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An experimental investigation of thermosolutal convection at marginal stability

By T. G. L. SHIRTCLIFFE
Department of Physics, Victoria University of Wellington,
Wellington, New Zealand

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Experiments are described in which the stability of a liquid stabilized by a gradient of concentration of a solute has been investigated. In all the cases observed, convection began at marginal stability with an oscillatory motion. The boundary of stability in the (R, R_s)-plane departs somewhat from that predicted by the simplest theories; the boundary is a curve, concave to the origin, and the critical Rayleigh number is high. The total rate of transport of solute increases much more rapidly than that of heat when the oscillations begin. The motions begin in cells which are separate from one another.

1. Introduction

The first stability analysis for the case of a fluid containing a vertical gradient of solute concentration in addition to a vertical temperature gradient was given by Vertgeim (1955). This paper considered only the possibility of monotonic instability, when the liquid was contained in a vertical cylinder. A number of analyses has since been published in which different geometries have been considered and the possibility of overstable (oscillatory) motions allowed for; notably those of Veronis (1965), Sani (1965) and Nield (1967). Both Veronis and Sani limited their investigations to the case of a horizontal layer of infinite extent having ideal (free, constant temperature, constant concentration) boundaries above and below, but considered the effects of finite amplitude motions. Nield considered the same geometry for the case of marginal stability only, but in such a way that, at least in principle, a wide variety of boundary conditions could be allowed for.

It appears from these investigations that marginal stability will occur in an infinite horizontal layer with ideal boundaries when

$$R+\frac{P_s}{P}R_s = 27\pi^4/4 \quad \text{(monotonic instability)},$$

or

$$R+\frac{P}{P+1}R_s = 27\pi^4/4 \quad \text{(overstability)},$$

where the second equation is an approximation appropriate to aqueous solutions. The angular frequency ω of the overstable oscillations is given by

$$\omega^2 = \frac{\nu^2}{3P_s^2h^4}\left\{\frac{P_s(P_s-P)}{P(P+1)}(-R_s)-\frac{27\pi^4}{4}\right\}.$$

In the above equations,

$$R = g\alpha\beta h^4/(\nu K) \quad \text{(positive destabilizing)}$$

is the thermal Rayleigh number, and

$$R_s = g\eta\gamma h^4/(\nu K) \quad \text{(positive destabilizing)}$$

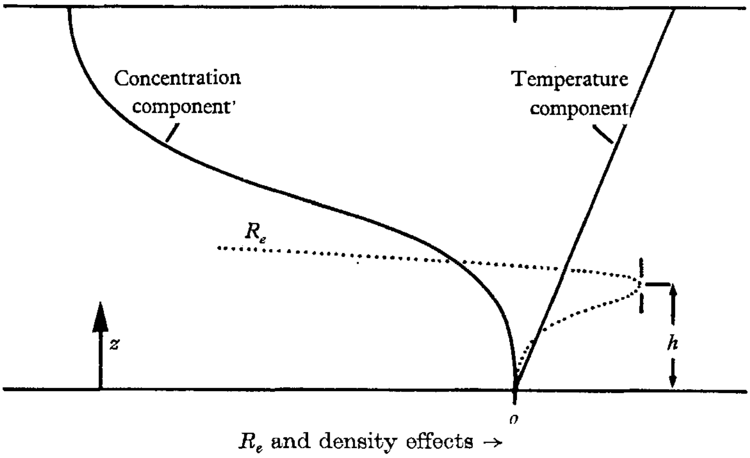

FIGURE 1. Temperature and concentration components of a density profile, and the resulting profile of R_e.

is its solutal analogue; g is the acceleration due to gravity, α is the coefficient of expansion and η its solutal analogue, β is the (constant) temperature gradient and γ the (constant) concentration gradient in the layer, h is the depth of the layer, ν is the kinematic viscosity and K the thermal diffusivity of the liquid, and P is the Prandtl number ν/K and P_s its solutal analogue ν/D, where D is the diffusion constant for the solute.

In the (R, R_s)-plane, the boundaries of stability are therefore straight lines for this ideal case. This paper describes experiments which were designed to test the predicted criterion for overstability by defining the observed boundary of stability for this case.

Of necessity, the conditions under which the experiments were performed were not the same as those assumed in the analyses. In the first place, the surfaces of the tank in which the experiments were conducted were impermeable to the solute (sugar). Thus it was impossible to establish a steady flux of solute through the solvent (water), and the concentration profile was non-linear. Secondly, the convection occurred in a thin bottom layer of the solution, and the boundary conditions were far from ideal; the lower boundary was rigid, with a constant (zero) concentration gradient and a thermal condition which probably approximated constant temperature; while the upper boundary, where the convecting liquid adjoined the stable region, was characterized by continuity of the horizontal velocity, the vertical solute and heat fluxes, the temperature, and the concentration.

