

DOUBLE-DIFFUSIVE PHENOMENA

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INTRODUCTION

The study of convective motions produced by unstable density distributions in a fluid is now highly developed. Most attention has been given to the case of a thin horizontal layer of fluid, heated below and cooled above, and results obtained using a combination of Boussinesq theory and laboratory experiments have been successfully applied in many contexts (Chandrasekhar 1961, Spiegel 1971). The problem of buoyant convection from isolated sources has also been extensively studied (Turner 1969). Though the effect of adding other processes, such as rotation and magnetic fields, to the buoyant motion has been considered (Spiegel 1972), it has been assumed that the driving density differences are produced by the spatial variations of a single diffusing property (such as heat or a solute). Comparatively recently it has been shown that the simultaneous presence of two components with different diffusivities can lead to a whole range of new phenomena, and these form the subject of the present review.

A striking feature of many systems of interest is that instabilities can develop even when the net density decreases upwards. Diffusion, which is generally stabilizing in a fluid containing a single solute, can now act so as to allow the release of the potential energy in the component that is heavy at the top. Much of this work was initiated with an application to the ocean in mind, and because heat and salt (or some other dissolved substance) are then important, the process has been called *thermohaline* (or *thermosolutal*) convection. Related effects have now been observed in other contexts, to be described below, and the name *double-diffusive convection* has been used to encompass this wider range of phenomena. The minimum requirements for the occurrence of double-diffusive convection, in the sense implied here, are the following:

- (i) The fluid must contain two or more components having different molecular diffusivities. It is the *differential* diffusion that produces the density differences required to drive the motion.
- (ii) The components must make opposing contributions to the vertical density gradient.

(It is assumed throughout that the fluids are completely miscible, so that surface-tension effects do not arise. Some of the motions produced by the Marangoni

effect and buoyancy changes at boundaries between immiscible solvents probably have some features in common with the phenomena discussed here, but a detailed comparison is not attempted. For a review of the immiscible case, see Berg, Acrivos & Boudart 1966.)

The form of the motion depends on the relation between the diffusivities and the density gradients, i.e., on whether the driving energy is provided by the substance of higher or lower diffusivity. Physical intuition based on ordinary thermal convection experiments is of little direct help here, but two "thought experiments" do bring out the main features of the two possible kinds of motion. The first, due to Stommel, Arons & Blanchard (1956), provided the impetus for the development of the whole subject. They considered what would happen if a long narrow metal tube were inserted vertically through a region of the ocean where warm salty water overlies colder, fresher, and denser water (Figure 1a). Water pumped upwards (say) would reach thermal equilibrium with its surroundings at the same level, while remaining fresher and therefore lighter, because the walls of the tube present a barrier to salt but not to heat. The fluid in the tube will experience an upward buoyancy force, so motion in this "salt fountain" will still occur when the externally applied pressure difference is removed. The flow is drawing on the potential-energy supply in the salinity distribution and will continue so long as this is renewed by evaporation at the sea surface. A flow started downwards would also continue without further pumping.

In the opposite case, where the warmer salt water is underneath colder, fresher, lighter water, let us suppose that a parcel of fluid is isolated from its surroundings by a heat-conducting shell and displaced upwards (Figure 1b). It will lose heat but not salt, and buoyancy forces will drive it back towards its initial position, thus producing an oscillation. This can grow in amplitude (i.e., become "overstable"), provided the viscous damping is not too large, because the lag in temperature between the parcel and its surroundings leads to an asymmetry of the motion; the parcel experiences a net accelerating buoyancy force that causes it to return to its equilibrium position faster than it leaves. Both fountain and oscillator are in fact very easy to demonstrate in the laboratory (Martin 1971).

