundary conditions in the next section.

The velocity components u_θ and u_ϕ can be obtained from u_r and ζ_r by a relation due to Sani (6);

$$\vec{u} = \hat{e}_r u_r + [r^2/n(n+1)] [\nabla_{II} R u_r - \hat{e}_r \nabla_{II} \zeta_r], \quad (6)$$

where \hat{e}_r is the radial unit vector,

$$\nabla_{II} = \nabla - \hat{e}_r \frac{\partial}{\partial r} \quad (7)$$

is the surface gradient operator and R is the operator

$$\frac{1}{r^2} \frac{\partial}{\partial r} r^2. \quad (8)$$

Eqn. (4) and the results for u_r , u_θ , u_ϕ can be used to derive ζ_θ and ζ_ϕ .

The hydrodynamic part $p(r, \theta, \phi)$ of the pressure can be found by taking the divergence of (3a) which yields $\nabla^2 p = 0$.

Thus,
$$p(r, \theta, \phi) = F_{mn}^\sigma r^n Y_{mn}^\sigma(\theta, \phi). \quad (9)$$

The coefficient F_{mn}^σ can also be obtained from (3a) by

$$\vec{r} \cdot \nabla^2 \vec{u} = \nabla^2 (ru_r) = (r/\mu) (\partial p / \partial r), \quad (10)$$

from which it can be shown that

$$F_{mn}^\sigma = 2(2n+3)\mu A_{mn}^\sigma / n. \quad (11)$$

PHYSICAL BOUNDARY CONDITIONS

The balance of stresses at the surface requires:

- 1) that the normal stress on the surface due to the hydrostatic pressure and to the motion of the fluid is balanced by the surface tension; and
- 2) that the shear stresses due to the variation of the surface tension are balanced by the fluid motion.

The deformation of the surface must be included in these conditions. The deformation is assumed to be small, so that

$$R + \Delta R = R [1 + \varepsilon(\theta, \phi)], \quad (12)$$

where ΔR is the displacement of the surface and

$$\varepsilon(\theta, \phi) = E_{mn}^\sigma Y_{mn}^\sigma(\theta, \phi) \quad (13)$$

is a radial strain. The coefficient E_{mn}^σ also must be obtained from the boundary conditions.

The normal stress due to the fluid

$$-p_{rr} = p_0 + F_{mn}^\sigma R^n Y_{mn}^\sigma - 2\mu (\partial u_r / \partial r) /_{r=R} \quad (14)$$

is balanced by the surface tension produced stress

$$\gamma \left(\frac{1}{R_1} + \frac{1}{R_2} \right), \quad (15)$$

*A simplification of the surface conditions is introduced at this point by neglecting the interfacial dilational elasticity and the interfacial shear elasticity which contribute an interfacial viscosity term to the normal stress equations--see Scriven (7) and Miller and Scriven (5) for discussions of these properties.

where γ is the local value of the surface tension and R_1 and R_2 are the principal radii of curvature. For small deformations of the surface of a spherical drop, Lamb (8) has shown that

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{1}{R} [2 + (n-1)(n+2) E_{mn}^\sigma Y_{mn}^\sigma]. \quad (16)$$

The surface tension coefficient is expanded in terms of the mean value γ_0 and the temperature coefficient γ_T as

$$\gamma = \gamma_0 + \gamma_T [T_{mn}^\sigma + L_{mn}^\sigma E_{mn}^\sigma] R^n Y_{mn}^\sigma, \quad (17)$$

where L_{mn}^σ is the coefficient obtained in the expansion of $T(R+\Delta R)$ of the deformed surface about the temperature $T(R)$ of the undeformed surface and $T_{mn}^\sigma R^n Y_{mn}^\sigma$ is the (n,m,σ) -term in the expansion of the surface temperature field.

Setting the hydrostatic pressure $p_0 = 2\gamma_0/R$, and considering terms to first order in Y_{mn}^σ in the normal stress condition, we obtain the first equation for A_{mn}^σ and E_{mn}^σ ,

$$6\mu R^{n+1} A_{mn}^\sigma / n = [\gamma_0 (n-1)(n+2) + 2\gamma_T L_{mn}^\sigma R^n] E_{mn}^\sigma = 2\gamma_T T_{mn}^\sigma R^n. \quad (18)$$

Instead of solving the shear stress conditions directly, it is more convenient to take the surface divergence and the surface curl of the force on an element of surface. The surface divergence equation is

$$\nabla_{II}^2 \gamma = \mu \left[\frac{\partial}{\partial r} (R u_r) - \nabla_{II}^2 u_r \right]_{r=R}. \quad (19)$$

Eqn. (19) can be simplified since $\nabla_{II}^2 u_r \big|_{r=R} = 0$. The divergence condition, then, yields a second equation for A_{mn}^σ and E_{mn}^σ ,

$$2(2n+1) \mu R^{n+1} A_{mn}^\sigma / n - (n+1) \gamma_T L_{mn}^\sigma R^n E_{mn}^\sigma = (n+1) \gamma_T T_{mn}^\sigma R^n. \quad (20)$$

The radial part of the surface curl equation yields the result that $B_{mn}^\sigma = 0$, or $\zeta_r = 0$.

