Onset of Thermosolutal Convection in a Liquid Layer Having Deformable Free Surface – I. Stationary Convection

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The effect of surface deformation on the onset of thermosolutal convection in a horizontal thin layer heated from below and salted from above is examined, using linear stability theory. It is found that two critical Crispation numbers Cr_1 and Cr_2 exist. For $\mathrm{Cr} \leqq \mathrm{Cr}_1$ the instability mechanism remains uneffected by the deformation of the free surface, whereas for $\mathrm{Cr}_1 < \mathrm{Cr} < \mathrm{Cr}_2$ the critical Marangoni number (M_c) decreases, showing instability due to deformation. If $\mathrm{Cr} = \mathrm{Cr}_2$, M_c is obtained for two values of the wave number. When $\mathrm{Cr} > \mathrm{Cr}_2$, M_c decreases as the wave number tends to zero. Further, the effect of the Marangoni number, Biot number, Bond number etc. on the stability characteristics of the problem is discussed.

1. Introduction

Calculations of the onset of thermal convection due to variations of the surface tension with temperature were first performed by Pearson [1]. He showed that these variations can lead to convection in a fluid layer even in the absence of buoyancy. This effect is due to the shearing forces produced in the surface layer by gradients of the surface tension and is known in the literature as Marangoni effect. Later several authors [2–5] studied the effect of different kinds of body forces and thermal conditions on Marangoni convection. The frequency of these attempts has increased recently because it was realized that Marangoni convection plays a role in crystal growth under microgravity conditions. In most of these studies the free surface is considered to remain undeformed, which is unrealistic.

Scriven and Sternling [6] considered the deformation of the free surface and showed that disturbances with zero wave number are always unstable. Smith [7] considered a deformable free surface and studied the gravity effects. Zeren and Reynolds [8] and Davis and Homsy [9] additionally included buoyancy effects, but in all these attempts exchange of stabilities has been assumed without proof. Therefore these studies are incomplete in the sense that they have not explored the possibility of other modes of instability, viz. the case of overstability. Takashima [10, 11] considered the gravity wave effect on both stationary and oscillatory instabilities for a pure Marangoni convection and pointed out that overstability is possible only when the rigid wall is cold. Recently Benguria and Depassier [12, 13] reported that oscillatory instability may occur at a smaller Rayleigh number than steady convection if the heat flux is fixed on the free surface and the lower surface is rigid and isothermic.

One of the problems associated with the growth of large semiconductor single crystals with uniform material properties either from melts or from aqueous solutions in microgravity conditions is the development of striations or segregation bands parallel to the melt-crystal interface. These striations are regions of varying impurity concentration, the underlying cause of which seeming to be a time dependent crystal-growth speed brought about by temperature oscillations in the melt. The origin of the temperature oscillations is due to the presence of instabilities in the flow field of the melt (Wilcox [14]). Chun and Schwabe [15] pointed out that the crystal growth from the melt is controlled by heat and mass transfer, i.e. by convection.

Hurle [16] has reviewed the hydrodynamics of crystal growth and pointed out the need for further detailed study of the problem under near-zero-gravity.

The problem of thermosolutal convection in a layer heated from below and subject to the distribution of solute was discussed by Veronis [17]. He neglected the variation of surface tension with temperature and concentration and assumed that the surface is flat. This discussion was extended by McTaggart [18] to the case of a concentration and temperature dependent surface tension for an undeformed free surface. In the present paper we study the onset of convection in a horizontal layer of fluid heated from below in presence of varying

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solute concentration. We assume that the surface tension varies with both temperature and concentration and that the free surface is deformable.

2. Formulation of the Problem

Consider a horizontal liquid layer of mean thickness d with a free upper surface open to the ambient air. The origin of the cartesian coordinate system (x', y', z') is at the rigid bottom plane and the z' direction is upward. Let T_0 and S_0 be the initial constant temperature and concentration, respectively, at the bottom. Further we assume that all physical properties of the liquid are constant except the surface tension, which is a linear function of temperature and concentration.

In most of the crystal growing processes the melt zone is very thin. Specially in the floating zone technique the thickness of the melt zone varies from 1 to 7 mm. In this thin layer one may neglect the effect of buoyancy, as pointed out by Takashima [10, 11] and Tan, Bankoff, and Davis [19] by treating the density ϱ as constant.

