J. Fluid Mech. (1973), *vol.* 57, *part* 1, *pp.* 103–110
Printed in Great Britain

The stability of a layer of binary gas mixture heated below

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(Received 12 April 1971 and in revised form 5 June 1972)

This paper investigates the Bénard problem in a binary mixture of dilute gases in which an imposed vertical temperature gradient induces a concentration gradient owing to the thermal diffusion effect. The transfer equations are derived by first-order perturbation theory which leads to instability criteria. Numerical results indicate that instability will set in only as stationary convection. This is distinctly different from the cases of liquids and concentrated gases, in which the thermal diffusion (or Soret) effect gives rise to oscillatory instability. It is disclosed in the study that the destabilization of the dilute gas-mixture layer is enhanced by an increase in the thermal diffusion ratio and/or the molecular weight ratio of the species.

1. Introduction

In a multi-component system the momentum flux depends only upon the velocity gradients, while the heat and mass fluxes depend both on the temperature gradient and on the mechanical driving forces (Bird, Stewart & Lightfoot 1960, § 18.4). The diffusion of matter induced by an applied temperature gradient is called the Soret effect in liquid mixtures or the thermal diffusion effect in gaseous mixtures. Coupled with this effect is a mechanical driving force, called the Dufour effect or diffusion-thermo effect, which tends to produce the diffusion of energy resulting from a concentration gradient.

The Bénard problem dealing with the onset of convective instability in a liquid mixture on which both temperature and concentration gradients are imposed has recently been investigated theoretically (Veronis 1965; Sani 1965; Nield 1967) and experimentally (Shirtcliffe 1967). Hurle & Jakeman (1969) predicted and later (1971) demonstrated that oscillatory instability may be induced in the thermosolutal liquid owing to the action of the Soret effect. In the case of gases, however, overstable oscillations were predicted only in concentrated mixtures. Their analysis is valid for the mixtures of liquids and dense gases but is not justified for the mixtures of dilute gases. In the case of dilute gases, the transport equations and coefficients are developed on the basis of kinetic theory. While the Dufour effect is still negligible, the thermal diffusion effect on mass transfer and the heat flux due to interdiffusion together with the ordinary thermal and mass diffusion mechanisms play important roles in the transport phenomena.

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This paper investigates the Bénard problem in a binary mixture of dilute gases in which an imposed vertical temperature gradient induces a concentration gradient owing to thermal diffusion. The transport equations are derived by first-order perturbation theory, which leads to instability criteria. The numerical results reveal that instability will set in only as stationary convection. This is in sharp contrast to the finding of Hurle & Jakeman (1971) that the Soret effect can give rise to oscillatory instability in liquids and concentrated gases. The effect of thermal diffusion on the stability criteria is examined.

2. Formulation of the problem

The system to be studied consists of a binary gas mixture confined between two infinite horizontal plates, the lower one being at a higher temperature T_a and the upper one at lower temperature T_b. The distance between the plates is h. Cartesian co-ordinates (x, y, z) are employed with the origin fixed at the midplane between the plates and z measuring the distance normal to the plates.

The temperature gradient in a binary fluid mixture induces a redistribution of concentration. According to Bird, Curtiss & Hirschfelder (1955), one can write the mass flux $\mathbf{J}_1$ as the sum of terms describing ordinary diffusion and thermal diffusion as

$$\mathbf{J}_1 = -(n^2/\rho)\, m_1 m_2 D_x[\nabla C + k_T \nabla(\ln T)], \tag{1}$$

where $n = n_1 + n_2$. The subscripts 1 and 2 denote the heavier and lighter species, respectively, ρ the mixture density, n the number density (number of molecules per unit volume), m_1 and m_2 the mass of a molecule of each species, C the molecular concentration fraction of the heavier species ($= n_1/n$), D_x the diffusion coefficient, k_T the thermal diffusion ratio and T the temperature.