The reason for the occurrence of the convection in a thin layer at the bottom of the tank may be seen as follows. Consider the case of a layer of solution in which there is a stabilizing gradient of concentration with impermeable horizontal boundaries, and a linear destabilizing profile of temperature. The contributions to density of the concentration and the temperature are sketched in figure 1. Superimposed is a sketch of the corresponding profile of $R_e(z)$, where

$$R_e(z) = R(z) + \frac{PR_s(z)}{P+1}$$

is the parameter which is expected to control the stability; here R and R_s are defined in terms of the temperature and concentration at the lower boundary and those at z, the level at which R_e is evaluated. Because R_e is proportional to z^3, it is zero at the lower boundary $z = 0$; also it is positive just above it where the temperature effect predominates, and negative at greater z where the solute effect predominates. Hence R_e goes through a maximum at some level $z = h$, and it is to be expected that if its maximum value exceeds some critical value, convection will occur in the layer of depth h.

2. Equipment

The tank used in these experiments had two sides made of plate glass, chosen for good optical flatness. Internally, it was 25 cm long and 6·4 cm wide, the largest vertical faces being glass and the ends Tufnol. It was filled to a depth of 9·7 cm. In order to ensure even heating of the liquid, the base of the tank was made of brass 1·25 cm thick, and it rested on a bath of oil which was electrically heated. The glass sides could be insulated with plastic foam during experiments, to ensure that heat flow was vertical.

A scanning schlieren device rather similar to that of Longsworth (1939) was used to record the profile of concentration gradient in the solution. It was designed for rapid conversion to a standard schlieren system, so that both quantitative and qualitative investigations were possible. The method exploits the fact that the refractive index of sucrose solutions varies with concentration, so that a record of the profile of refractive index gradient can yield the required information on concentration once the system has been calibrated. The refractive index also varies with temperature (so that it is defined, at least approximately, by the resulting density), so this measurement was always made before heating began. The solution was introduced into the tank as a series of layers whose successive concentrations increased downwards; after several hours the contrast between adjacent layers decayed by diffusion to produce a smooth profile.

In order to indicate the onset of convection, a pair of copper-Constantan thermocouples was used in the manner of Fultz *et al.* (1955). The junctions were 0·5 and 1·0 cm from the bottom of the tank; an alternative pair, which could be chosen by switch, were at 1·5 and 2·0 cm. The chosen thermocouples were connected in series with a Tinsley vernier potentiometer used as a potential source variable in 1 μV steps, and the difference passed to a galvanometer amplifier and thence to a strip chart recorder. Full-scale deflexion on the recorder corresponded to about 2 μV change of thermocouple output, and the sensitivity of the system was such that an oscillation of amplitude 5×10^{-4} °C was readily detectable, corresponding to a displacement amplitude of 10^{-4} cm at the highest heating rates.

A miniature-bead thermistor was mounted in contact with the upper surface of the brass slab which formed the base of the tank. This was made one arm of a bridge circuit in which it was balanced against one of 24 resistors chosen by switches. The out-of-balance voltage of the bridge was recorded on a strip chart recorder, and the temperature could thus be determined continuously to 0·01 °C. The ranges were individually compared with a calibrated mercury-in-glass thermometer.

3. Experiments

The experiments were run in three series, the tank being refilled at the start of each. Each experiment of a series began with the liquid isothermal; a concentration gradient profile was measured under these conditions, so that at any later time up to the onset of convection the profile could be deduced from a knowledge of the diffusion constant. In practice, change in the profile during this period was very small.

After the concentration gradient profile had been measured, the heater was switched on and the current adjusted to give the desired rate of increase of temperature at the bottom of the tank. Both this temperature and the amplified output of the differential thermocouples were recorded continuously throughout the experiment. Both records showed an increase with time; the former was kept on scale by switching to successively lower balance resistors in the bridge circuit, the latter by switching in successively greater bucking potentials from the potentiometer. The rate of rise of both records fell off with time, and the heating rate was modified from time to time to maintain progress towards marginal stability. It was not known in advance when this condition would occur in the case of the first run of a series. Thereafter the steady decrease of concentration gradient in the tank, which resulted from diffusion and such limited convection as occurred during individual experiments, was accompanied by a steady decrease in the temperature gradient necessary to bring about marginal stability. Thus for runs other than the first of a series it was possible to predict roughly the critical output of the differential thermocouple. As this condition was approached the rate of increase of temperature gradient was maintained at a modest value.

Marginal stability was reached in different runs after times varying from 15 min to nearly 3 h, depending on the heating rate and the magnitude of the concentration gradient.

4. Nature of the marginal state

A total of 15 runs were made in the three series. The nature of marginal stability was in every case similar to that already described in a preliminary note (Shirtcliffe 1967). When the critical state was reached, an oscillation appeared which was superimposed on the steady rise in output of the differential thermocouple. Initially, this oscillation was quite regular. However, its amplitude generally grew quite rapidly, and this growth was accompanied by increasing disorder. About the time that the oscillation could be said to have become irregular, it became possible to see a cellular structure near the bottom of the tank; this is discussed further below.