Under this assumption the governing equations are given by

$$\nabla \cdot \boldsymbol{q}=0, \tag{1}$$

$$\frac{\partial \boldsymbol{q}}{\partial t}+\boldsymbol{q}\cdot\nabla\boldsymbol{q}=g\,\hat{e}_{z'}-\frac{1}{\varrho}\nabla p+\vartheta\nabla^2\boldsymbol{q}, \tag{2}$$

$$\frac{\partial T}{\partial t}+\boldsymbol{q}\cdot\nabla T=k_T\nabla^2 T, \tag{3}$$

$$\frac{\partial S}{\partial t}+\boldsymbol{q}\cdot\nabla S=k_S\nabla^2 S, \tag{4}$$

where $\boldsymbol{q}(\equiv u, v, w)$, $\hat{e}_{z'}(\equiv 0,0,1)$, p, ϑ, g, T, k_T, S, and k_S denote velocity, unit vector along z' direction, pressure, kinematic viscosity, acceleration due to gravity, temperature, thermal diffusivity, concentration and mass diffusivity, respectively.

The boundary conditions are:
At the rigid plane $(z'=0)$

$$\boldsymbol{q}=0, \quad T=0 \quad \text{and} \quad S=0. \tag{5}$$

At the free surface $(z'=d+\eta'(x', y', t'))$,

$$\frac{\partial \eta'}{\partial t'}+u\frac{\partial \eta'}{\partial x'}+v\frac{\partial \eta'}{\partial y'}=w, \tag{6}$$

$$S_{ij}\,n_j\,n_i=2\Lambda S_T, \tag{7}$$

$$S_{ij}\,n_j\,t_i=S_{Ts}, \tag{8}$$

$$T_n+H(T-T_g)=0, \tag{9}$$

$$S_n+GS=0. \tag{10}$$

In the above conditions the subscripts n and s represent the normal and tangential derivatives at the interface, respectively. Equation (6) is the kinematic boundary condition at the liquid gas interface. The stress balance at the interface in normal and tangential directions are given by (7) and (8), respectively. The jump in the normal stress across the interface is balanced by the surface tension times the mean curvature Λ, and the jump in the shear stress equals the surface tension gradient along the interface. In these equations S_{ij}, $\hat{n}$, $\hat{t}$, S_T, H, and G denote the stress tensor for the incompressible viscous liquid, unit outward normal vector, unit tangent vector, surface tension, heat transfer coefficient in the gas liquid interface and the mass transfer coefficient in the interface, respectively. Here we assume that the temperature of the gas T_g is constant.

It is to be noted that there exist the following steady solutions

$$\bar{u}=\bar{v}=\bar{w}=0,$$
$$\bar{p}=p_A+\varrho g(d-z'),$$
$$\bar{T}=T_0-\beta z',$$
$$\bar{S}=S_0+\beta_1 z',$$

where p_A is the atmospheric pressure, $\beta=\dfrac{H(T_0-T_g)}{1+\mathrm{Hd}}$ and $\beta_1=-GS_0/(1+\mathrm{Gd})$ are the adverse temperature and solute gradients, respectively.

Following the standard method of linearised stability analysis, this basic state is perturbed as follows:

$$\boldsymbol{q}=\bar{\boldsymbol{q}}+\boldsymbol{q}'(x', y', z', t'),$$
$$p=\bar{p}+p'(x', y', z', t'),$$
$$T=\bar{T}+T'(x', y', z', t'),$$
$$S=\bar{S}+S'(x', y', z', t'),$$

and expresses the system (1)–(10) in its linearised form.

Now, introducing the scales for the variables as

$$(x', y', z')=\mathrm{d}(x, y, z),\ t'=(d^2/k_T)t,\ \boldsymbol{q}=(k_T/d)\boldsymbol{q},$$
$$p'=(\varrho k_T^2/d^2)p,\ T'=(\beta d)T,\ S'=(\beta_1 d)S \text{ and } \eta'=\mathrm{d}\eta$$

in the linearised system of (1)–(10), we obtain the following dimensionless parameters:

$P_r = \vartheta/k_T$ the Prandtl number,
$\mathrm{Cr} = \varrho\,\vartheta\,k_T/\tau_0\,d$ the Crispation/Capillary number,
$B_0 = \varrho\,g\,d^2/\tau_0$ the Bond number,
B_i = Hd the Biot number,
B_i' = Gd the solute Biot number,
$M = \gamma\,\beta\,d^2/\varrho\,\vartheta\,k_T$ the Marangoni number,
$M' = \gamma'\,\beta_1\,d^2/\varrho\,\vartheta\,k_T$ the solute Marangoni number and
$\tau = k_S/k_T$.