Eqns. (18) and (20) can be solved simultaneously for A_{mn}^σ and E_{mn}^σ ,

$$A_{mn}^\sigma = n(n+1)(n+2) \gamma_0 \gamma_T T_{mn}^\sigma / 2\mu R D_{mn}^\sigma \quad (21)$$

$$E_{mn}^\sigma = \gamma_T T_{mn}^\sigma R^n / D_{mn}^\sigma, \quad (22)$$

where $D_{mn}^\sigma = (2n+1)(n+2) \gamma_0 + \gamma_T L_{mn}^\sigma R^n$. (23)

THE VELOCITY FIELD AND THE VORTICITY FIELD

Defining a characteristic rate of flow

$$U_{mn}^{\sigma} = -n(n+1)(n+2)\gamma_0 \gamma_T T_{mn}^{\sigma} R^n / 2\mu D_{mn}^{\sigma}, \quad (24)$$

the components of the velocity \vec{u} can be written as

$$u_r(r, \theta, \phi) = U_{mn}^{\sigma} \bar{r}^{n-1} (1-\bar{r}^2) Y_{mn}^{\sigma}(\theta, \phi) \quad (25a)$$

$$u_{\theta}(r, \theta, \phi) = U_{mn}^{\sigma} (\bar{r}^{n-1}/n) [1 - \frac{n+3}{n+1} \bar{r}^2] \cdot (\partial Y_{mn}^{\sigma} / \partial \theta) \quad (25b)$$

$$u_{\phi}(r, \theta, \phi) = U_{mn}^{\sigma} (\bar{r}^{n-1}/n) [1 - \frac{n+3}{n+1} \bar{r}^2] (1/\sin\theta) \cdot (\partial Y_{mn}^{\sigma} / \partial \theta), \quad (25c)$$

where $\bar{r} = r/R$.

Although $\zeta_r = 0$ within the drop, ζ_{θ} and ζ_{ϕ} generally do not vanish. Using the definition of the vorticity, Eqn. (4), and Eqns. (25a,b,c),

$$\zeta_{\theta} = Z_{mn}^{\sigma} r^n (1/\sin\theta) (\partial Y_{mn}^{\sigma} / \partial \phi) \quad (26a)$$

$$\zeta_{\phi} = -Z_{mn}^{\sigma} r^n (\partial Y_{mn}^{\sigma} / \partial \theta), \quad (26b)$$

where

$$Z_{mn}^{\sigma} = 2(U_{mn}^{\sigma}/R)(2n+3)/n(n+3). \quad (27)$$

The rate of flow, or speed, is

$$v = [u_r^2 + u_{\theta}^2 + u_{\phi}^2]^{1/2}, \quad (28)$$

where in general,

$$u_r = \sum_{n=1}^{\infty} \sum_{m=0}^n \sum_{\sigma} u_r(n, m, \sigma) \quad (29a)$$

$$u_{\theta} = \sum_{n=1}^{\infty} \sum_{m=0}^n \sum_{\sigma} u_{\theta}(n, m, \sigma) \quad (29b)$$

$$u_{\phi} = \sum_{n=1}^{\infty} \sum_{m=0}^n \sum_{\sigma} u_{\phi}(n, m, \sigma). \quad (29c)$$

Thus, the general expression for v can be very complicated. Only expressions for the pure modes will be investigated here.

For the special case of the lowest order mode $(n, m, \sigma) = (1, 0, e)$ and where $L_{01}^e = 0$, we obtain equations for a levitated drop in a constant vertical temperature gradient. In this case, the temperature in the drop is

$$T = T_0 + T_1 R \bar{r} \cos\theta, \quad (30)$$

which results in the velocity components

$$u_r = -(\gamma_T T_1 R / 3\mu) (1 - \bar{r}^2) \cos \theta \quad (31a)$$

$$u_\theta = (\gamma_T T_1 R / 3\mu) (1 - 2\bar{r}^2) \sin \theta \quad (31b)$$

$$u_\phi = 0.$$

These results can be obtained from the solutions of Young, et. al (5), as mentioned in the Introduction. In addition, we find that

$$E_{mn}^e = \gamma_T T_1 R / 9\gamma_o, \quad (32)$$

so that the drop is spherical in the limit $\gamma_T / \gamma_o \rightarrow 0$.

SOME CHARACTERISTICS OF AXIALLY SYMMETRIC MODES

For an axially symmetric mode $(n, 0, e)$, the expression for the rate of flow, Eqn. (28), is

$$\bar{v} = (\bar{r}^{n-1} / n) [n^2 (1 - \bar{r}^2)^2 (P_n^0)^2 + (1 - \frac{n+3}{n+1} \bar{r}^2)^2 (P_n^1)^2]^{1/2} \quad (33)$$

where $\bar{v} = v/U^e$, P_n is the n th order Legendre polynomial and P_n^1 is the associated Legendre polynomial of first degree. Eqn. (33) can be used to identify the stagnation points since $\bar{v} = 0$ at these points.