A decisive step was taken by Stern (1960), who pointed out that solid containing boundaries are not essential to the above arguments—it is not necessary

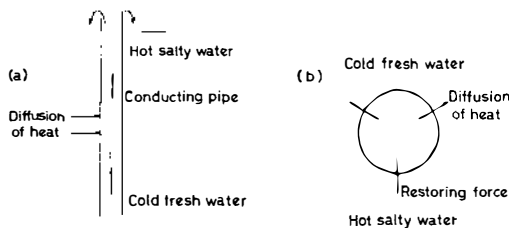


Figure 1 Sketch showing the two kinds of motion that can arise in a fluid with opposing vertical gradients of heat and salt: (a) salt fountain, (b) oscillating element.

to block the transfer of salt entirely. Similar motions can be set up in the interior of a fluid because a slower transfer of salt relative to heat is assured by its smaller molecular diffusivity. In the first case, long narrow convecting cells, called *salt fingers*, have been predicted and observed (see Figure 3), and these can lead to a very rapid vertical transport of salt. The existence of an oscillatory mode of instability has also been confirmed in the case of a stabilizing gradient of a solute heated from below.

STABILITY ANALYSES

Linear Theory

Much of the theoretical work in this field has developed directly from the linear stability calculations for a simple fluid heated from below. Stern (1960) was the first to consider the case of linear opposing gradients of two properties between horizontal boundaries at fixed concentrations, and since then many others, including Gershuni & Zhukhovitskii (1963), Veronis (1965, 1968), and Nield (1967) have developed the ideas. A good summary of this work has been given by Baines & Gill (1969). With the introduction of two "concentration" differences ΔT and ΔS between the boundaries (which need not be limited to temperature and salinity), and two different molecular diffusivities κ_T and κ_S , four nondimensional parameters are required to specify the system. In addition to the ordinary thermal Rayleigh number $Ra = g\alpha\Delta T d^3/\kappa_T \nu$ and the Prandtl number $Pr = \nu/\kappa_T$ (where α is the coefficient of expansion, d is the depth, ν is the kinematic viscosity), one can use the ratio $\tau = \kappa_S/\kappa_T$ (with $\kappa_T > \kappa_S$) and the density ratio $R_\rho = \beta\Delta S/\alpha\Delta T$, where β is the corresponding "coefficient of expansion" relating salinity to density variations. A frequently used combination is the salinity Rayleigh number

$$Rs = (\beta\Delta S/\alpha\Delta T)Ra = g\beta\Delta S d^3/\kappa_T \nu \quad (1)$$

The characteristic equation for the complex frequency $p = p_r + ip_i$ of small disturbances (obtained in the usual way using a normal-mode expansion $T \sim e^{pt} \sin \pi ax \cdot \sin \pi bz$, for example) is a cubic in p , with coefficients that depend on Ra , Rs , Pr , and τ . In a given fluid (fixed Pr and τ) the form of the solutions, and therefore the criterion for instability and the character of the unstable motion, change systematically with Ra and Rs . The various regions of the (Ra, Rs) plane are shown schematically in Figure 2, where the sign convention is such that negative Ra and positive Rs imply distributions of the respective components that are heavy at the bottom.

The stability boundaries, for the case of free horizontal boundaries above and below, are the straight lines whose equations are

$$XZ: Ra = Rs/\tau + 27\pi^4/4 \quad (2)$$

$$XW: Ra = [(Pr + \tau)/(Pr + 1)]Rs + (1 + \tau)[1 + (\tau/Pr)](27\pi^4/4) \quad (3)$$

When $Ra < 0$ and $Rs > 0$, both gradients are stabilizing and no growth is possible. When $Ra > 0$ and $Rs < 0$, all points in this quadrant above XZ are unstable;

