

## Convective instabilities in a closed vertical cylinder heated from below. Part 2. Binary gas mixtures

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Non-reactive binary gas mixtures (Xe + He, SiCl<sub>4</sub> + H<sub>2</sub>) have been investigated for convective instabilities in closed vertical cylinders with conductive walls heated from below. Critical Rayleigh numbers  $N_{Ra}^i$  for the onset of various convective modes (including the onset of marginally stable and periodic flow) have been determined with a high resolution differential temperature sensing method. It is found that the second component can significantly alter the hydrodynamic state of the fluid compared to the monocomponent behaviour. Considerably lower critical *thermal* Rayleigh numbers for steady and time dependent convective modes are observed. The Xe : He system shows stable oscillatory modes similar to those observed in monocomponent gases (periodic disturbances of the mean flow,  $T_0 \approx 5$  s) from  $N_{Ra} = 713$  to 780, where a new mode with  $T_0 = 15$  s sets in. The frequency of these slower temperature oscillations can be fitted by an equation of the form  $f^2 = k'(N_{Ra} - N_{Ra}^0)$  where  $k'$  and  $N_{Ra}^0$  are constants, which supports the contention that these oscillations are the result of vertical vorticity. For SiCl<sub>4</sub> : H<sub>2</sub> the high frequency oscillations occur only as a transient mode eventually evolving into the low frequency mode characteristic of binary gas mixtures. This low frequency state is degenerate with a stable time-independent state over a considerable range of  $N_{Ra}$ . Finite amplitude perturbations can lead to (1) transient oscillatory phenomena accompanied by reorientations of the roll cells with mean periods of 3–5 min; and (2) stable oscillatory flow at  $N_{Ra}$ 's considerably below  $N_{Ra}^{osc}$ . The unique behaviour of these binary fluids is tentatively assigned to thermal diffusion.

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### 1. Introduction

Studies of the convective behaviour of a multicomponent fluid are relatively new. The first experiments (Legros, Van Hook & Thomaes 1968*a, b*) were stimulated by the thought that thermal diffusion in a temperature gradient could alter the stability of a multicomponent fluid layer heated from below. Since then, a number of experiments have been performed and theories developed which confirm this hypothesis. For a review of these and other works concerned with the convective behaviour of multicomponent fluids, see Schechter, Velarde & Platten (1973). It appears, however, that there are several large gaps in the current understanding of binary fluids.

For instance, the large thermal Rayleigh number convective behaviour is not known for a binary fluid in which thermal diffusion plays an important role. It has been suggested by several authors (Legros *et al.* 1968*b*; Platten & Chavepeyer 1973)

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	$\mu \times 10^{-6}$ (g/cm s)†	$k \times 10^{-6}$ (cal/cm s °K)†	$c_p$ (cal/g °K)	$k_T$
Xe:He (0.8:0.2 mole ratio)	247.9‡	36.75‡	0.0472	0.042§
SiCl <sub>4</sub> :H <sub>2</sub>	?	?	—	?

†  $T = 313$  K.    ‡ Touloukian (1970).    § Atkins *et al.* (1939).

TABLE 1. Transport properties of gases used in these experiments

that the mass density gradient which results from thermal diffusion is ‘...destroyed upon the onset of convection because characteristic relaxation times for thermal diffusion are much longer than those for convection’ (Legros *et al.* 1968*b*). This suggests that for supercritical Rayleigh numbers, the convective behaviour of a binary fluid should not differ significantly from the behaviour of a similar mono-component fluid. In view of this rather intriguing conclusion, we have conducted a series of experiments which are designed to study the time-independent and time-dependent convective behaviour of a laterally confined non-reactive binary gas (Xe:He and SiCl<sub>4</sub>:H<sub>2</sub>). Deviations from monocomponent behaviour for large Rayleigh numbers are detected by comparing the results of these experiments with similar results from the preceding paper (Olson & Rosenberger 1979), hereafter called part 1.

## 2. Experimental considerations

### 2.1. Apparatus

The experimental apparatus, convection cell and detection scheme are basically the same as that described in part 1. The main experimental differences between part 1 and part 2 occur in the preparation of the binary mixture to be discussed in §2.3.

### 2.2. Choice of systems

Quantitative convection studies require a precise knowledge of the thermophysical properties of the fluid. These quantities are needed for an accurate determination of the thermal Rayleigh number,

$$N_{Ra} = g \frac{\alpha \rho^2 c_p}{\mu k} \frac{\Delta T a^4}{h}. \quad (1)$$

See part 1 for description of variables. The viscosity  $\mu$  and thermal conductivity  $k$  of many pure species are known with reasonable precision (approximately 1–2%), but in most cases, they are not known for mixtures of gases. Fortunately there are some exceptions, see table 1. Binary mixtures of the noble gases are well characterized, the transport properties  $\mu$  and  $k$  of these mixtures having been measured *and* calculated (Touloukian 1970) as a function of composition and temperature. In addition, the thermal diffusion ratio  $k_T$  has been measured as a function of composition for all the various binary mixtures of the noble gases (Atkins, Bastick & Ibbs 1939).