In the foregoing analysis we have assumed the variation of surface tension as

$$S_T = \tau_0 - \gamma(T-T_0) + \gamma'(S-S_0), \tag{11}$$

where

$$-\gamma = \frac{\mathrm{d}S_T}{\mathrm{d}T} \quad \text{and} \quad \gamma' = \frac{\mathrm{d}S_T}{\mathrm{d}S}.$$

Here τ_0 is the surface tension for $T = T_0$ and $S = S_0$, γ is positive and γ' may be either positive or negative. It is to be noted that when $\gamma' > 0$, the free energy will be minimum if the impurity is distributed over the melt surface nonuniformly. Although the actual redistribution of the impurity over the surface and in the bulk of the melt is more complicated, yet the impurity distribution will be a maximum in the low temperature zone (Avduyevsky [20]).

Eliminating the pressure and the horizontal components of velocity, viz. u and v, from the linearised momentum equation, we follow the procedure for linear stability analysis (Chandrasekhar [21]) by decomposing the spatial dependence in terms of normal modes:

$$(w, T, S, \eta) = [W(z), \theta(z), \phi(z), Z] \cdot \exp[i(k_x x + k_y y) + \omega t] \tag{12}$$

and finally obtain the following set of equations after using $a^2 = k_x^2 + k_y^2$ as

$$(D^2 - a^2)(D^2 - a^2 - \omega P_r^{-1})\,W = 0,$$
$$(D^2 - a^2 - \omega)\,\theta = -W, \tag{13a–c}$$
$$[\tau(D^2 - a^2) - \omega]\,\phi = W,$$

subject to the boundary conditions

$$W = DW = \theta = \varphi = 0 \quad \text{at} \quad z = 0, \tag{14a–d}$$

$$W = \omega Z = 0,$$
$$\mathrm{Cr}(D^2 - 3a^2 - \omega P_r^{-1})\,DW - a^2(B_0 + a^2)\,Z = 0,$$
$$(D^2 + a^2)\,W + a^2(M\theta - M'\varphi) - a^2(M + M')\,Z = 0,$$
$$(D + B_i)\,\theta - B_i Z = 0,$$
$$(D + B_i')\,\varphi + B_i' Z = 0. \qquad \text{at} \quad z = 1 \tag{15a–e}$$

For $\mathrm{Cr} = 0$, we arrive at the same set of equations as considered by McTaggart [18]. The differences arise due to the use of different characteristic scales. McTaggart considered the problem as heated and salted from above, whereas here we have considered the case when heated from below and salted from above. To get the same notation as that of Vidal and Acrivos [22] and with McTaggart, the following changes are to be made:

$$\theta = -\theta, \quad \varphi = \tau^{-1} C, \quad M'\tau^{-1} = B_S,$$
$$M = B_T \quad \text{for} \quad B_0 = \mathrm{Cr} = 0.$$

3. Stationary Convection

In this paper we shall only study the stability characteristic for stationary convection. The guiding equations can be obtained from (13)–(15) by setting $\omega = 0$. They read

$$(D^2 - a^2)^2\,W = 0, \tag{16a–c}$$
$$(D^2 - a^2)\,\theta = -W \quad \text{and} \quad \tau(D^2 - a^2)\,\varphi = W,$$

subject to the boundary conditions,

$$W = DW = \theta = \varphi = 0 \quad \text{at} \quad z = 0, \tag{17a–d}$$

and

$$W = 0,$$

$$D^2 W + a^2(M\theta - M'\varphi) - \frac{\mathrm{Cr}(M + M')(D^2 - 3a^2)\,DW}{(B_0 + a^2)} = 0,$$

$$(D + B_i)\,\theta = \frac{B_i\,\mathrm{Cr}}{a^2(B_0 + a^2)}(D^2 - 3a^2)\,DW = 0,$$

$$(D + B_i')\,\varphi + \frac{B_i'\,\mathrm{Cr}}{a^2(B_0 + a^2)}(D^2 - 3a^2)\,DW = 0.$$

$$\text{at} \quad z = 1 \tag{18a–d}$$

4. Solution and Numerical Results

The solution of (16a) subject to the boundary conditions $W(0) = DW(0) = W(1) = 0$ can be obtained as

$$W = A\,[z \sinh az + K_0(\sinh az - az\cosh az)], \tag{19}$$

similarly the solutions for the set (16b, 17c, and 18c) and (16c, 17d, and 18d) give as

$$\theta = \frac{A}{4a^2}[K_1 \sinh az - az^2\cosh az + z\sinh az - K_0\,a(3z\cosh az - az^2\sinh az)] \tag{20}$$