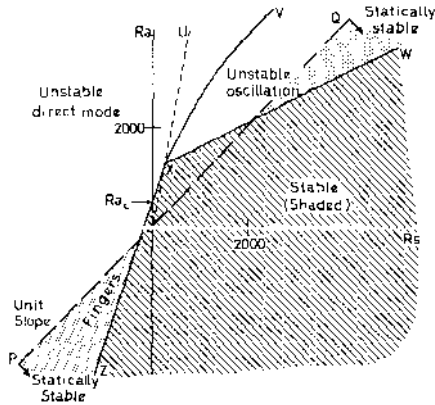


Figure 2 Diagram of the various convection regimes in the Ra, Rs plane, where positive Ra corresponds to a destabilizing gradient of the component with larger diffusivity κ_T and positive Rs to a stabilizing gradient of the second component (κ_S). The cross-hatched region is stable; in the stippled regions, small disturbances can grow, in spite of the fact that the net density distribution decreases upwards.

$Ra_e = Ra - Rs/\tau$ is an effective Rayleigh number that plays the same role as Ra in ordinary convection, and has the same critical value.

In the lower left quadrant ($Ra < 0, Rs < 0$, the “finger” regime), instability sets in at values of Rs only slightly larger, when τ is small, than the critical value for a salinity gradient alone, and certainly while the net density gradient is still statically stable. The motion is a direct, growing mode (i.e., the imaginary part of p is zero), and draws its energy from the potential energy in the component of lower diffusivity. Linear growth-rate calculations at supercritical $|Rs|$, with $|Ra| \rightarrow \infty, \tau \ll 1$, show that the fastest-growing cells are tall and thin, with a width scale of

$$l \sim \left(\frac{g\alpha\Delta T/d}{\kappa_T \nu} \right)^{-1/4} \tag{4}$$

Though such calculations are not strictly applicable to finite-amplitude motions, they do suggest that this will be the most efficient configuration for the release of energy. The result also agrees qualitatively with the observed form of salt fingers in their fully developed state.

When Ra and Rs are both positive (called the *diffusive regime*), instability occurs in an overstable or oscillatory mode, since $p_1 \neq 0$, just above XW . For $\tau \ll 1$ the critical thermal Rayleigh number is

$$Ra_c^0 = [Pr/(1 + Pr)]Rs + 27\pi^4/4 \tag{5}$$

At large values of Rs , instability occurs when the net density distribution is statically stable, since the slope of XW is less than that of PQ and these lines must cross at some Rs . The frequency of the most unstable mode is always less than

the buoyancy frequency $N = [(g/\rho)(d\rho/dz)]^{1/2}$, the factor being about 1/5 for heat and salt, and the cells are roughly as high as they are wide through the whole of the top of the diagram. Above the line XV the motion becomes direct, rather than oscillatory. As Ra is increased at fixed Rs , the two complex roots of p move towards the p_r axis, coalesce at parameter values corresponding to XV , and then separate along the p_r axis.

Linear calculations have also been made for a variety of boundary conditions by Nield (1967), and for an unbounded fluid by Walin (1964). These introduce no new qualitative effects though the numerical values of the critical parameters are changed. Hurle & Jakeman (1969, 1971) have shown that the Soret effect (i.e., mass diffusion caused by a temperature gradient) can produce a concentration gradient near a boundary that influences the form of the instability. Even in very dilute solutions the resulting Rs may be large enough to lie to the right of X on Figure 2, so that the breakdown is oscillatory rather than direct.

Finite-Amplitude Calculations

Veronis (1965, 1968) has extended the stability calculations for the case $Ra > 0$, $Rs > 0$ into the nonlinear regime, using numerical methods with a series expansion technique. When $\tau = 10^{-1/2}$ and $Pr = 1$ or larger, the onset of instability appears as an oscillatory mode at the $Ra = Ra_c^0$ predicted by linear theory. As Ra is further increased, a finite-amplitude transition to a steady mode occurs below the line XV (Figure 2). When Ra is sufficiently large, the strong convective motions that develop tend to mix the solute more rapidly than diffusion can make it nonuniform again, so the interior is nearly neutrally stratified. The mixed layer will thus transport nearly as much of the destabilizing T component as it does when this is present alone, whereas before this stage is reached the S gradient inhibits the transport of T . The fluxes of both T and S (F_T and F_S , say) are increased by convection, and Veronis has suggested that in the limit $Rs \rightarrow \infty$, $Rs/Ra \rightarrow 1$, we have

$$\beta F_S / \alpha F_T \rightarrow -\tau^{1/2} \quad (6)$$

This is related to an experimental result to be described later; but note that most of Veronis' calculations were made assuming an overall density distribution that increases upwards, unlike the experiments, in which the net density gradient is hydrostatically stable.