The system SiCl<sub>4</sub>:H<sub>2</sub> (non-reactive near room temperature) was chosen for its importance in the silicon industry, but unfortunately it is not well characterized with respect to the transport properties of the mixture. However, if one employs the

method of (Ulybin 1959; Ulybin, Burgrov & Il'in 1966) to account for the changing mean temperature of the fluid (see part 1, §2.3) then accurate *relative* measurements (e.g.  $N_{Ra}^{osc}/N_{Ra}^{cr} \equiv \Delta T^{osc}/\Delta T^{cr}$ ) are indeed possible and, as will be shown, very informative.

### 2.3. Preparation of gas mixtures

For the preparation of a nonreactive binary gas mixture, known quantities (accurate to within 1%) of the two components are introduced into a glass mixing vessel. The vessel also contains several teflon chips, so that the slow diffusional mixing process can be aided by periodic mechanical agitation. The procedure used for filling the convection cell with the binary mixture is identical to that described in part 1.

## 3. Experimental results and discussion

### 3.1. Time-independent behaviour

The typical time-independent behaviour of a binary gas mixture† is summarized in figures 1(*a*, *b*). The fluid is a mixture of Xe and He, the mole fraction of Xe being 0.80.

The ordinate  $2T'$  is the transverse temperature difference as measured between pairs of small (0.25 mm) thermistors immersed at symmetric points within the fluid. When the fluid is at rest,  $2T' = 0$ . When convection is present,  $2T'$  deviates from zero, and the sign of  $2T'$  gives the direction of flow. See part 1 for further details.

The abscissa of the graph in figure 1 is the *thermal* Rayleigh number  $N_{Ra}$  calculated from the transport properties of the *mixture*. Hence the mixture is treated as a monocomponent fluid, so that deviations from monocomponent behaviour can be attributed to properties of the binary mixture (such as thermal diffusion and doubly diffusive phenomena).

A comparison of figure 1 with figure 7(*a*) in part 1 reveals several interesting results.

(1) The basic flow geometry is essentially unchanged. For slightly supercritical Rayleigh numbers, the planform is a unicellular asymmetric roll. For larger Rayleigh numbers the flow reverses direction in the lower two-thirds of the cell. This vertically asymmetric, 2-roll planform persists with few variations all the way up to the onset of time-dependent convection.

(2) There is a distinct shift of all three curves to lower values of  $N_{Ra}$ . The first critical Rayleigh number  $N_{Ra}^1$  has been shifted by approximately 20% to a value of 180. The zero-crossing of  $2T'$  for the thermistor pairs 2–3 and 6–5 occurs at around 230. This again is about 20% below the corresponding point ( $N_{Ra}^2 = 280$ ) in figure 7(*a*), part 1. In other words, this binary system appears to be less stable (by approximately 20%) than a fluid-dynamically similar monocomponent system. Our error estimates indicate that the thermal  $N_{Ra}$  is accurate to within 7–8%. This estimate is based upon a maximum error of 0.5% in the mole fraction of Xe.

A qualitative understanding of this shift can be developed by considering the effects of thermal diffusion. As is well known, an initially homogeneous binary fluid mixture will tend to segregate when subjected to a temperature gradient; complete segregation being limited by back transport due to ordinary concentration diffusion. For almost all binary gas mixtures, the heavier component moves toward the colder region.

† Liquid binary mixtures are expected to behave differently, mainly because of the large deviation from unity of the Prandtl number  $N_P$ , and the Schmidt number  $N_{Sc}$ .