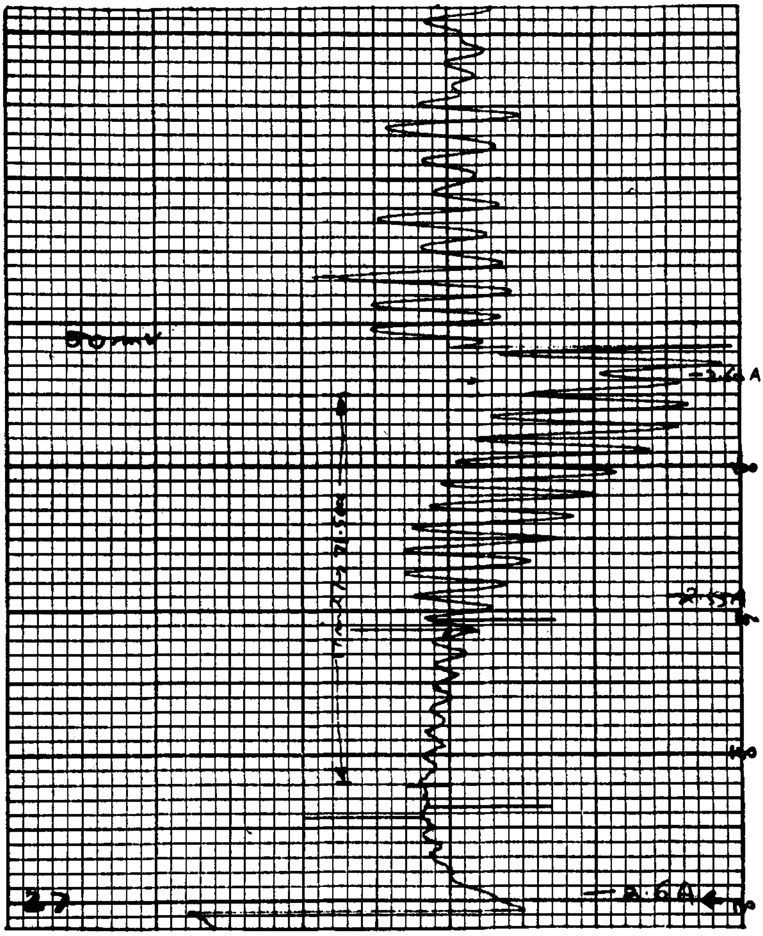

FIGURE 2. A section of the differential thermocouple record, run 4. The amplitude of the sudden change at the start is 1 μV, and the initial output is 27 μV. The sensitivity was reduced by a factor 5 at the mark '50 mV'. While the amplitude of oscillation was nearly constant at 0·2 μV the temperature gradient was falling at the rate of 0·2 % per minute.

It is worth noting that the growth in amplitude of the oscillations bears out the analysis of Veronis indirectly. His numerical integration of the equations for finite amplitude oscillations showed that convection always became monotonic after an initial oscillatory period, and that this steady convection was due to a subcritical instability which manifested itself only when the system was given a sufficiently large disturbance of appropriate form. Since the oscillations were initially very small, it follows that they always grew in amplitude until the finite-amplitude instability was initiated. Indeed the inevitability of this growth (when R and R_s are constant) is suggested by the form of the curve of equilibrium heat flow versus R for constant R_s. In the region of interest it appears that higher heat flow is associated with lower R; it was shown by Sani (1965) that, in a rather similar (monotonic) situation, such an equilibrium is unstable and there is a tendency for the motion to increase.

5. Solute transfer

Figure 2 shows an extract from the thermocouple record of run 4, in which an attempt was made to vary the heating rate in such a way as to prolong the period of small oscillations. The result was a period of nearly 10 min during which the amplitude of the oscillations was approximately constant. In order to maintain zero growth rate it was necessary to reduce the temperature gradient continuously, at a rate of 0·2 % per minute. Presumably, therefore, the difference in concentration between bottom and top of the convecting layer was being reduced at about the same rate. Diffusion alone would have changed the concentration contrast by only about 0·006 % per minute. Hence even this low amplitude oscillation was apparently transferring solute 30 times faster than pure diffusion would. The corresponding figure for the convective heat flow could not be determined but was apparently small. This result is consistent with the prediction of Veronis (1965) that the increase in solute transfer rate due to small-amplitude convection would be $(K/D)^2$ (i.e. about 10^4) times as great as that for heat, at least in the monotonic case.