Within the drop, n rings of stagnation points about the axis of the drop can be readily identified: take $\bar{r}^2 = (n+1)/(n+3)$ and $P_n^0 = 0$; since P_n has n nodes and $0 \leq \phi \leq 2\pi$, n rings have been identified. On the surface of the drop ($\bar{r} = 1$), the rate of flow $\bar{v} = 0$ if $P_n^1 = 0$. The associated Legendre polynomial P_n^1 has $(n+1)$ nodes, including one at each pole. These two stagnation points at the poles and the $(n-1)$ stagnation rings on the surface coincide with the hot and cold spots of the temperature field and define the boundaries of the convection cells in the drop. If $n > 1$, $v = 0$ when $r = 0$. That is, flow does not occur through the center of the drop when $n > 1$. Finally, we must consider the possibility of other internal stagnation points. That is, are there other points such that

$$n^2 (1 - \bar{r}^2)^2 (P_n^0)^2 + (1 - \frac{n+3}{n+1} \bar{r}^2)^2 (P_n^1)^2 = 0 ?$$

The answer is no. Since each term is either positive or zero, and since neither $(1 - \bar{r}^2)$ and $(1 - (n+3)/(n+1)\bar{r}^2)$ nor P_n^0 and P_n^1 vanish at identical values of \bar{r} and $\cos \theta$, respectively, there are no other internal stagnation points.

For an axially symmetric problem, a streamfunction $\psi(\bar{r}, \theta)$ can be obtained from

$$\bar{u}_r = - \frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta} \quad (34a)$$

$$\bar{u}_\theta = \frac{1}{\bar{r} \sin \theta} \frac{\partial \psi}{\partial r} \quad (34b)$$

where $\tilde{u}_a = u_a / U_{mn}^\sigma$ ($a = r, \theta$). For the $(n, 0, e)$ -mode, the streamfunction is—

$$\psi = -r^{n+1}(1-r^2) \sin\theta P_n^1(x) / n(n+1), \quad (35)$$

where $x = \cos\theta$. For the mode $(1, 0, e)$, the streamfunction is that for Hill's spherical vortex (9). Stagnation points, indicated by "N", and typical streamlines for Hill's spherical vortex are illustrated in Fig. 1 and for the modes $n = 2, 3, 4, 5$ in Figs. 2-5, respectively. The drops are seen in vertical cross-sections. The model rings lie in planes perpendicular to the axis in each illustration. The straight lines within the circles represent the boundaries of the convection cells, and here $\psi = 0$. The positions of the stagnation points are listed in Table 1 for these five modes.

Table 1. Stagnation "Points" for Axially Symmetric Convection Cells in Nominally Spherical Drops

Mode n	\tilde{r}	Interior		Type	Surface ($\tilde{r} = 1$)		
		θ			θ	Type	
1	1/√2	--		ring	0°	point	
					180°	point	
2	0	--		point	0°	point	
	√3/5	54° 44' 8"		ring	90°	ring	
		125° 15' 52"		ring	180°	point	
3	0	--		point	0°	point	
	√2/3	39° 13' 54"		ring	63° 26' 6"	ring	
		"	90°		ring	116° 33' 54"	ring
		"	140° 46' 6"		ring	180°	point
4	0	--		point	0°	point	
	√5/7	30° 33' 20"		ring	49° 6' 24"	ring	
		"	70° 7' 28"		ring	90°	ring
		"	109° 52' 32"		ring	130° 53' 36"	ring
		"	149° 26' 40"		ring	180°	point
5	0	--		point	0°	point	
	1/2√3	25° 1' 2"		ring	40° 5' 17"	ring	
		"	57° 25' 14"		ring	73° 25' 38"	ring
		"	90°		ring	106° 34' 22"	ring
		"	122° 34' 46"		ring	139° 54' 43"	ring
		"	154° 58' 58"		ring	180°	point