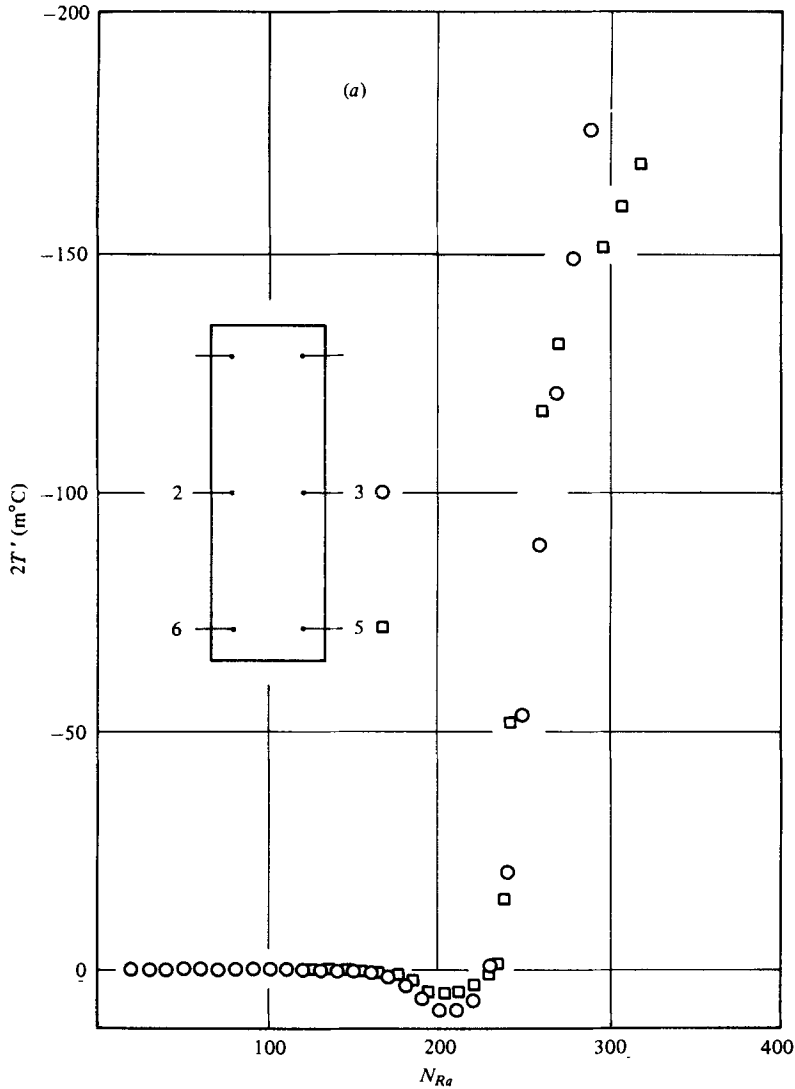


FIGURE 1 (a). For legend see facing page.

Applied to the present system, one would then expect the concentration of Xe(He) to increase in the upper (lower) portion of the cell, respectively. For a vertical  $\Delta T$ , there is now an additional source for an adverse density gradient. The net effect is to decrease, for a given  $\Delta T$ , the overall stability of the fluid column.

One can attempt to obtain a semiquantitative understanding of this effect by comparing the linear contributions of thermal volume expansion and thermal diffusion to the overall density gradient. To facilitate this comparison, equation (1) is rewritten in the following form:

$$N_{Ra} = \frac{ga^4}{\nu\kappa h} \cdot \frac{\Delta\rho}{\rho}, \quad (2)$$

where  $\nu = \mu/\rho$  and  $\kappa = k/c_p\rho$ .

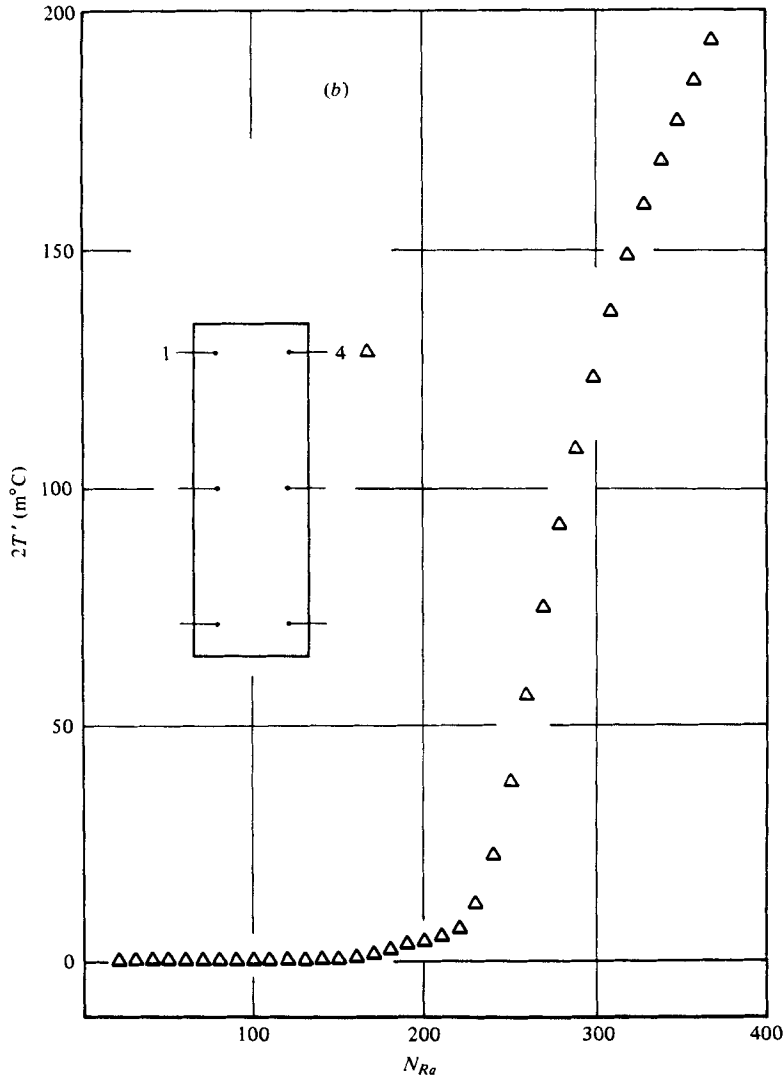


FIGURE 1. (a, b) Measurements of the transverse temperature difference  $2T'$  taken simultaneously at two symmetric locations, vs. the thermal Rayleigh number  $N_{Ra}$ , in an 0.8:0.2 mole fraction mixture of Xe:He,  $5.45 \times 10^{-3} \text{ g cm}^{-3}$ ,  $\alpha = 1 \text{ cm}$  and  $h/a = 6$ .