The heat flux $\mathbf{q}$ in the mixture consists of three contributions associated with the molecular diffusion, interdiffusion and the Dufour or diffusion-thermo effect. The Dufour heat flux is usually of minor importance. One can write the total heat flux relative to the mass-average velocity as

$$\mathbf{q} = -K\nabla T + C_p T(m_1 - m_2)\, n D_x[\nabla C + k_T \nabla(\ln T)], \tag{2}$$

in which K and C_p are the thermal conductivity and specific heat under constant pressure of the mixture, respectively.

If the confining plates are impermeable, so that $\mathbf{J}_1$ vanishes, then (1) yields

$$\frac{\partial C}{\partial z} = -\frac{k_T}{T}\frac{\partial T}{\partial z}. \tag{3}$$

A concentration gradient is therefore established at the plates and will extend into the entire mixture. In other words, an applied temperature gradient can establish a concentration gradient owing to the thermal diffusion effect. If a uniform vertical temperature gradient α is imposed on the mixture, i.e.

$$T = T_m - \alpha z, \tag{4}$$

where $T_m = \frac{1}{4}(T_a + T_b)$ and $\alpha = (T_b - T_a)/h$, the integration of (3) yields the induced concentration as

$$C = C_m + \frac{k_T \alpha}{T_m}\left[z + \frac{\alpha}{2T_m} z^2 + \dots\right] \tag{5}$$

for $(\alpha z/T_m)^2 < 1$, in which C_m is the value of C at $z = 0$. For a thin layer only the first term in square brackets in (5) is retained.

The density of the mixture ρ can be expressed as the sum of the partial densities as

$$\rho = n_1 m_1 + n_2 m_2. \tag{6}$$

The equation of state for an ideal gas reads

$$P = n\nu T, \tag{7}$$

in which ν is the Boltzmann constant. The combination of (6) and (7) together with the definition $n = n_1 + n_2$ yields

$$\rho = \frac{P}{\nu T}[m_2 + (m_1 - m_2)\,C] = \frac{P}{\bar{R}T}[M_2 + (M_1 - M_2)\,C], \tag{8}$$

where $\bar{R}$ is the universal gas constant and M_1 and M_2 are the molecular weights of the two species.

Since the mixture density is a function of both temperature and concentration, one can write a first-order Taylor-series expansion for ρ with respect to its mean value $\rho_m(T_m, C_m)$ in the form

$$\rho = \rho_m + (\partial\rho/\partial T)_m\,(T - T_m) + (\partial\rho/\partial C)_m\,(C - C_m),$$

where the subscript m refers to the base state (T_m, C_m). With the aid of (7) and (8), this equation can be rewritten as

$$\rho = \rho_m[1 - \beta_T(T - T_m) + \beta_c(C - C_m)], \tag{9}$$

in which β_T denotes the thermal cubical expansion coefficient and β_c relates density increases to increases in solute concentration:

$$\beta_T = -1/T_m, \quad \beta_C = (m_2 n/\rho)\,(M_1/M_2 - 1). \tag{10}$$

Let w', C' and T' be the z velocity component, the concentration and temperature of the perturbation, respectively. If (9) and the continuity equation are used to eliminate the pressure terms in the momentum equations, the Boussinesq approximation for a quasi-incompressible fluid gives the z component of the momentum, energy and mass-transfer equations as

$$\frac{\partial}{\partial t}\nabla^2 w' = g\beta_T\left(\nabla^2 T' - \frac{\partial^2 T'}{\partial z^2}\right) - g\beta_C\left(\nabla^2 C' - \frac{\partial^2 C'}{\partial z^2}\right) + \nu\nabla^4 w', \tag{11}$$

$$\frac{\partial T'}{\partial t} - \alpha w' = k\nabla^2 T - n\frac{T_m}{\rho_m}(m_1 - m_2)\,D_x\left(\nabla^2 C' + \frac{k_T}{T_m}\nabla^2 T'\right) \tag{12}$$

and

$$\frac{\partial C'}{\partial t} + \frac{\alpha k_T}{T_m}w' = D_x\left(\nabla^2 C' + \frac{k_T}{T_m}\nabla^2 T'\right) \tag{13}$$

respectively. Here, t is the time, k the thermal diffusivity and g the acceleration due to gravity.