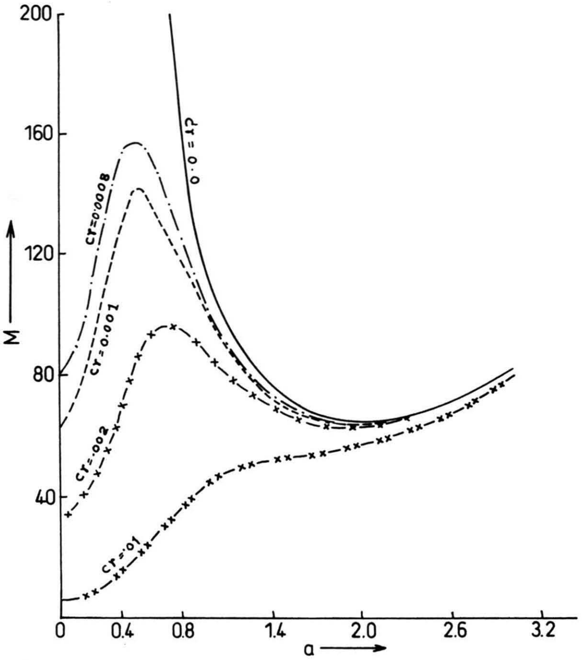

Fig. 1. M versus a for different values of Cr and fixed values $M'=1, \tau=0.07, B_0=0.1, B_i=B_i'=0$. —— for Cr = 0.0, –·– for Cr = 0.0008, - - - - for Cr = 0.001, –×–×–×– for Cr = 0.002 and –××–××– for Cr = 0.01.

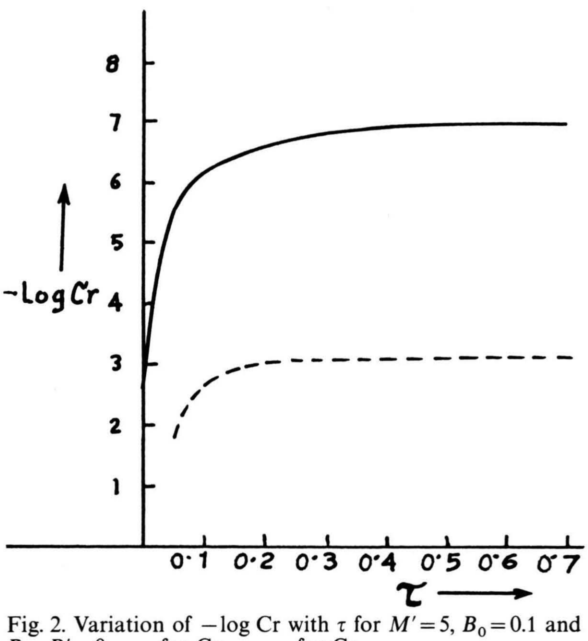

Fig. 2. Variation of $-\log$ Cr with τ for $M'=5$, $B_0=0.1$ and $B_i=B_i'=0$. —— for Cr_1, - - - - for Cr_2.

and

$$\varphi = \frac{A}{4a^2\tau}\,[K_2 \sinh a z - a z^2 \cosh a z + z \sinh a z + K_0\, a(3 z \cosh a z - a z^2 \sinh a z)], \quad (21)$$

where

$$K_0 = S_a/(a C_a - S_a),$$

$$K_1 = [(B_0+a^2)\{a^2 C_a^2 + S_a^2 + a S_a C_a + B_i(a^2 + a S_a C_a + S_a^2)\} - 8 B_i \mathrm{Cr}\, a^4]/[(B_0+a^2)(a C_a + B_i S_a)(a C_a - S_a)], \quad (22)$$