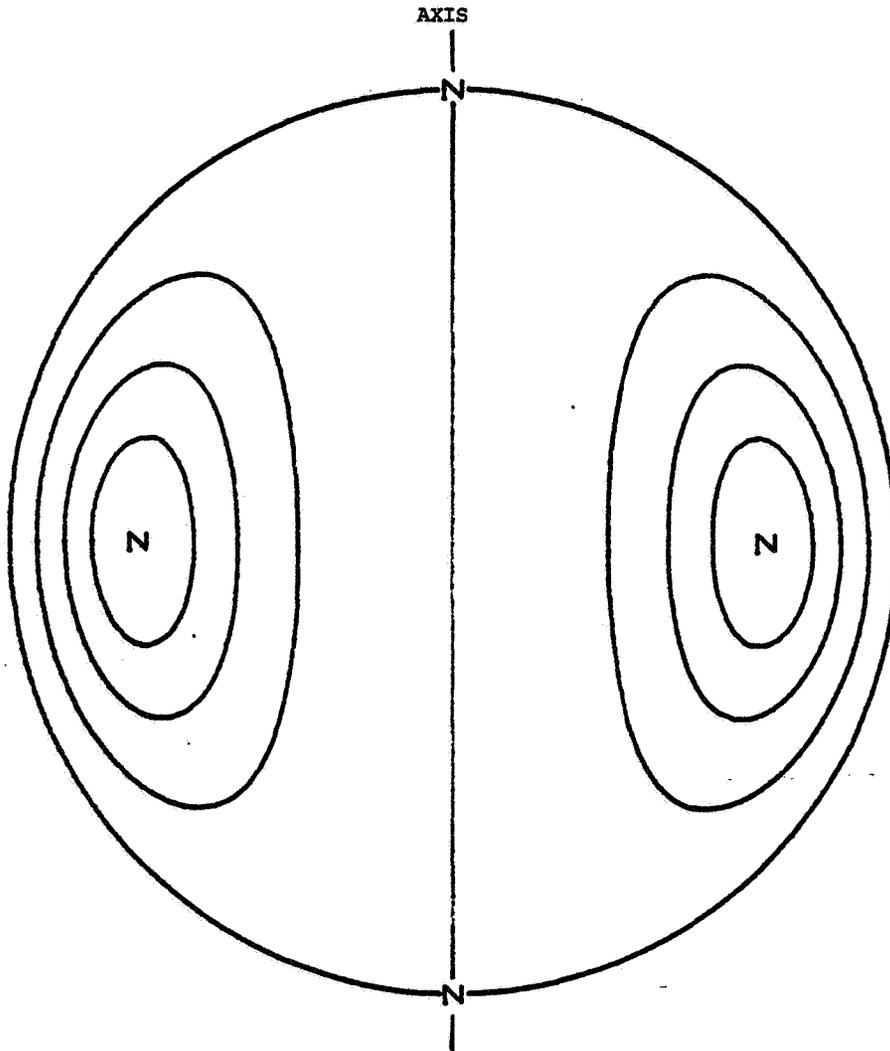


Figure 1. Streamlines and Stagnation Points ("N") for the $(1,0,e)$ -Mode: Hills' Spherical Vortex.