For convenience in the analysis, a new variable defined by

$$H' = C' + k_T T'/T_m \tag{14}$$

is introduced. The appropriate boundary conditions at $z = \pm\frac{1}{2}h$ can be expressed as

$$w' = \partial w'/\partial z = 0, \quad T' = 0, \quad \partial H'/\partial z = 0, \tag{15}$$

which describe the situation at the two rigid boundaries, namely no flow, specified surface temperature, and zero mass flux.

With the introduction of the non-dimensional variables

$$\eta = x/h, \quad \xi = y/h, \quad \zeta = z/h, \quad \tau = kt/h^2$$

together with the non-dimensional quantities

$$\begin{aligned} w' &= (kT_m/h^2\alpha)\, W(\tau,\zeta) f(\eta,\xi), \\ T' &= T_m \Lambda(\tau,\zeta) f(\eta,\xi), \\ H' &= \frac{kk_T}{D_x}\left(\frac{\rho}{m_2 n}\right)_m H(\tau,\zeta) f(\eta,\xi), \end{aligned}$$

(11) becomes

$$\frac{1}{Pr}(D^2-b^2)\frac{\partial W}{\partial \tau} = -b^2 Ra\Lambda(1+\phi Le/S) + (D^2-b^2)^2 W + Ra\phi H Le b^2 \tag{16}$$

while (12) and (13) are reduced to

$$\partial\Lambda/\partial\tau - W = (D^2-b^2)\Lambda - \phi(D^2-b^2)H, \tag{17}$$

$$S\frac{\partial H}{\partial \tau} - \frac{\partial \Lambda}{\partial \tau} = -W + (D^2-b^2)H\frac{S}{Le}, \tag{18}$$

respectively. Here, D is the differential operator $\partial/\partial\zeta$ and the function $f(\eta,\xi)$ satisfies

$$\frac{\partial^2 f}{\partial \eta^2} + \frac{\partial^2 f}{\partial \xi^2} + b^2 f = 0, \tag{19}$$

in which b is the horizontal wavenumber. Pr is the Prandtl number k/ν, Ra is the Rayleigh number, defined as

$$Ra = \alpha\beta_T g h^4/(k\nu),$$

S is a dimensionless parameter defined by

$$S = \rho_m k/(nm_2 D_x)$$

and Le is k/D_x, the Lewis number. The parameter S is equivalent to the ratio of the Schmidt number to the Prandtl number, or the Lewis number. The thermal diffusion parameter ϕ is defined by

$$\phi = k_T(M_1/M_2 - 1),$$

where M_1 and M_2 are the molecular weights of the heavier and lighter species, respectively. The boundary conditions (15) become

$$H = DW = 0, \quad \Lambda = DH = 0 \tag{20}$$

at $\zeta = \pm\frac{1}{2}$.

3. Solution

An examination reveals that it is a difficult task to solve (16)–(18) subject to the boundary conditions (20) for an expression which predicts the criteria for the onset of stationary or oscillatory convection. Consequently, solutions will be obtained separately for the neutral stability and overstability. The type of convection which would eventually occur in the system can be determined from the criterion that gives the lower critical Rayleigh number. The criteria for the onset of stationary or oscillatory convection can be obtained by solving (16)–(18) by means of the Finlayson's approximate method (Finlayson 1968).

As the first approximation it is postulated that

$$\left.\begin{aligned} W &= A(\tau)\left[\frac{\cosh(\mu\zeta)}{\cosh(\frac{1}{2}\mu)}-\frac{\cos(\mu\zeta)}{\cos(\frac{1}{2}\mu)}\right], \\ \Lambda &= 2^{\frac{1}{2}}B(\tau)\cos(\pi\zeta), \\ H &= 2^{\frac{1}{2}}E(\tau)\cos(2\pi\zeta), \end{aligned}\right\} \tag{21}$$

which satisfy the boundary conditions (20). Here, the eigenvalue is the first root of the transcendental equation

$$\tanh(\tfrac{1}{2}\mu)+\tan(\tfrac{1}{2}\mu) = 0.$$

After substitution of (21), equations (16)–(18) are reduced to the first-order ordinary differential equations