6. Cells

Figure 3 (*a*)–(*e*) shows the tops of the cells formed at an early stage of a typical run (run 15). These pictures were taken with the optics arranged as a standard schlieren system, which would pass only that light which had penetrated the tank in a region where the refractive index was increasing downwards at more than a certain rate. Thus the image of the lowest part of the tank, where the density (and hence the refractive index) actually decreased downwards, was not illuminated. The cell tops were illuminated, and hence were regions in which the density increased downwards more rapidly than in surrounding liquid at the same level. This is consistent with the observation that solute was transferred more rapidly than heat by the convection; in terms of their respective effects on the density, the build-up of solute at the top of the cell exceeded that of heat, producing a relatively large (stable) density gradient just above the cell. This view was confirmed by limited observations which suggested that the temperature gradient in the cell was not greatly disturbed by the convection at this stage.

The sequence in figure 3 shows that the cells were independent and dynamic. They migrated sideways, and in addition were continually appearing and disappearing. The system only revealed those in which the solute flux was sufficiently high at a given time; so it is not known whether the whole layer was populated by cells of which only a few were sufficiently active at any time, or whether their apparent independence was genuine. The first alternative appears more reasonable hydrodynamically, though there is evidence that the second is in fact true. In either case the situation was clearly not the orderly array of rectangular contiguous similar cells assumed by the theories of this type of convection.

7. Stability diagram

To define the boundary of stability it was necessary to evaluate R and R_s for the convecting layer at marginal stability for each run. Unfortunately there was no direct measure of the depth of the layer, and it was therefore necessary to assume that this depth was equal to that at which $R_e(z)$ was a maximum. Thus it was necessary to know the profiles of both temperature and concentration throughout the layer.

The temperature at any level z was calculated from the measurement of the temperature at the bottom of the tank as a function of time. The liquid was initially entirely at temperature θ_0, and the temperature at time t at z was given by (Carslaw & Jaeger 1959)

$$\theta = \theta_0 + \frac{z}{2(\pi K)^{\frac{1}{2}}} \int_0^t T(\lambda) \frac{\exp \dfrac{z^2}{-4K(t-\lambda)}}{(t-\lambda)^{\frac{3}{2}}} \, d\lambda$$

where T is the temperature at $z = 0$. The integral was evaluated numerically from readings of T taken every 45 seconds during heating.

The concentration gradient profile supplied by the optical system was approximated by a Fourier series, which was then integrated to give an expression for the concentration profile. Allowance was made for the decay of harmonics between the time at which the gradient profile was measured and that at which marginal stability was reached, although this was usually very small.

$R_e(z)$ was calculated at the time of marginal stability at depth intervals of 0·1 cm in the region of interest. The value of z for which R_e was a maximum was then taken to represent the top of the convecting layer, and the required R and R_s were taken to be those calculated for that level. In all these calculations, the values adopted for physical constants of the solution were those available in tables, principally the International Critical Tables, due allowance being made for the solute content and temperature variation. The thermal expansion coefficient of sugar solutions was not available, and was therefore measured in a dilatometer for 5 and 10 % solutions between 0 °C and 35 °C; a double power series approximation was obtained by least-squares and used for calculation of the coefficient at other concentrations and temperatures.

The resulting stability diagram is shown in figure 4, from which it may be concluded that the theoretical linear boundary of stability is approximately confirmed. However, the experimental points show a marked tendency to occur at excessive values of R when R is large, and to approach the predicted relationship only for small R. The critical value of R_e, R_{ec}, is about 2,500; this is much greater than the predicted value of $27\pi^4/4 (= 657)$, but most of this difference is accounted for by the non-ideal nature of the boundary conditions, and more interest attaches to the failure of the points to follow the line of predicted slope.

The departure from the predicted stability relationship cannot be accounted for by experimental error. Uncertainty in the thermal expansion coefficient and its solutal analogue amounts to $\pm 1\%$ in R and R_s independently; and that in $(K\nu)$, which is the denominator in both R and R_s, amounts to $\pm 5\%$ in R and R_s jointly. A further uncertainty of 1–2 % in R and 1 % in R_s independently arises from measurement of temperature and concentration respectively. The greatest uncertainty, amounting to about $\pm 10\%$, arises from uncertainty in the depth of the convecting layer; but this affects both R and R_s in such a way that the locus of possible positions (R, R_s) is nearly parallel to the trend of the graph, and does not greatly modify the result. The only remaining possible source of error is the determination of the time of onset of overstability. Except in runs (notably run 1) where the heating rate was excessive, this uncertainty is negligible, particularly as this effect also acts almost along the trend of the graph.