Let us suppose that the total relative density difference  $(\Delta\rho/\rho)|_{\text{total}}$  is the relevant parameter which determines the onset of marginally stable flow and that the critical  $(\Delta\rho/\rho)|_{\text{total}}^{\text{cr}}$  is given by

$$\frac{\Delta\rho}{\rho}\Big|_{\text{total}}^{\text{cr}} = \frac{\Delta\rho}{\rho}\Big|_E + \frac{\Delta\rho}{\rho}\Big|_T, \quad (3)$$

where the expansive term is (for an ideal gas)

$$\frac{\Delta\rho}{\rho}\Big|_E = \frac{1}{\rho} \frac{\partial\rho}{\partial T} \Delta T^{\text{cr}} = \alpha \Delta T^{\text{cr}} = \frac{\Delta T^{\text{cr}}}{T}, \quad (4)$$

and the thermal diffusion contribution is

$$\left. \frac{\Delta\rho}{\rho} \right|_T \simeq \frac{\Delta X_A}{X_A} = \frac{k_T}{X_A} \ln \left( 1 + \frac{\Delta T^{\text{cr}}}{T} \right) \simeq \frac{k_T}{X_A} \frac{\Delta T^{\text{cr}}}{T}. \quad (5)$$

In (5),  $X_A$  is the mole fraction of the heavier, more abundant species. The derivation of equation (5) can be found in Grew & Ibbs (1952). One then assumes that

$$N_{Ra}^{\text{cr}} = \frac{ga^4}{\nu\kappa h} \cdot \left. \frac{\Delta\rho}{\rho} \right|_{\text{total}}^{\text{cr}} = \frac{ga^4}{\nu\kappa h} \cdot \frac{\Delta T^{\text{cr}}}{T} \cdot \left( 1 + \frac{k_T}{X_A} \right) = N_{Ra}^{\text{cr}}|_T \cdot \left( 1 + \frac{k_T}{X_A} \right). \quad (6)$$

Application of equation (6) to the Xe:He system of figure 1 setting  $N_{Ra}^{\text{cr}} = 230$ , yields

$$N_{Ra}^{\text{cr}}|_T = 218.4 \pm 11,$$

which does not compare well with the results in figure 1 where  $N_{Ra}^{\text{cr}} = 180 \pm 14$ . If we attempt to include the effects of the Schmidt and Prandtl numbers by using the results of Platten & Chavepeyer (1977) with the assumption that their analysis for finite, horizontal stress-free boundaries is applicable here, then equation (6) becomes

$$N_{Ra}^{\text{cr}} = N_{Ra}^{\text{cr}}|_T \cdot \left( 1 + \frac{k_T}{X_A} \cdot \frac{N_{Pr} + N_{Sc}}{N_{Pr}} \right). \quad (7)$$

Notice that even when  $N_{Pr} = N_{Sc}$  (i.e. heat and mass diffuse at the same rate) equations (6) and (7) yield different results. That is, neglecting the complication from double-diffusive phenomena, the combination of thermal expansion and thermal diffusion has a greater effect than would be expected from a linear addition of their separate contributions. For the Xe:He system in figure 1,  $N_{Pr} = 0.32$  and  $N_{Sc} = 0.10$ . Equation (7) then yields  $N_{Ra}^{\text{cr}} = 215.4 \pm 12$ , which still does not compare well with the results in figure 1. A quantitative interpretation of our finding can only be expected from an analysis that also accounts for viscous and thermal interactions with the lateral boundaries which dominate in our case.

### 3.2. Time-dependent behaviour

The steady or time-independent behaviour of these binary gas mixtures was not the main focus of this research. The shift of  $N_{Ra}^{\text{cr}}$  due to the destabilizing (or stabilizing) influence of thermal diffusion has already been well established (Legros *et al.* 1968*a, b*). But the time-dependent behaviour of fluids for which thermal diffusion acts destabilizing (and  $N_{Ra}^{\text{osc}} \gg N_{Ra}^{\text{cr}}$ ) has received virtually no attention. As mentioned in the introduction the work of Legros *et al.* (1968*b*) and Platten & Chavepeyer (1977) suggests that for supercritical Rayleigh numbers, the convective behaviour of a binary fluid should not differ significantly from that of a similar monocomponent fluid. As we shall see, however, our findings don't support this expectation.