$$\frac{dA}{d\tau} = e_1A-e_2B-e_3E, \tag{22}$$

$$\frac{dB}{d\tau} = 0{\cdot}986A-(\pi^2+b^2)B+\frac{4}{3\pi}\phi(4\pi^2+b^2)E, \tag{23}$$

$$\frac{dE}{d\tau} = -\frac{4}{3\pi S}(\pi^2+b^2)B-\frac{1-\phi}{S}(4\pi^2+b^2)E, \tag{24}$$

in which

$$e_1 = -\frac{(500{\cdot}564+24{\cdot}652b^2+b^4)Pr}{12{\cdot}326+b^2},$$

$$e_2 = -\frac{0{\cdot}986b^2Ra(1+\phi S_0/S)Pr}{12{\cdot}326+b^2}, \quad e_3 = \frac{0{\cdot}5495b^2S_0\phi Ra}{12{\cdot}326+b^2}.$$

The characteristic equation of (22)–(24) is

$$\sigma^3+a_1\sigma^2+a_2\sigma+a_3 = 0,$$

where σ is the eigenvalue and

$$a_1 = (\pi^2+b^2)+(S/S_0-\phi)(4\pi^2+b^2)/S-e_1,$$

$$\begin{aligned} a_2 = {} & (\pi^2+b^2)[(4\pi^2+b^2)(S/S_0-\phi+16\phi/9\pi^2)/S-e_1]-e_1(S/S_0-\phi) \\ & \times(4\pi^2+b^2)/S+0{\cdot}986e_2, \end{aligned}$$

$$\begin{aligned} a_3 = {} & (4\pi^2+b^2)[0{\cdot}986(S/S_0-\phi)e_2-(\pi^2+b^2)(S/S_0-\phi/9\pi^2)e_1]S \\ & -1{\cdot}315(\pi^2+b^2)e_3/S. \end{aligned}$$

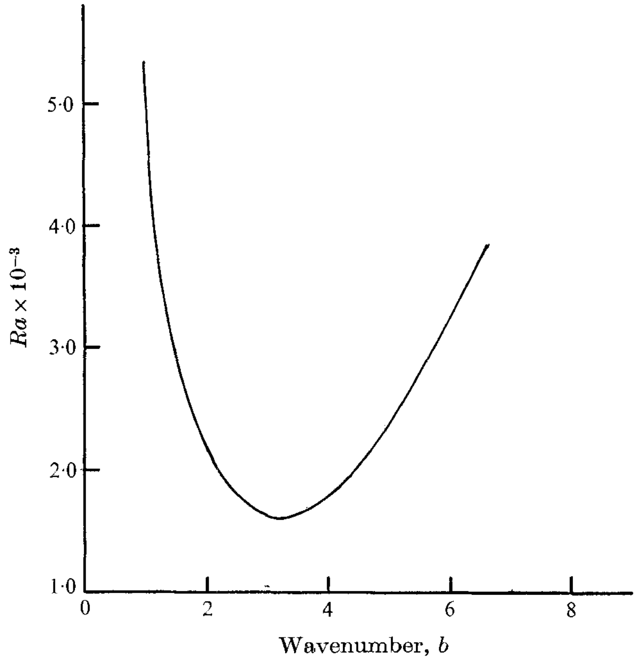

FIGURE 1. Rayleigh number versus wavenumber for $S = 2{\cdot}5$ and $\phi = 0{\cdot}2$.

It can be shown that the condition for overstability is

$$a_1a_2 - a_3 = 0, \tag{25}$$

which yields a relationship between the overstability Rayleigh number and the wavenumber b. The corresponding frequency is

$$\Omega = a_2^{\frac{1}{2}}. \tag{26}$$

The criterion for stationary convection is

$$a_3 = 0. \tag{27}$$

4. Results and discussion

The values of the Prandtl and Lewis numbers are taken to be 0·7 and 1, respectively, which are typical of most gases. It is known (Grew & Ibbs 1952, § 2.6) that the value of ϕ depends upon the ratio of the molecular weights, the molecular diameters and the nature of intermolecular forces. At STP the values of ϕ are 0·6, 0·441, 0·049, 0·0007 and −0·00093 for N_2–He, Ne–Xe, Ne–A, N_2–O_2 and Ne–NH_3 mixtures, respectively. The values of S are 6·0, 1·09 and 1·04 for N_2–He, Ne–NH_3 and N_2–O_2 mixtures, respectively.