Sani (1965) has used a different perturbation method to investigate the stability of the same system to a particular form of disturbance, a finite-amplitude roll cell. Elder (1969) has considered the case of a step or sharp interface between two well-mixed layers, with higher T and S (and density) below. At first the interface thickens by diffusion, the T interface more rapidly than the S interface. The outer part becomes unstable and breaks away, and the interface is sharpened by the increased mixing at its edges. At this stage there is a balance between diffusion, which tends to spread the interface, and mixing on either side, which sharpens it, though later the calculations show a further disruption for reasons that are not clear. Lindberg (1971), using an upper-bound technique, has obtained results that suggest that convection will be completely inhibited at large values of R_p (though

this state has never been reached in laboratory experiments). More recent unpublished numerical calculations by D. O. Gough and J. Toomre have begun to incorporate some of the more complicated features of the experiments described below.

THE FORMATION OF LAYERS AND INTERFACES

Some of the above theoretical predictions have been verified in the laboratory, but there are still considerable gaps between theory and experiment. The attainable initial and boundary conditions are not the ideal ones assumed, and the physical models rapidly achieve large amplitude and even turbulent states that are not so amenable to detailed theoretical treatment. It is characteristic of double-diffusive convection that well-mixed layers form, separated by interfaces across which there are larger gradients of the several properties (Turner 1973). We now discuss the various ways in which such layers can be set up.

Superposition of Two Fluids

The most straightforward method is to pour a lighter layer of fluid with one combination of T and S carefully on top of another. In the diffusive case, for example with a layer of colder fresh water over hot salty water, the more rapid vertical transfer of heat relative to salt keeps the two layers well stirred and the

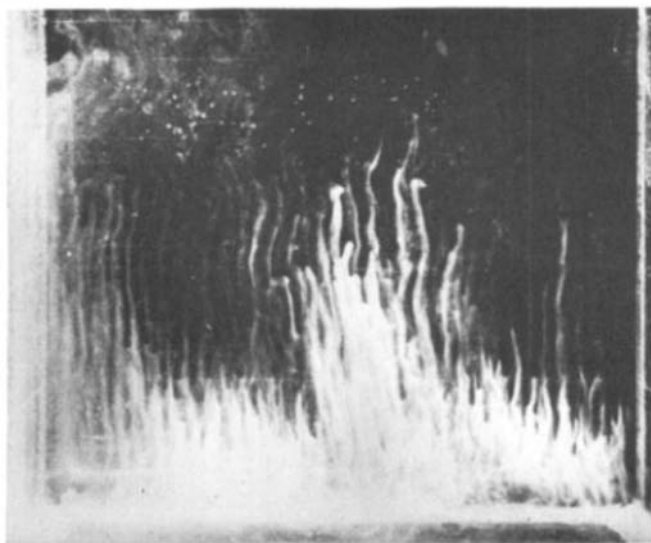


Figure 3 Vertical cross section of salt fingers, marked by fluorescein dye added to the upward moving fingers. (The experimental tank is 25 cm wide.)