The time-independent behaviour of the  $\text{SiCl}_4:\text{H}_2$  system is shown in figure 2. The nominal thermal  $N_{Ra}$  used for the abscissa is to a good approximation proportional to the actual thermal  $N_{Ra}$ , so that ratios of  $N_{Ra}$  (e.g.  $N_{Ra}^{\text{osc}}/N_{Ra}^{\text{cr}}$ ) should be quantitatively reliable. For increasing values of  $N_{Ra}$ , oscillations set in at a nominal  $N_{Ra}^{\text{osc}} = 900$ , as shown in figure 3. These oscillations are fluctuations of the transverse temperature difference  $2T''$  as measured by two thermistors placed at symmetric points in the

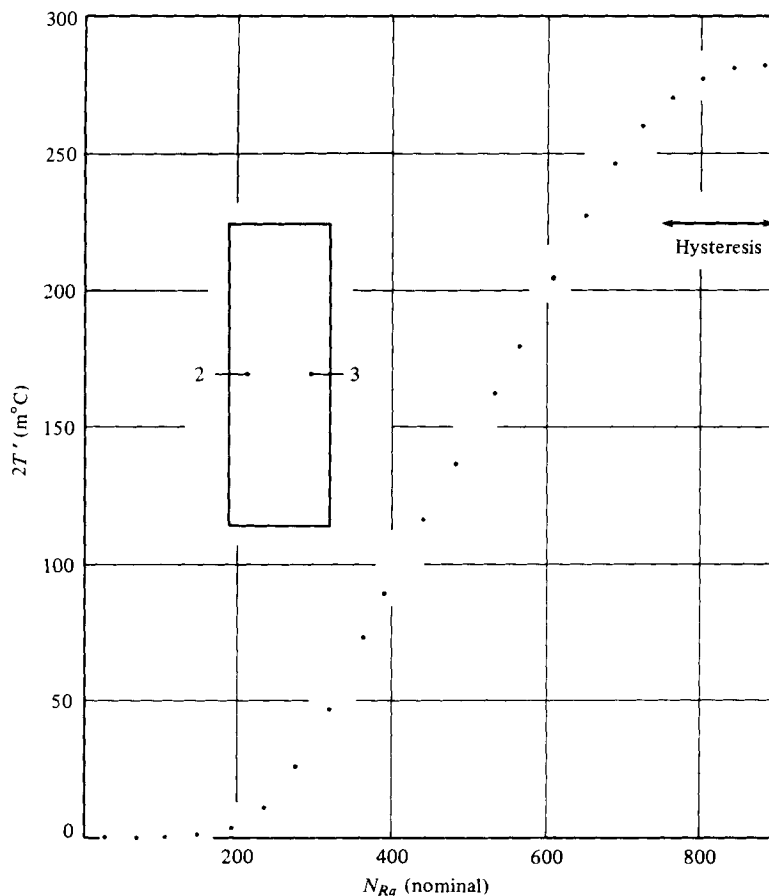


FIGURE 2. Transverse temperature difference  $2T'$  vs. a nominal thermal  $N_{Ra}$  for the non-reactive equimolar gas mixture of  $\text{SiCl}_4$  and  $\text{H}_2$ ,  $3.3 \times 10^{-3} \text{ g cm}^{-3}$ ,  $a = 1 \text{ cm}$  and  $h/a = 6$ .

midplane of the cell. The ratio  $N_{Ra}^{\text{osc}}/N_{Ra}^{\text{cr}} \approx 3.9$  is considerably lower than the corresponding value for a mono-component fluid where  $N_{Ra}^{\text{osc}}/N_{Ra}^{\text{cr}} \approx 6$  (see part 1).

The results for the Xe:He system are quite similar to those for the  $\text{SiCl}_4$ : $\text{H}_2$  system. The onset of oscillations of the transverse temperature difference  $2T'$  in the midplane of the cell is shown in figure 4(a). This state was attained by small positive increments ( $\sim 0.05 \text{ }^\circ\text{C}$ ) of the vertical  $\Delta T$ . The critical Rayleigh number for the onset of these oscillations is  $N_{Ra}^{\text{osc}} = 713$ . Comparison of this number with the corresponding number for monocomponent fluids ( $N_{Ra}^{\text{osc}} = 1348$ , see part 1) shows that the presence of the second component has significantly altered the hydrodynamic state of the fluid for a given thermal  $N_{Ra}$ . In addition, the ratio  $N_{Ra}^{\text{osc}}/N_{Ra}^{\text{cr}} = 4$ , as it was for  $\text{SiCl}_4$ : $\text{H}_2$ . From these results we are forced to the conclusion that the absolute ( $N_{Ra}^{\text{osc}} = 713$ ) and the relative ( $N_{Ra}^{\text{osc}}/N_{Ra}^{\text{cr}} = 4$ ) stability of these binary mixtures is lower than that of a similar, monocomponent fluid even though the fluid is far from the point of marginal instability  $N_{Ra}^{\text{cr}}$ .