Equation (27) is depicted graphically in figures 1 and 2. Figure 1 illustrates the relationship between the Rayleigh number and wavenumber for a gas mixture having $S = 2{\cdot}5$ and $\phi = 0{\cdot}2$. The minimum Rayleigh number is where instability sets in as stationary convection. The critical Rayleigh number is lower than 1717, the value for a pure-gas layer, which corresponds to $\phi = 0$. However, the wavenumber corresponding to the critical Rayleigh number is 3·17. Since this wavenumber is approximately equal to π, the wavelength in a two-dimensional

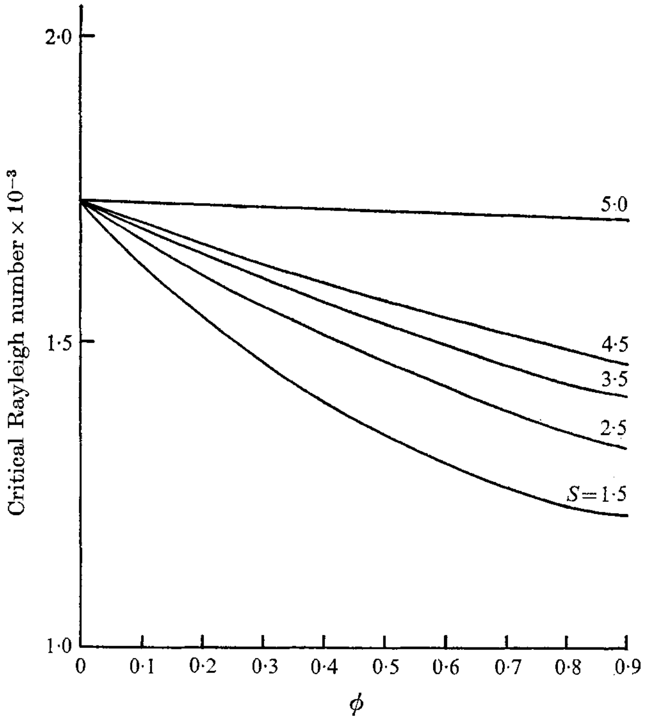

FIGURE 2. Critical Rayleigh number versus ϕ.

disturbance is nearly twice the depth of the gas-mixture layer. Therefore in analogy with the case of a pure-gas layer in the presence of a temperature gradient, the cells generated in a vertical plane bounded by the solid surfaces and by neighbouring upward and downward currents would be nearly square. Equation (25) has also been examined numerically for overstability. The *Ra vs. b* curve for overstability, if plotted in figure 1, would lie above the existing curve for neutral stability. This indicates that instability will set in only as stationary convection. No overstability is found to occur within the ranges of values of the parameters being studied: $S \leqslant 50$ and $-0{\cdot}001 \leqslant \phi \leqslant 1{\cdot}0$.

In figure 2 the critical Rayleigh number is plotted against ϕ with S as a parameter. It is seen in the figure that, for a given value of S, the critical Rayleigh number decreases with an increase in ϕ, indicating destabilization of the layer owing to the thermal diffusion effect, which is enhanced as the thermal diffusion ratio k_T and/or the molecular weight ratio M_1/M_2 increase.

5. Conclusions

A necessary condition for linear-theory overstability is the existence of two opposing forces: a destabilizing temperature gradient in conjunction with rotation, surface tension or a stabilizing solutal gradient. While the Soret effect gives rise to overstable solutions of the thermosolutal Bénard problem, instability can set in only as stationary convection in a layer of binary gas mixture owing to the action of thermal diffusion effect. Thermal diffusion is a destabilizing force which aids the temperature gradient to promote the occurrence of stationary stability in the gas mixtures at a lower critical Rayleigh number than in a single-component gas layer. The destabilization of the layer due to the thermal diffusion effect is enhanced as the thermal diffusion ratio and/or the molecular weight ratio increase.

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