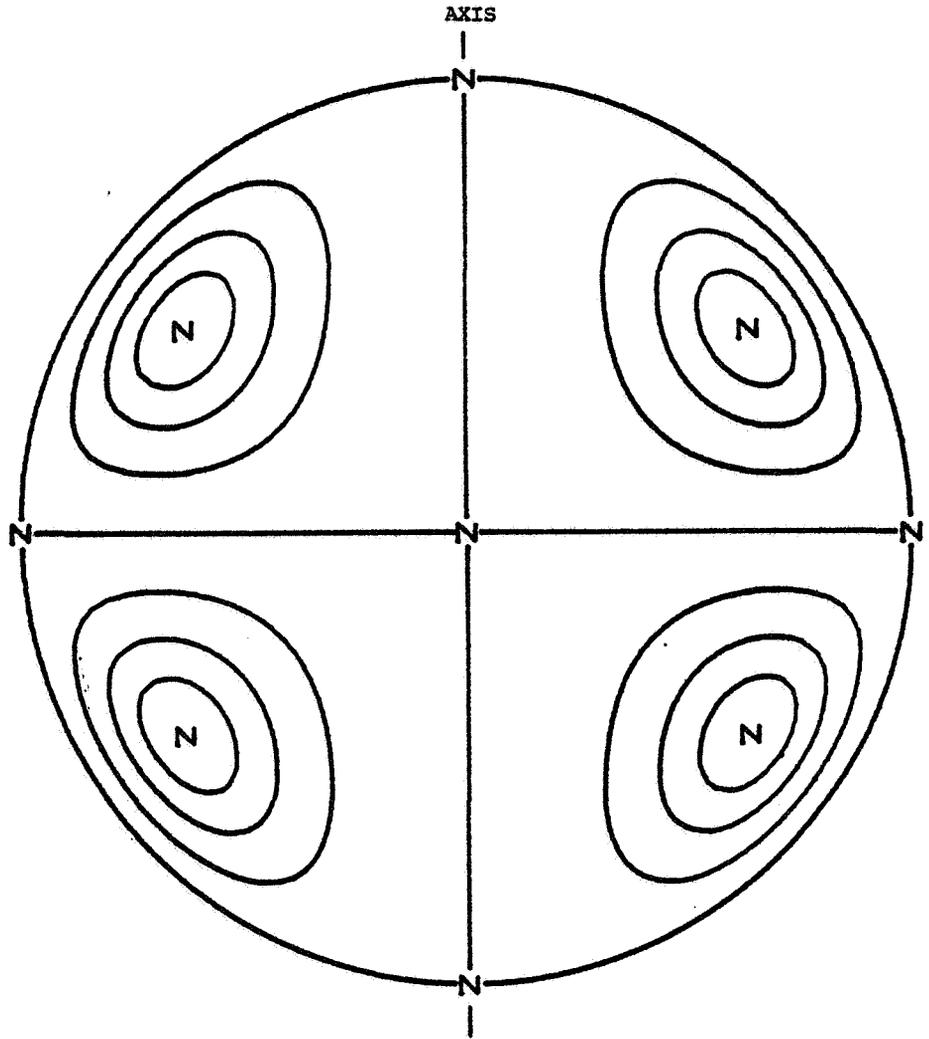


Figure 2. Streamlines and Stagnation Points for the $(2,0,e)$ -Mode

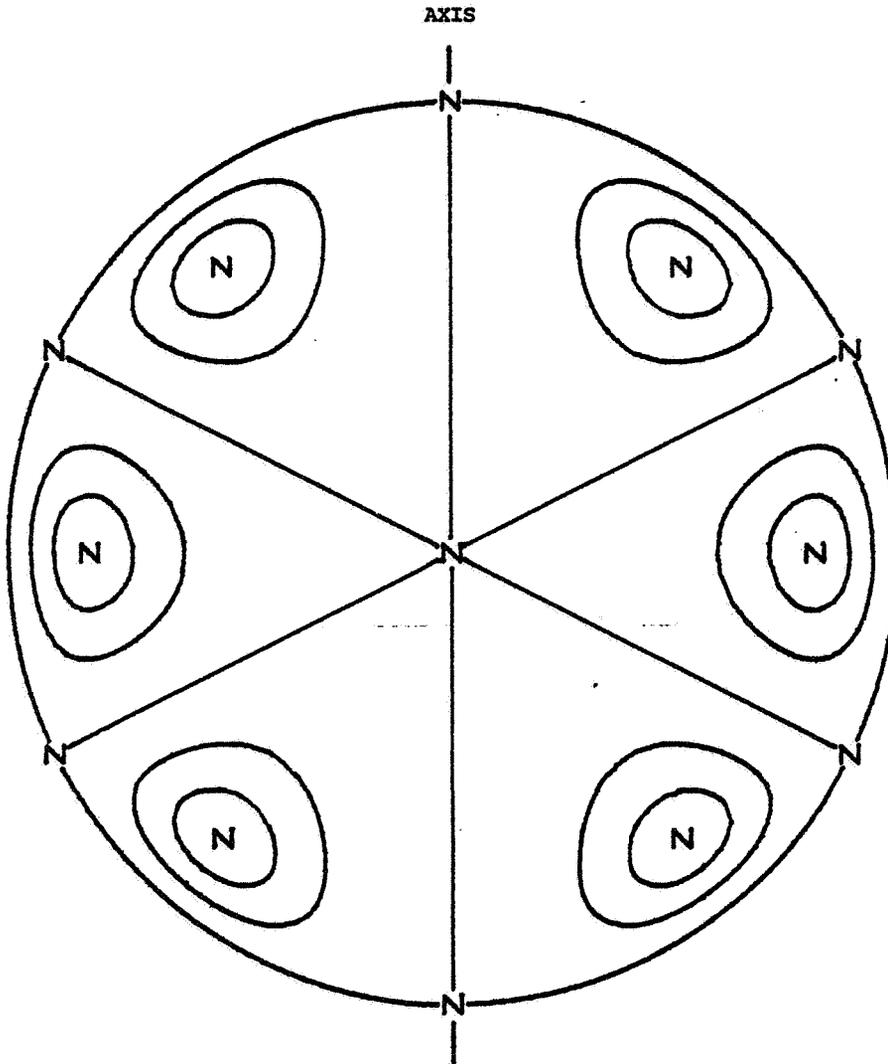


Figure 3. Streamlines and Stagnation Points for the $(3,0,e)$ -Mode

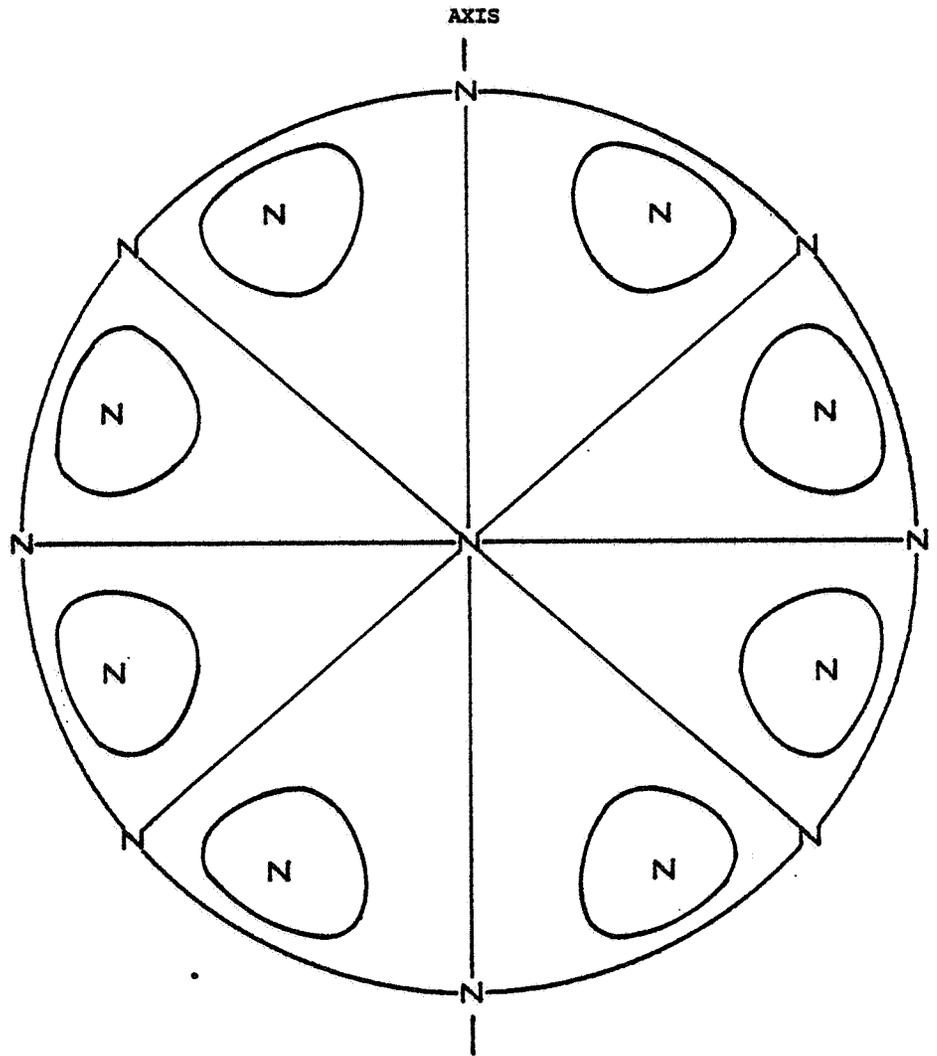


Figure 4. Streamlines and Stagnation Points for the $(4, 0, e)$ -Mode

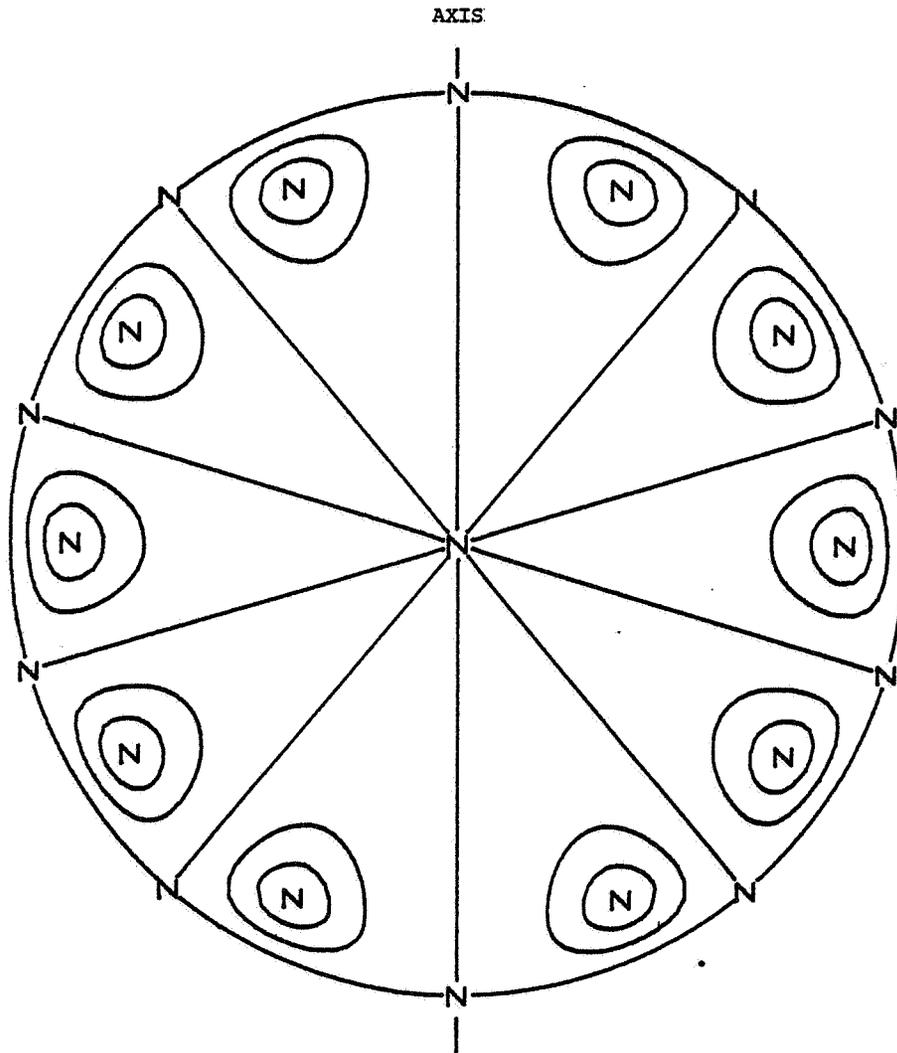


Figure 5. Streamlines and Stagnation Points For the $(5,0,e)$ -Mode

THE CIRCULATION TIME

The circulation time τ will be introduced in this section as a means of characterizing the rate of convective mass transfer from the interior of the convection cell to the vicinity of the surface of the drop. The reciprocal circulation time τ^{-1} is defined by