There are other characteristics of these binary fluids which distinguish them from monocomponent fluids. The transient temperature oscillations in figure 3 are similar

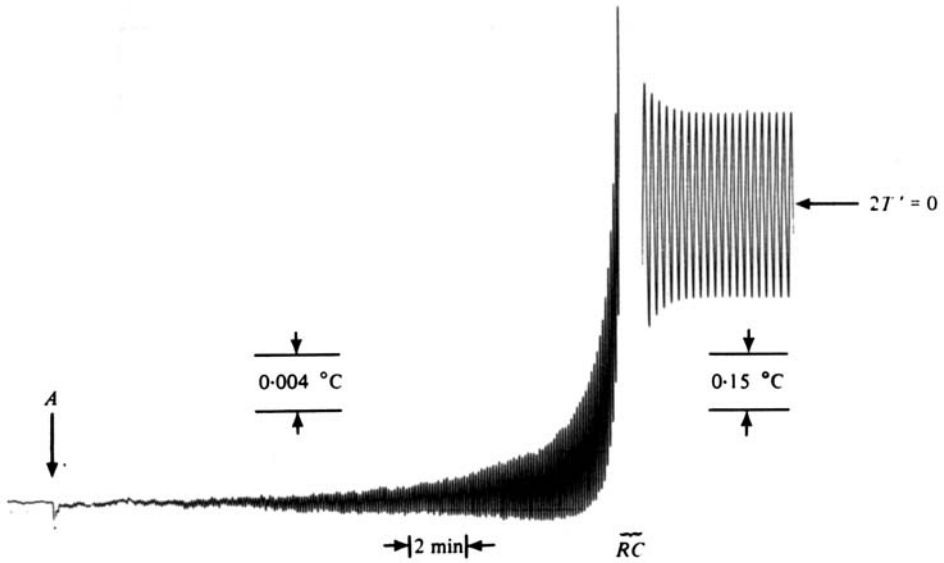


FIGURE 3. Onset of oscillations of the transverse temperature difference  $2T'$  in the midplane of the cell for  $\text{SiCl}_4:\text{H}_2$  (see figure 2). Nominal  $N_{Ra}^{\text{osc}} = 900$ ,  $RC$ -range change, time scale uninterrupted. At point  $A$ ,  $N_{Ra}$  was increased by about 0.5%.

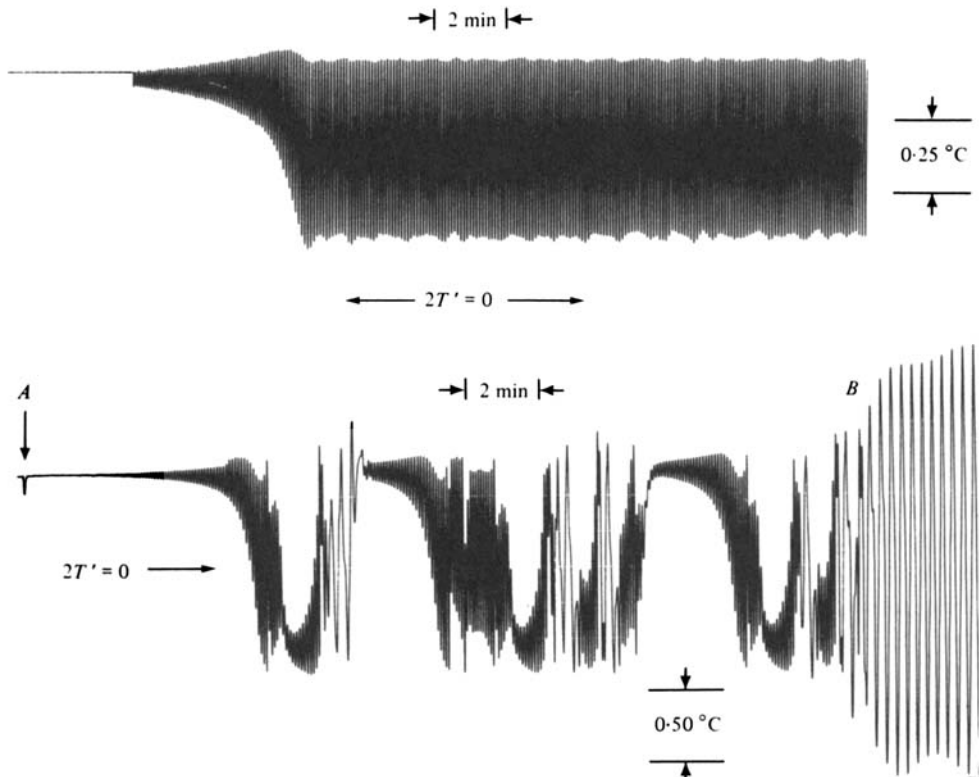


FIGURE 4. (a) Quasi-steady approach to the onset of time-dependent convective flow at  $N_{Ra}^{\text{osc}} = 713$  for  $\text{Xe}:\text{He}$  (see figure 1). (b) 'Finite amplitude' approach at  $N_{Ra} = 663$ .