$$K_2 = -[(B_0+a^2)\{a^2 C_a^2 + S_a^2 + a S_a C_a + B_i'(a^2 + a S_a C_a + S_a^2)\} - 8 B_i' \tau \mathrm{Cr}\, a^4]/[(B_0+a^2)(a C_a + B_i' S_a)(a C_a - S_a)],$$

$$S_a = \sinh a \quad \text{and} \quad C_a = \cosh a.$$

It is clear that to obtain the solutions for W, θ, and φ we have not used the boundary condition (18 b) so far. Using (19), (20) and (21) in (18 b) we get an eigenvalue relationship among the parameters M, M', Cr, τ, B_0, B_i, B_i', and a as

$$8a(B_0+a^2)(a - S_a C_a) + M\left[\frac{8\,\mathrm{Cr}\, a^5 C_a + (B_0+a^2)(S_a^3 - a^3 C_a)}{(a C_a + B_i S_a)}\right] + \frac{M'}{\tau}\left[\frac{8\tau\,\mathrm{Cr}\, a^5 C_a + (B_0+a^2)(S_a^3 - a^3 C_a)}{(a C_a + B_i' S_a)}\right] = 0. \quad (23)$$

Setting $M'=0$ in (23) we arrive at the relation obtained by Takashima [10] for pure Marangoni convection with deformable free surface. Again for $B_0 = \mathrm{Cr} = 0$ and replacing M by B_T and $M'\tau$ by B_S one can arrive at the expression obtained by McTaggart [18].

Figure 1 shows the variation of M versus a for different values of Cr and fixed values of M', τ, B_0, and B_i and B_i'. It is clear from Fig. 1 that there exist two values of the Crispation number, Cr_1 and Cr_2 (say), such that when $\mathrm{Cr} \leqq \mathrm{Cr}_1$ the instability mechanism is not effected by the deformation of the free surface while for $\mathrm{Cr}_1 < \mathrm{Cr} < \mathrm{Cr}_2$, the critical Marangoni number M_c decreases, showing instability due to deformation. It is to be noted that the critical wave number a_c takes the value 1.99 for $\mathrm{Cr} \leqq \mathrm{Cr}_1$ but for $\mathrm{Cr}_1 < \mathrm{Cr} < \mathrm{Cr}_2$, $a_c = 1.96$. For $\mathrm{Cr} = \mathrm{Cr}_2$, M_c can be obtained at two values of the wave number, viz. $a_c = 0.008$ and 1.96. When $\mathrm{Cr} > \mathrm{Cr}_2$, M_c decreases as a tends to zero. Figure 2 shows the variation of Cr

Table 1. Effect of τ when salted from above.

τ	Cr_1	a_{c_1}	M_{c_1}	Cr_2	M_{c2}	a_{c21}	a_{c22}
0.05	4×10^{-6}	3.11	2.516×10^{-1}	1.4×10^{-2}	0.1	0.003	3.11
0.07	1×10^{-6}	1.99	8.178	5.3×10^{-3}	7.24	0.008	1.96
0.1	7×10^{-7}	1.99	29.607	1.9×10^{-3}	28.715	0.008	1.96
0.7	1×10^{-7}	1.99	72.464	8.48×10^{-4}	71.55	0.008	1.96

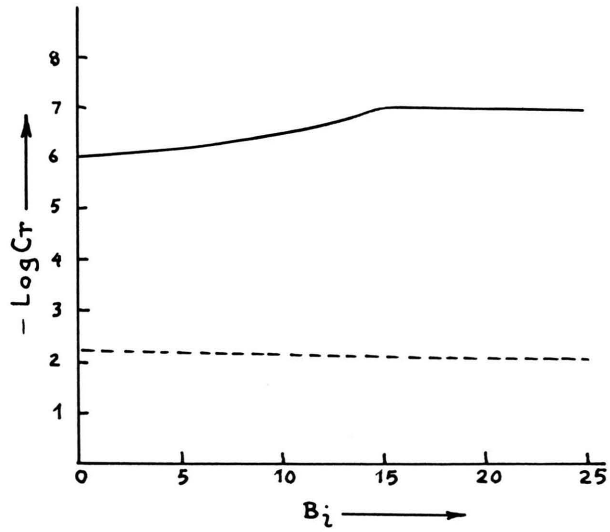

Fig. 3. $-\log$ Cr versus B_i for $M'=5$, $\tau=0.07$, $B_0=0.1$, $B_i'=0$, —— for Cr_1 and - - - - for Cr_2.