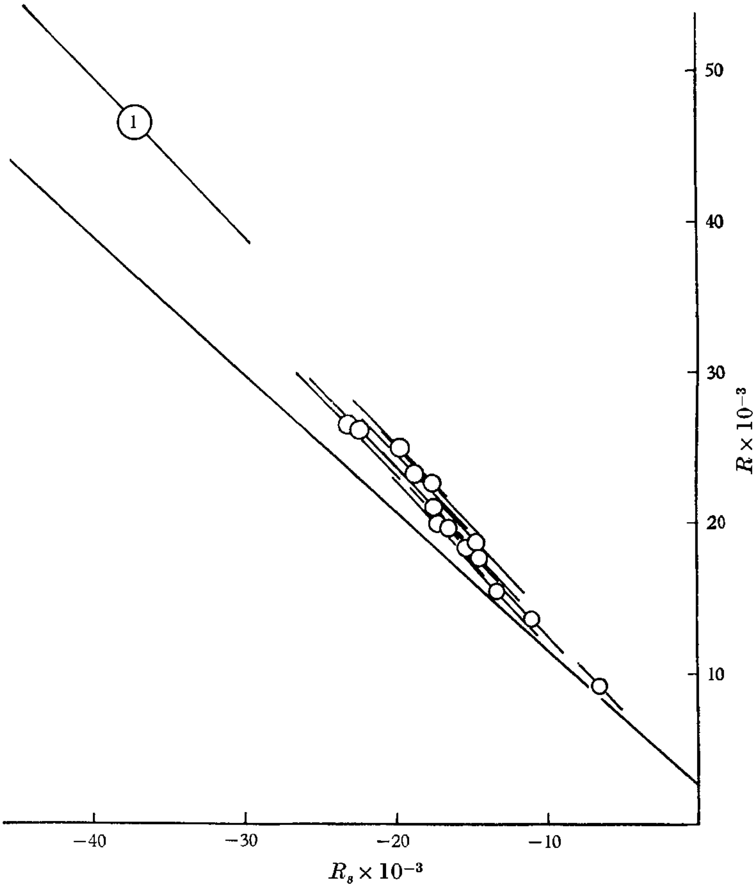

FIGURE 4. Stability diagram for overstability, assuming a linear R_s.

The predicted line has slope $P/(P+1)$. It is tempting to conclude from figure 4 that this slope should be modified, but in fact this would not produce agreement between theory and experiment. If a different value is assumed for this slope, for example, unity, this affects the calculations of convecting depth in such a way that the same disparity exists between the experimental points and the new theoretical line.

Thus it seems very probable that, although the theoretical linear stability criterion is approximately correct, it is not exact; and that the true criterion is represented by a curve rather than a line in the (R, R_s)-plane. This result accords with the conclusion of Nield (1967) for the case of monotonic instability. The curvature can result from the fact that, at the lower boundary, the temperature and the concentration were subjected to formally different conditions.

In view of the method used to calculate R and R_s, it is not possible to decide directly from figure 4 what form the non-linearity of the stability criterion takes. To do this satisfactorily it would be necessary to have an independent measure of the depth of the convecting layer. The only measure available is the record of observations of cell heights. These were not very accurate, and were measured sufficiently long after convection began to throw doubt on their relevance to the marginally stable state. However, the graph of R versus R_s, where these are the quantities calculated at marginal stability for the level later occupied by the tops of the observed cells, is displayed in figure 5, and shows reasonably systematic results. The boundary of stability is undoubtedly curved, and is represented quite well by the equation

$$R(1+2{\cdot}5\times 10^{-6}R)+0{\cdot}88R_s = R_{ec},$$

where R_{ec} is about 5,000.

With this relationship as a guide, a search was made for a similar one which would give better agreement with that derived from the experiments, where the assumed relationship was used, as previously, to determine the convecting depth. To ensure that R_e had a maximum in the region of interest, a cubic term had to be included. Figure 6 shows the stability diagram which resulted from assuming that its form would be

$$R(1+4\times 10^{-6}R-10^{-11}R^2)+0{\cdot}88R_s = R_{ec},$$

together with the graph of this expression for $R_{ec} = 5{,}000$. Agreement is still not complete, but the data do not justify a more intensive search for a better fit. One virtue of this choice is that it gives a good agreement in the main between calculated convecting depths and observed cell heights, as shown in table 1. Discrepancies are mainly associated with the fact that the maximum of R_e is now rather broad, so that the depth h is ill-defined. It should be noted that the significance of these particular expressions for the boundary of stability is probably limited to a demonstration that the boundary is curved, and that the curvature has a particular sense.

The results of run 1 do not conform with the general pattern. No source of error could be found to account for this, so it might be associated with the particularly high rate of heating used on this occasion, which led to a very markedly non-linear temerature profile. In the case of monotonic pure-thermal convection, Currie (1967) has predicted that very rapid heating may have a substantial effect on the critical Rayleigh number.