$$\begin{aligned}\tau^{-1} &= \oint \vec{u} \cdot d\vec{l} / \int dS \\ &= \oint \vec{\zeta} \cdot \vec{n} dS / \int dS,\end{aligned}\tag{36}$$

where the circulation integral

$$\oint \vec{u} \cdot d\vec{l}$$

is taken around the boundary of the cell formed by the intersection of the cell with a vertical plane containing the axis of the drop, such as any of the cell boundaries shown in Figures 1-5. The second equality follows from Stokes's theorem, where the surface integrals

$$\int \vec{\zeta} \cdot \vec{n} dS \text{ and } \int dS$$

are taken over the region bounded by the circuit of the line integral; \vec{n} is the unit normal vector to the surface and here is equal to \hat{e}_z . According to Eqn. (36), the reciprocal circulation time is the average vorticity of the cell. The circulation time resembles the period of rotation of a rigid body. Indeed, if the fluid circulated about the vortical center as a rigid body, Eqn. (36) would yield 4π times the frequency of rotation and τ would be the period of the rotation reduced by 4π . Unfortunately, such a simple interpretation of thermocapillary convection cells is not possible, but τ can still be used to characterize the rate of circulation within the cell.

For the axially symmetric mode $(n, 0, e)$, the integrals in Eqn. (36) can be performed yielding

$$\tau_{ns}^{-1} = (n+2) [P_n(x'_s) - P_n(x'_{s+1})] / 2Z_{0n}^e (\theta'_s - \theta'_{s+1})\tag{37}$$

for the circulation time for the s -cell, where $1 \leq s \leq n+1$, and x'_s and x'_{s+1} are roots of

$$P_n^1(x) = 0.\tag{38}$$

Cell- s has its vortical center at the stagnation point whose angular position θ'_s is given by the root x'_s of

$$P_n(x) = 0;\tag{39}$$

θ_s lies within the range whose lower bound is θ' and whose upper bound is θ'^{s+1} . The circulation time may be either positive or negative, the sign depending upon the direction of circulation in the cell. Using Eqns. (1), (24) and (27), we can show that, apart from the geometric factor, the circulation time τ depends only on the material parameters μ and γ_T and on the temperature gradient at the surface.

The circulation times for the mode $n = 1$ are given in Tables 2-5 for a variety of liquids. Here, we assume "unit conditions": $L_{01}^e = 0$, and a unit temperature gradient, $T_{01}^e = -1^\circ/\text{cm}$. The assumption of unit conditions is indicated for the circulation time by τ^o , where the superscript "o" designates unit conditions. Table 2 lists circulation times for some liquids at room temperature; Table 3, for five liquid metals; Table 4, for four molten oxides; and Table 5 for three molten sodium halide salts.

Table 2. Circulation Times* for Some Liquids at Room Temperature (298.15 K)

Material	γ_T mN/K.m**	Viscosity mPa.s**	$ \tau $, s
Acetone	-0.112 (11)	0.316 (10)	0.0323
DC 200, 20 cs [†]	-0.062 (5)	19 (5)	3.5
200 cs	-0.065 (5)	193 (5)	34
1000 cs	-0.061 (5)	793 (5)	180
Ethanol	-0.9832 (11)	1.092 (10)	0.150
n-Hexadecane	-0.106 (5)	3.086 (12)	0.334
Krytox 143 AZ ^{††}	-0.1 (13)	32.4 (13)	4
Water	-0.1477 (11)	0.8904 (13)	0.06908

*temperature gradient at the poles has a magnitude of $1^\circ/\text{cm}$.

**1 mN/m = 1 dyn/cm; 1 mPa.s = 1 cp = 10^{-2} dyn.s/cm².

†Dow Corning silicone oils of the DC 200 series.

††A perfluoroalkylpolyether.

Table 3. Circulation Times*--Metals

Materials	γ_T mN/K·m	Temp. T_o K	Viscosity mPa.s	$ \tau , s$
Aluminum	-0.356 (10)	930	4.5 (15)	0.14
		1070	2.5	0.08
Copper	-0.06 (10)	1370	4.5 (15)	0.9
		1470	3.9	0.7
Mercury	-0.2049 (11)	290	1.554 (15)	0.0869
		470	1.052	0.0588
Sodium	-0.09833 (14)	470	0.450 (15)	0.00524
		670	0.284	0.00331
Tin	-0.0706 (13)	500	1.97 (13)	0.320
		870	1.05 (13)	0.170

*The temperature gradient at the poles has a magnitude of 1°/cm.