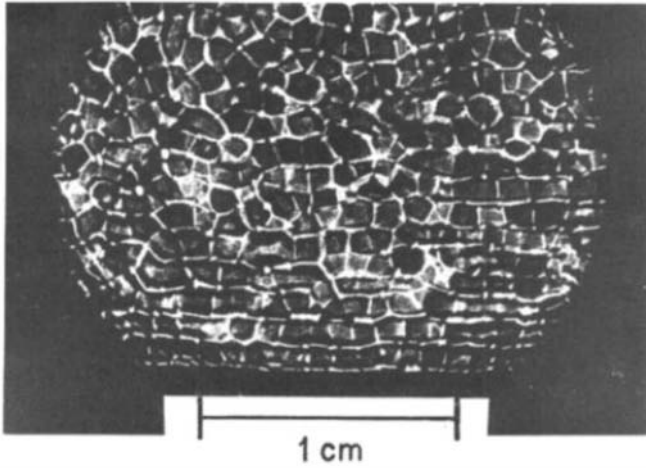


Figure 4 Plan view of sugar-salt fingers, obtained by a shadowgraph method. The cells tend to be square, but with gradually changing orientation, except where they are aligned by boundaries (as at bottom of picture).

interface sharp, while the density step is maintained by the excess salinity in the lower layer.

A sharp diffusive interface with strong convection on either side of it can also be formed using a layer of common salt solution (now the property T with larger diffusivity κ_T) over denser sucrose (S). Strong double-diffusive effects can thus be produced even though the diffusivities are much closer ($\tau = 1/3$ for salt and sugar compared with $\tau \approx 10^{-2}$ for heat and salt). This is a very convenient laboratory system, since it is useful to be able to set up such experiments without requiring any special precautions to prevent heat losses at the side walls.

In the finger case, when a little hot salty water is poured on top of cooler fresh water, salt fingers rapidly form. A vertical cross section of the long narrow convection cells, marked by fluorescein, is shown in Figure 3. When there are strong contrasts in properties, the fingers are confined to an interface, through which the density decreases upwards. This is bounded by sharp edges where the fingers break down and feed an unstable buoyancy flux into the convecting layers on either side. A plan view obtained by shining light vertically through such an interface is reproduced in Figure 4. This shows that upward and downward motions alternate in a close-packed array, which in the idealized case has a square planform (Shirtcliffe & Turner 1970).

It is also of interest to mention here the elegant color schlieren technique devised by Shirtcliffe (1972), which colors the image of a region according to the refractive-index gradient through it. This gives a much better qualitative picture of the motions at both diffusive and finger interfaces than does a simple shadowgraph.

Some definite quantitative results have been obtained regarding the initial stability of a finger interface and its dependence on τ . When a layer of S is placed on a layer of T , the sharp boundary is thickened by diffusion, at different rates for the two components. When Ra and Rs are large, as they usually are in practical cases, viscosity can be neglected and (2) shows that the criterion for instability is just $Rs > \tau Ra$. The appropriate Rayleigh numbers are based on the corresponding gradients; evaluating these in terms of the overall differences between the layers gives

$$\beta \Delta S / \alpha \Delta T > \tau \sqrt{\tau} \quad (7)$$

as the condition for the formation of fingers. This is easily achieved in the heat-salt case, but is a more serious limitation when the diffusivities are comparable. Huppert & Manins (1973) have verified this criterion experimentally using NaCl and sucrose and MgSO₄ and sucrose. They have also analyzed the quasi-steady finite-amplitude finger configuration (corresponding to Figure 4), and shown that it leads to the same condition (7) for the existence of fingers.

Step-by-Step Layer Formation

Layers can form when a smooth, statically stable gradient of one property has a vertical flux of a second imposed on it. The simplest example of a diffusive system of this kind (defined as above to be one in which the driving energy comes from the component having the larger diffusivity) is a stable salinity gradient, heated from below (Turner & Stommel 1964, Turner 1968). Initially a temperature boundary layer develops over a thickening region near the boundary; Shirtcliffe (1967, 1969) has shown that the first instability does occur as a growing oscillation, and that both the critical Rayleigh number and the frequency are in fair agreement with the predicted values. As heating is continued, a well-mixed layer develops. When the initial salinity gradient is linear, the layer depth d increases in time like

$$d = 2^{1/2} B^{1/2} N_s^{-1} t^{1/2} \quad (8)$$

where $B = -g\alpha F_T / \rho C$ is the buoyancy flux corresponding to the heat flux F_T