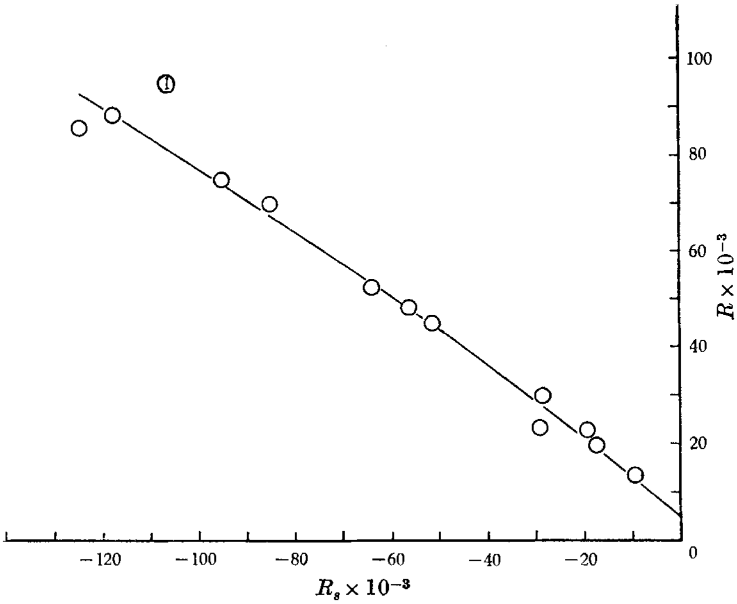

FIGURE 5. Stability diagram for overstability, using cell heights to estimate the convecting depths.

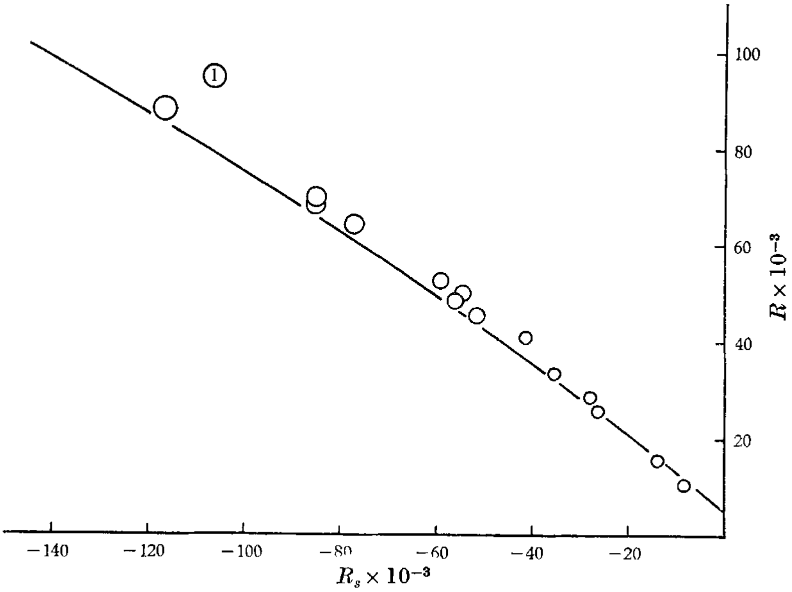

FIGURE 6. Stability diagram for overstability, assuming a non-linear (cubic) R_e.

Run	Calculated (cm)	Observed (cm)
1	1·4	1·4
2	1·2	1·4
3	1·2	1·2
4	1·3	1·3
5	1·0	0·8
6	1·0	0·7–1·0
7	1·0	0·7–1·0
8	1·1	0·8
9	1·1	0·9
10	1·0	0·9–1·1
11	0·8	0·9
12	0·8	0·5–1·0
13	0·8	1·0
14	0·9	0·9
15	0·8	0·8

TABLE 1. Comparison of observed cell heights with calculations of the layer depth using the non-linear (cubic) stability criterion

		Calculated period (sec)		
Run	Observed period (sec)	Linear R_e	Cubic R_e	Cell height
1	53 ± 5	56	49	49
2	85 ± 2	63	59	53
3	73 ± 2	58	50	50
4	69 ± 2	55	48	48
5	45 ± 2	39	35	39
6	48 ± 5	41	36	43–36
7	56 ± 2	41	37	44–37
8	55 ± 2	41	35	41
9	53 ± 1	43	37	40
10	32 ± 2	29	24	26–23
11	30 ± 5	31	29	27
12	47 ± 1	35	33	41–30
13	65 ± 5	59	59	49
14	53 ± 2	43	40	40
15	59 ± 4	45	45	45

TABLE 2. Periods of the overstable oscillations

Although the data do not provide an unequivocal value of R_{ec}, it is likely that the value is higher than expected. The overstable layer had been expected to behave as though its lower boundary was plane and rigid, and its upper boundary approximately plane and stress-free. By analogy with pure-thermal convection, R_{ec} would then have been about 1,200 (D. A. Nield, pers. comm.), or perhaps as high as 1,700 if the upper boundary was effectively rigid. A higher value implies that, in the thermosolutal case, more work has to be done against friction by the buoyancy force. The only possible cause for this seems to be that the cells which formed first were independent of one another, and not contiguous as suggested by studies of pure-thermal convection. In this case the buoyancy force in a cell would cause sympathetic motions in a relatively large volume of liquid in addition to the oscillation in the cell itself, against the frictional effect of viscosity.