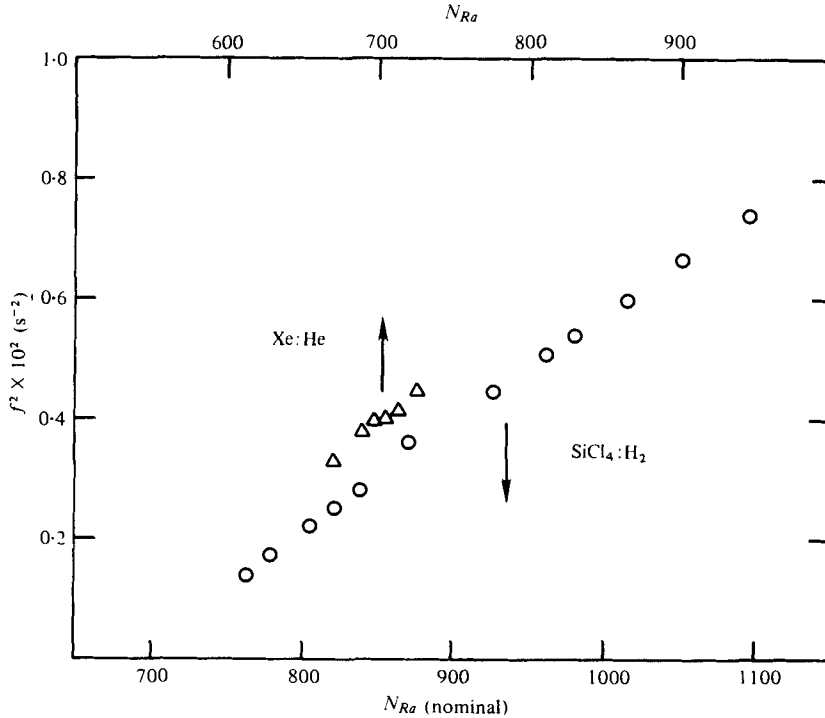


FIGURE 5. Frequency (squared) of  $2T'$  oscillations vs. the thermal  $N_{Ra}$  for the mixtures  $\text{SiCl}_4:\text{H}_2$  (see figure 2 and 3) and  $\text{Xe}:\text{He}$  (see figure 1).

to the steady temperature oscillations observed in monocomponent gases (see part 1). In both cases, the oscillations are symmetric disturbances of the mean flow with a period of  $T_0 = 5$  s. However, the final oscillatory state is one which is characteristic of binary gas mixtures. The distinguishing features of these 'binary oscillations' are: (1) a large amplitude,  $\Delta(2T') > 2T'|_{N_{Ra}^{osc}}$ ; (2) a long period,  $T_0 = 15\text{--}16$  s; and (3) a symmetric fluctuation about  $2T' = 0$ .

The last feature could be interpreted as either a rotation of the asymmetric roll configuration about the axis of the convection cell; or a periodic reversal of the flow within a roll, the orientation of the roll configuration being fixed. Measurements of  $T_0$  vs.  $N_{Ra}$  show that the first mechanism is, most likely, the correct one. In figure 5 we have plotted the square of the frequency vs.  $N_{Ra}$  for  $\text{SiCl}_4:\text{H}_2$  and  $\text{Xe}:\text{He}$ . The data for  $\text{SiCl}_4:\text{H}_2$  can be fit by an equation of the form

$$f^2 = k'(N_{Ra} - N_{Ra}^0),$$

where  $k'$  and  $N_{Ra}^0$  are constants. This functional relation is the same as that predicted for the Rayleigh-Bénard geometry by Busse (1972) for oscillations which accompany the onset of vertical vorticity. This result, coupled with the previous observations, suggests that these 'binary oscillations' are caused by a rotation of the vertically asymmetric roll-cell pattern about the vertical axis of the convection cell. Irregardless of the mechanism which generates these temperature oscillations, they are unique to the binary gases.

Another interesting and most fascinating feature of these binary gases is the hysteresis which accompanies the onset (and decay) of the oscillatory state. For  $\text{SiCl}_4:\text{H}_4$  the 'binary' oscillatory state, once established, is stable (for days) for  $N_{Ra} > 750$ . Conversely, if the fluid is already in a time-independent state, this state is stable for  $N_{Ra} < 900$ . Consequently, the region  $750 < N_{Ra} < 900$  is characterized by two states (as indicated in figure 2); one which is time-independent, the other time-dependent or oscillatory.