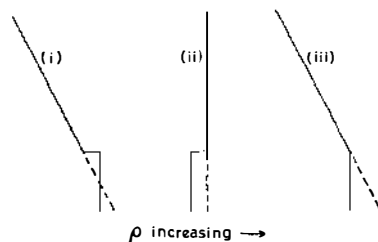


Figure 5 The observed distribution of density produced by heating a linear salinity gradient from below, showing the contributions due to (i) salinity, (ii) temperature, and (iii) the net density.

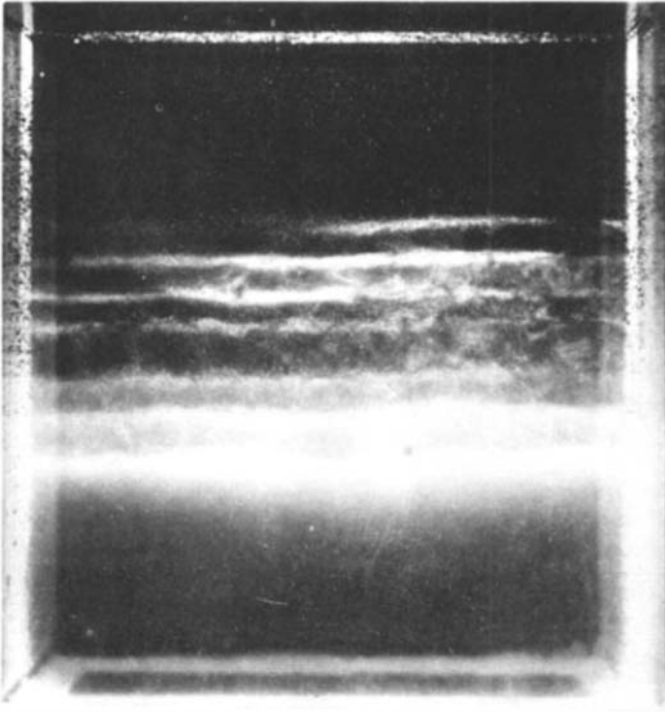


Figure 6 Photograph showing the layers formed in a laboratory tank of smoothly stratified salt solution, by heating from below. The layers are marked with fluorescein dye and aluminum powder, and the tank (which is 25 cm wide), is lit through a slit at the top.

(C being the specific heat) and $N_S^2 = -g\beta dS/dz$ is a measure of the initial salinity gradient. This relation has been shown by Turner (1968) to imply that the steps of T and S above the bottom layer are compensating, so there is no discontinuity of density, only of density gradient, as shown in Figure 5.

When the thermal boundary layer ahead of the convecting region reaches a critical Rayleigh number, it too becomes unstable. The first layer stops growing when d reaches the value

$$d_c = (\nu Ra_c / 4\kappa_T^2)^{1/4} B^{3/4} N_S^{-2} \quad (9)$$

and a second convecting layer is formed. Eventually many layers are built up, as shown in Figure 6, and the lowest layers merge successively while more appear at the top. Strong turbulent convection persists in each layer, driven by the heat flux across the interfaces, while much of the salt is left behind to maintain a mean density distribution that is lighter at the top.

It has been shown by Stern & Turner (1969) that layers are produced in the finger situation too, when a layer of lighter sugar solution (S) is placed over a gradient of common salt (T). This occurs because long fingers can become unstable to a collective instability having the form of an internal wave (Stern 1969). Fingers form, and then break down to produce a convecting layer which deepens, bounded below by an interface in which fingers persist. These in turn can become unstable, leading to a second convecting layer, and so on.