Table 4. Circulation Times*--Oxides

Material	γ_T mN/K·m	Temp. T_o K	Viscosity Pa.s	$ \tau , s$
Al ₂ O ₃	(0.1) ⁺	2400	0.11 (16)	13
		2600	0.062	7
B ₂ O ₃	0.0354 (18) at 720 K	1410	5.02 (17)	1620
		1670	2.01	651
GeO ₂	0.056 at 1390 K	1750	12.2 (17)	2500
		1930	0.787	160
SiO ₂	0.031 at 2000 K	2280	717 (17)	8 × 10 ⁵
		2680	102	1 × 10 ⁵
		2820	46.4	5 × 10 ⁴

*Temperature gradient at the poles has a magnitude of 1°/cm.

⁺Estimated.

Table 5. Circulation Times*--Sodium Halides

Material	γ_T (18) mN/K·m	Temp. T_0 K	Viscosity (17) mPa.s	$ \tau , s$
NaCl	-0.0719	1090	1.38	0.220
		1150	1.08	0.172
NaBr	-0.0809	1060	1.28	0.181
		1170	1.00 ₆	0.142
NaI	-0.129	1030	1.15	0.102
		1100	0.96	0.085

*Temperature gradient at the poles has a magnitude of $1^\circ/\text{cm}$.

The steady-state temperature gradient is obtained by balancing the conductive heat flux through the drop against the radiant heat flux away from the drop in the cooler hemisphere. Sufficient heat is applied to the hotter hemisphere to maintain the steady-state gradient. Since the radiant heat flux $\propto T^4$, liquids such as molten metals which have high thermal conductivities will only have steep steady-state gradients at high temperatures. Although copper has the highest thermal conductivity of the liquid metals considered in Table 3, it is the only one of the five metals which is considered at temperatures high enough for a rate of radiant heat loss to be attained which could produce a steady-state gradient of $1^\circ/\text{cm}$. Thus, the circulation times given for copper in the table are the only ones which are physically attainable under steady-state conditions. For the other metals, the physically attainable value of τ_1 will be longer than τ_1^* ; the attainable steady convective rates will be less than the rates produced by a gradient of $1^\circ/\text{cm}$.

The long circulation times estimated for the oxides, B_2O_3 , GeO_2 and SiO_2 show the importance of the viscosity in determining whether thermocapillary convection can occur in the liquid. For these three oxides, thermocapillary convection is expected to be negligible. However, Al_2O_3 has a lower viscosity than these other three oxides and, thus, a shorter circulation time. Since temperature gradients much larger than $1^\circ/\text{cm}$ can be attained in molten oxides, significant thermocapillary convection might be observed in molten alumina.

The $n = 1$ mode has been demonstrated on earth by the work of Young, Goldstein and Block (5) who observed the balancing of the thermocapillary and buoyancy forces on small bubbles in a vertical temperature gradient. The low-g of a space laboratory in addition to facilitating the levitation of a drop would allow the observation of the higher convective modes, in particular, the $n = 2$ mode. The circulation could be observed by the movement of a dye or of a radiotracer.

Relative circulation times for the axially symmetric modes $n=2,3,4,5$ are given in Table 6. These times are calculated relative to the single vortex ($n = 1$) time τ_1^0 . The sign of the circulation time for a particular cell indicates the direction of circulation with respect to that in the single vortex drop. Since the convective rate decreases as n increases, the higher modes appear to be less desirable than the $n = 1$ mode for obtaining rapid purification. However, the convective rates for some of these higher modes still can be large enough for the convective pattern to be studied and for these modes to have some utility for the purification of levitated melts in space.

SUMMARY

Equations describing the convective flow velocities in thermocapillary convection cells in drops have been derived. When the thermal conditions approximate a pure, symmetric mode--that is, when the temperature field can be described by a single Legendre polynomial, the streamlines of the vortex and the positions of the stagnation points in each cell can be calculated. The circulation time, which is the reciprocal of the average vorticity of the cell and which characterizes the rate of circulation, also can be calculated for these high symmetry cells. An estimate of the circulation time for a liquid drop is an indicator of the probable significance of thermocapillary convection in the drop when a temperature gradient is imposed across it.

Table 6. Relative Circulation Times, τ_{ns}^0/τ_1^0 , for Axially Symmetric Modes with $n > 1$

Mode n	Cells s	τ_{ns}^0/τ_1^0 *
2	1	3.968
	2	-3.968
3	1	9.981
	2	-7.365
	3	9.981
4	1	19.994
	2	-13.505
	3	13.505
	4	-19.994
5	1	34.960
	2	-22.691
	3	20.647
	4	-22.691
	5	34.960

* τ_{ns}^0 is the circulation time for the s -cell of the n th mode,

τ_1^0 is the circulation time for mode $n = 1$; a temperature gradient with a magnitude of $1^\circ/\text{cm}$ is assumed at the poles. The sign indicates the direction of circulation with respect to the $n = 1$ mode.

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