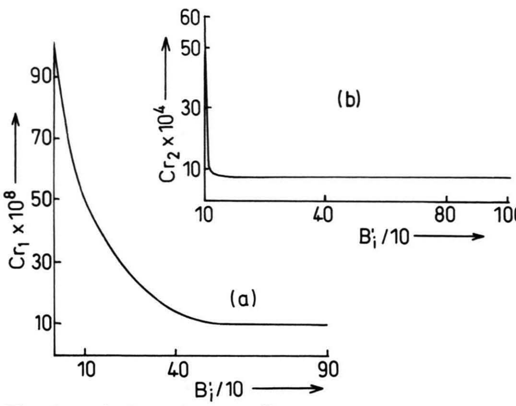

Fig. 4. Variation of $Cr\times10^8$ with $B_i'/10$ for $M'=5$, $\tau=0.07$, $B_i=0$, $B_0=0.1$. 4 (a) represents Cr_1 and 4 (b) Cr_2.

with τ for fixed values of M', B_0, B_i and B_i'. This shows that at first $-\log$ Cr increases as τ increases and takes an asymptotic value for large τ. Decrease of Cr_1 and Cr_2 with the increment of τ implies that the critical M_c increases, in other words τ stabilizes the system for $M'>0$. This result can be visualized in Table 1, which is obtained for $M'=5$, $B_0=0.1$ and $B_i=B_i'=0$.

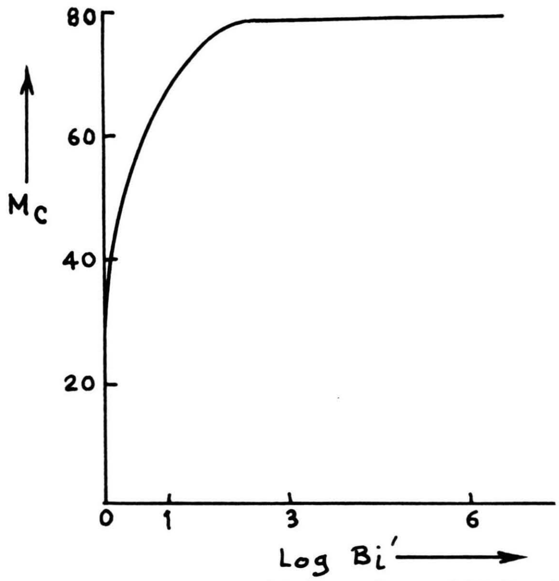

Fig. 5. Variation of M_c with $\log B_i'$ for $\tau=0.07$, $B_0=0.1$, $B_i=0$ and $M'=5$.

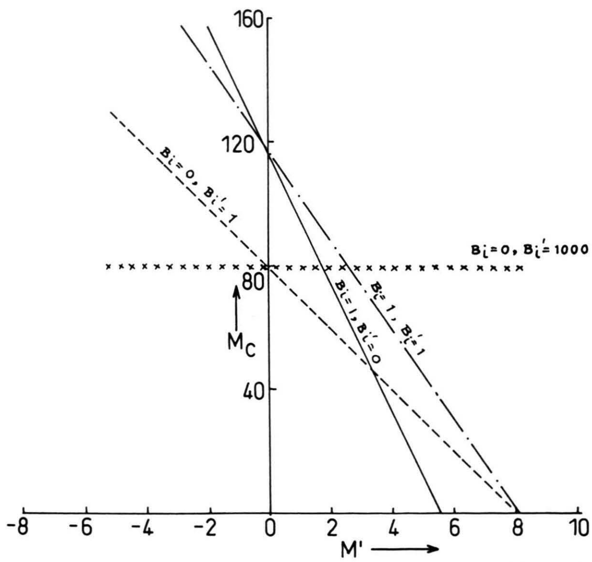

Fig. 6. Variation of M_c with M' for $\tau=0.07$, $Cr=0.0001$ and $B_0=0.1$. —— for $B_i=1$, $B_i'=0$, – · – for $B_i=1$, $B_i'=1$, - - - for $B_i=0$, $B_i'=1$, × × × × × for $B_i=0$, $B_i'=1000$.