8. Periods of the oscillations

The measured periods of the oscillations just after marginal stability are shown in table 2. In each case, the measurement was made over as many complete cycles as possible until the oscillation became irregular. This number varied from 2 to 20, producing a range of uncertainties in the periods.

The period expected for each run was calculated in accordance with the relationship given in §1. Since the period is a function of the convecting depth, three values were calculated for each run, corresponding to the three estimates of this depth; namely, by taking the maximum of the linear R_e, the maximum of the cubic R_e, or the observed cell depth. The values are included in table 2.

There is a strong tendency for the calculated periods to be less than the observed ones. This tendency is least obvious if the linear R_e is used to calculate the convecting depth, since it generally gives the smallest of the three depth estimates. The discrepancy is presumably associated with the difference between the actual form of the cells and that assumed in the theory. For a given solute distribution, the period is determined by the thermal diffusivity and the viscosity, solute diffusion being negligible. Since it increases with the viscosity of the liquid, the observation of excessive values may be ascribed to the same cause as the high R_{ec}, namely the high viscous damping of the motion when the cells are not in contact.

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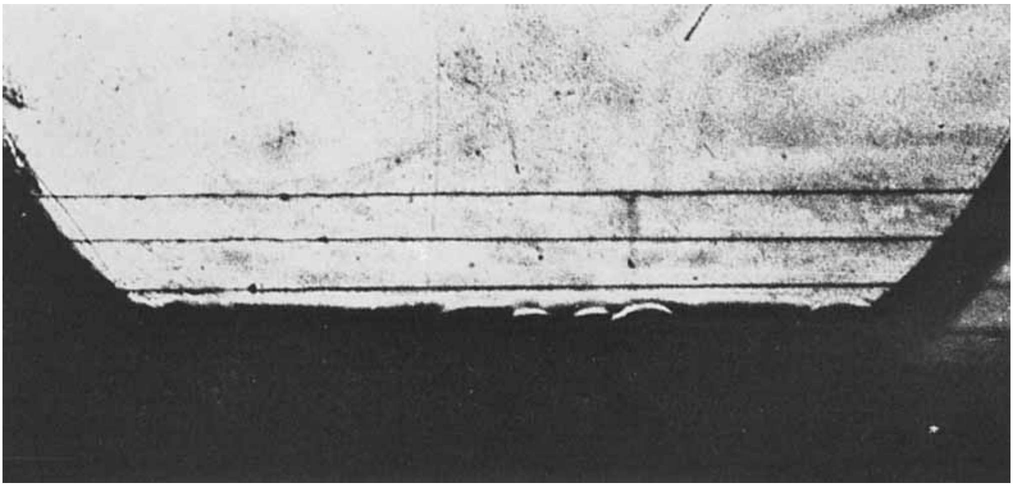


FIGURE 3(*a*)

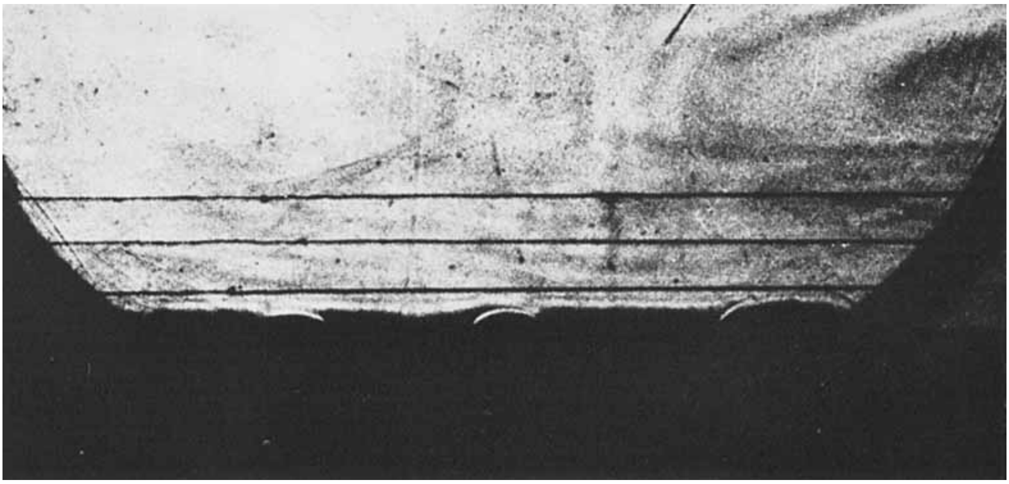
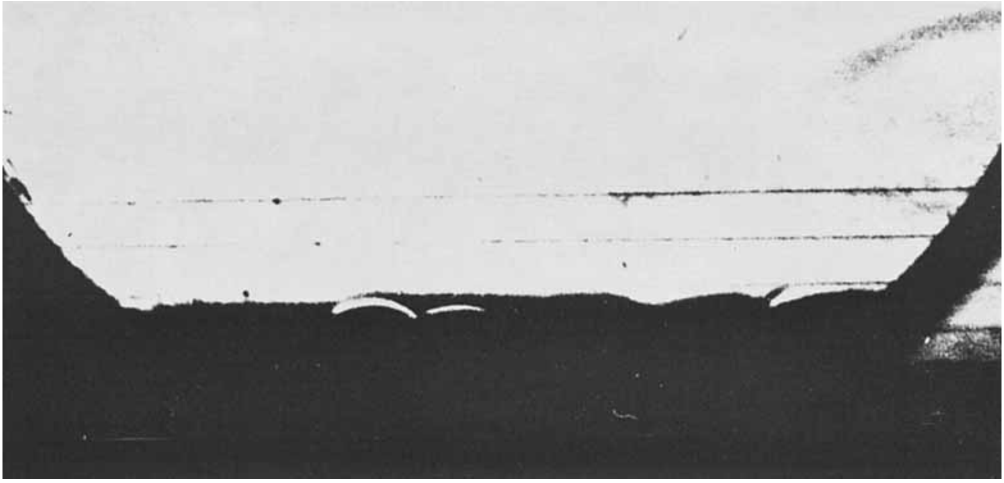