The time-dependent modal behaviour of the  $\text{Xe}:\text{He}$  system is somewhat different from that of  $\text{SiCl}_4:\text{H}_2$ , but a region of hysteresis of bistability still exists. For  $\text{SiCl}_4:\text{H}_2$ , the high frequency or 'monocomponent' oscillations ( $T_0 \simeq 5$  s) were a transient mode eventually evolving into the low frequency ( $T_0 \simeq 15$  s) mode characteristic of binary gases. For  $\text{Xe}:\text{He}$ , the high frequency mode is stable against small perturbations up to at least  $N_{Ra} \simeq 777$  (for  $N_{Ra}$  increasing) at which point the low frequency mode becomes stable. The sequence of events leading to these long period oscillations cannot be retraced by decreasing  $N_{Ra}$  in small amounts. The long period oscillations, once established, are stable for  $N_{Ra} > 650$ . At  $N_{Ra} = 650$  they suddenly disappear and are replaced by a time-independent regime. For decreasing  $N_{Ra}$  the 'monocomponent' oscillatory regime does not appear in this case, a characteristic which is similar to the  $\text{SiCl}_4:\text{H}_2$  system.

The various regimes of flow discussed previously were determined under conditions which were virtually steady state and relatively noise free. As mentioned in part 1, a particular state ( $N_{Ra}$ ) was attained by a series of small increments of  $\Delta T$  and the thermal noise in the experimental system was extremely low. Applications of larger increments showed, however, that the way in which a certain state was approached in monocomponent systems (part 1) had little effect on the final result. But for the binary gases considered here, large increments of  $\Delta T$  can have catastrophic consequences particularly in the region of hysteresis. Figure 4(b) is a strip chart recording of the transverse temperature difference in the mid-plane of the cell. Near point *A*,  $N_{Ra}$  was increased from 600 to 663 within a period of 10 min. Note that the final value of  $N_{Ra}$  is well below  $N_{Ra}^{\text{osc}} = 713$  which was obtained in the careful manner described above (figure 4a). Nevertheless, a time-dependent flow regime evolves.

The transient state between points *A* and *B* in figure 4(b) is an interesting superposition of high frequency ( $T_0 = 5$  s) 'monocomponent' oscillations and quasi-periodic reorientations of the roll configuration about  $2T' = 0$ . Near point *B*, this transient mode decays to a stable 'binary' mode. This occurs at  $N_{Ra} = 663$  which is well below  $N_{Ra} = 780$ , the quasi-steady critical  $N_{Ra}$  for the onset of 'binary' oscillations.

The hysteresis observed here has no resemblance to that observed by Platten & Chavepeyer (1975). They observed hystereses associated with the onset of oscillations at  $N_{Ra}^{\text{cr}}$ . This is a problem of overstability and only occurs when the Soret coefficient is negative. For our binary gas studies, the Soret number (or thermal diffusion ratio  $k_T$ ) is positive; the Principle of Exchange of Stabilities is obeyed and oscillations set in for  $N_{Ra}^{\text{osc}} \gg N_{Ra}^{\text{cr}}$ .

For the monocomponent fluids of part 1 there was virtually no hysteresis associated with the onset or decay of temperature oscillations. This once again suggests that the influence of thermal diffusion plays a definite role in determining the hydrodynamic state of the fluid even when  $N_{Ra} \gg N_{Ra}^{\text{cr}}$ . The hypothesis of Legros *et al*. (1968*b*) and

Platten & Chavepeyer (1973) is intuitively plausible and indeed may be true for the Bénard geometries. But in our geometry small corner regions and the core of roll-cells will be relatively stagnant and not subject to the mixing effects of the convective flow. Thermal diffusion will adversely influence the stability of these regions, even when  $N_{Ra} \gg N_{Ra}^{cr}$ . A mechanism similar to that proposed by Howard (1964) could then be responsible for the generation or onset of oscillatory temperature fluctuations.

#### 4. Summary and conclusions

We have presented and discussed the results of experiments designed to study the convective behaviour of binary gas mixtures.

The time-independent behaviour of the non-reactive binary gases (Xe:He and  $\text{SiCl}_4:\text{H}_2$ ) was similar to that of the monocomponent gases in part 1, except that the  $2T'$  vs.  $N_{Ra}$  curves were shifted to lower values of  $N_{Ra}$ . This shift occurs for all values of  $N_{Ra}^i$ , not just  $N_{Ra}^{cr}$ . In addition, the difference between monocomponent gases and binary gases is even more distinct when one compares their time-dependent behaviour.

These two observations are tentatively assigned to thermal diffusion and hence appear to disprove the predictions of Legros *et al.* (1968*b*) and Platten & Chavepeyer (1973). It may be true that the mass density distribution due to thermal diffusion is destroyed at the onset of motion by convective mixing. Once flow is established, however, its amplitude and behaviour is determined by the respective local density distribution in the boundary layer and stagnant (e.g. corner) regions of the fluid. In these regions, the homogenizing effects of convection flow is less efficient with the result that normal diffusion may still play an important role in determining the convective behaviour of the fluid.

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