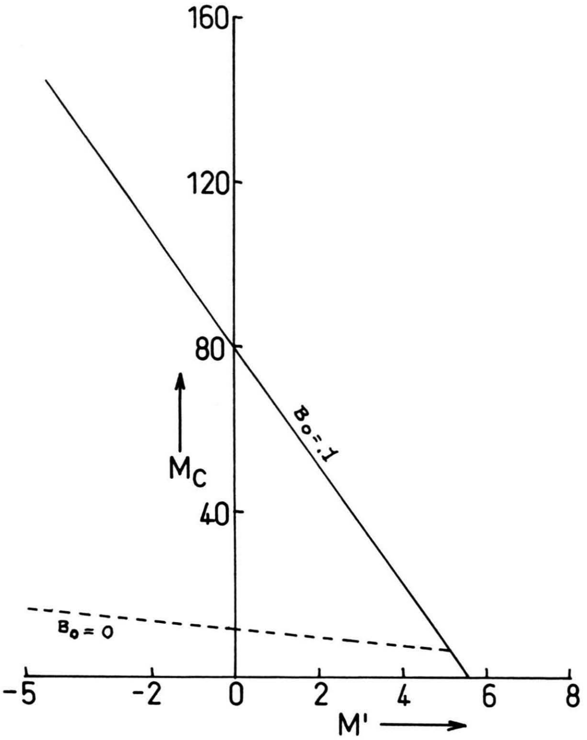

Fig. 7. M_c versus M' for $\tau = 0.07$, $Cr = 0.001$, $B_i = B_i' = 0$. — for $B_0 = 0.1$, - - - for $B_0 = 0$.

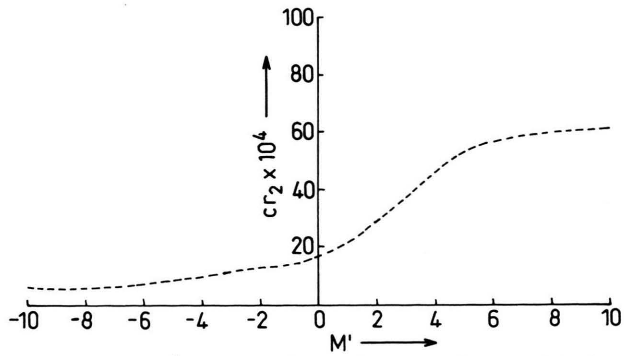

Fig. 8. $Cr_2 \times 10^4$ versus M' for $\tau = 0.07$, $B_0 = 0.1$, $B_i = B_i' = 0$.

It is to be noted that if we consider negative values of M', which corresponds to either salted from below or for the negativity of γ', then Cr increases initially for small values of τ and gradually takes the asymptotic value for large τ. This shows that τ destabilizes the system for $M' < 0$. This result confirms the earlier findings of Sengupta and Gupta [23] in absence of the rotation parameter Y in their paper. Figure 3 shows that Cr_1 decreases with increasing B_i and ultimately takes an asymptotic value for large B_i. Thus, as the conductivity of the free surface increases the system stabilizes. This result was also obtained by McTaggart [18]. Figure 4 depicts the variation of Cr_1 and Cr_2 with B_i for $M' > 0$. It is clear from Figure 4 that the effect of deformation is experienced for small values of B_i', i.e. for an impermeable boundary Cr is maximum. In otherwords, an impermeable boundary enhances the instability mechanism for $M' > 0$. Figure 5 confirms the above stated result more distinctly. Figure 6 shows the variation of M_c with M' for a particular set of combinations of B_i and B_i'. It is clear from the figure that the results obtained here are in agreement with the results of Figs. 3–5. Figure 7 gives the variation of M_c with M' for different values of B_0. Figure 8 shows that Cr_2 increases with M' and takes an asymptotic value for large M'. Thus, for $M' > 0$ the system destabilizes whereas for $M' < 0$ Cr_2 decreases as M' decreases, implying stabilization.

5. Conclusion

We have examined the thermosolutal convection problem in the frame of linearised stability analysis by assuming deformation of the free surface. This study states that surface deformation plays an important role in the calculation of the critical Marangoni number for the stationary convection problem. Further we shall see in the accompanying paper [24] that the frequency of the oscillatory mode and the Crispation number (Cr) are strongly coupled for large values of the latter. To understand the effect of surface deformation on the stability characteristics of this thermosolutal problem in detail, stationary and oscillatory modes of instability in the presence of buoyancy shall be considered in future studies.

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