FIGURE 3(*b*)

FIGURE 3. Schlieren observation of cell tops, run 15. Only the lower part of the field of view is shown. The three thermocouple wires visible are 1·0, 1·5 and 2·0 cm above the tank bottom. These photographs were taken between 9 and 17 min after marginal stability.



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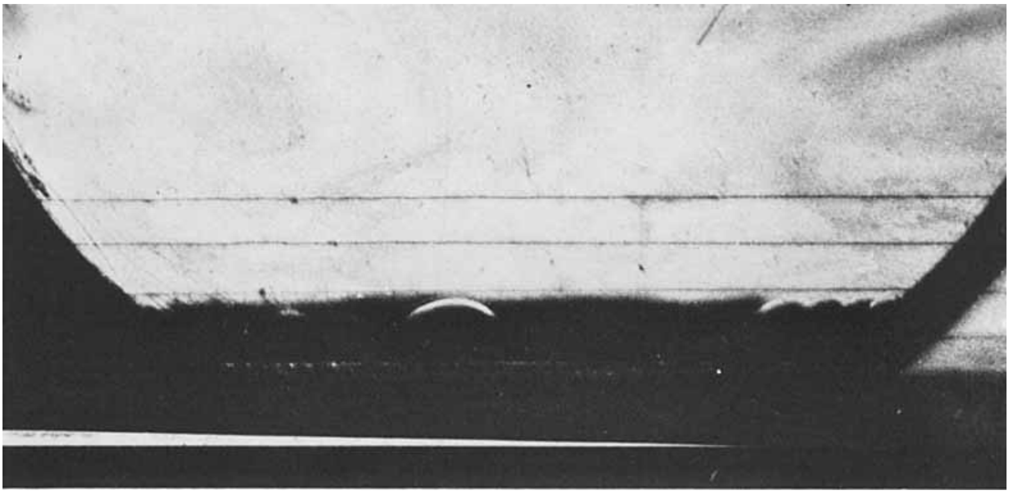

FIGURE 3(*d*)

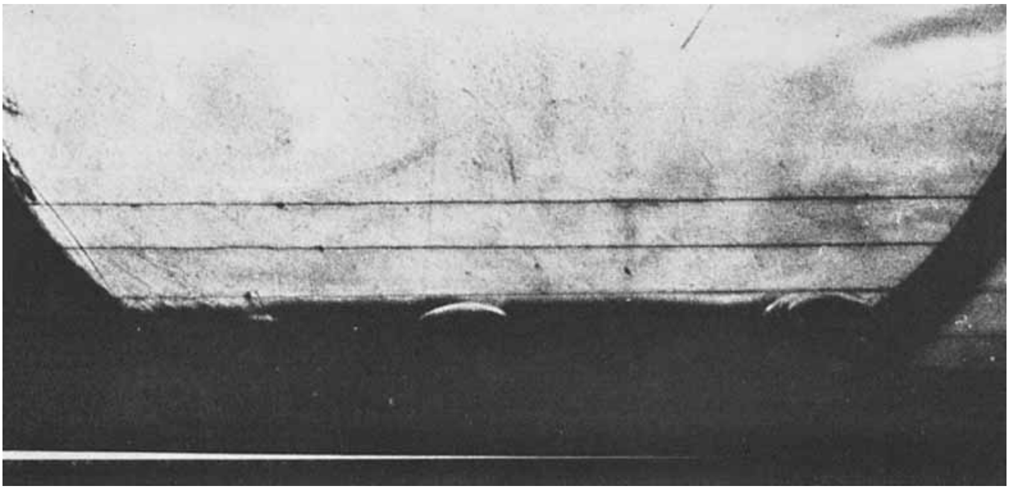

FIGURE 3(*e*)

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