There is in fact a complete correspondence between the finger and diffusive systems, when they are considered on the scale of the convecting layers. The inequality of κ_S and κ_T results in an unstable buoyancy flux across statically stable interfaces in both cases, and this maintains the convection above and below. Only the mechanism of transport is different; across a finger interface the buoyancy flux is dominated by the destabilizing S component, so salt is transported faster than heat.

Side Boundaries

When a horizontal heat flux is imposed on a tank containing stratified salt solution, layers can form in another way. The sidewall thermal boundary layer grows by conduction and begins to rise. Salt is lifted to a level where the net density is close to that in the interior, and then fluid flows out away from the wall, producing a

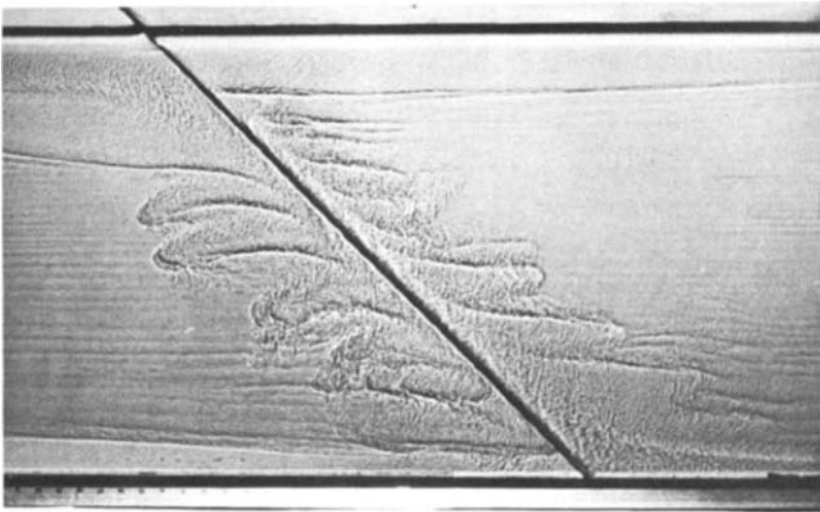


Figure 7 The growth of layers in a two-component system, following the introduction of a sloping boundary. The fluid was set up with linear gradients of NaCl (maximum concentration at the top and zero at the bottom) above sugar, and was stable when contained by vertical walls. The total depth of fluid was 21 cm.

series of layers that form simultaneously at all levels and grow inwards from the boundary. Thorpe, Hutt & Soulsby (1969), and Chen, Briggs & Wirtz (1971) have carried out laboratory experiments that demonstrate the effect beautifully. The latter have verified that when τ is small, the layer thickness is close to the lengthscale

$$l_1 = \frac{\alpha \Delta T}{\beta dS/dz} \quad (10)$$

which is the height to which a fluid element with temperature difference ΔT would rise in the initial density gradient. The first investigations of this phenomenon were reported by Mendenhall & Mason (1923) in another context, the separation of powdered minerals by differential settling in a fluid. This produced a density stratification that was not smooth, but occurred in the form of layers that they correctly associated with external heating. Their layers formed successively from the bottom up, so the horizontal boundaries also played a significant role.

The stability problem corresponding to sidewall heating of a wide container has not been solved, though Stern (1967) has shown theoretically how lateral gradients could be responsible for the generation of layers. Thorpe, Hutt & Soulsby (1969) have analyzed the simpler case of a narrow vertical slot, containing a fluid with compensating linear horizontal gradients of S and T . They have predicted the form and spacing of the cellular motions, which are steady and extend right across the gap, in fair agreement with their measurements using a narrow gap. Hart (1971), taking into account the mean flow due to the sidewall heating and more realistic boundary conditions, subsequently obtained better agreement with these same experiments. Wirtz, Briggs & Chen (1972) have produced a numerical model that exhibits the same features.

Analogous effects can be obtained using two solutes instead of salt and heat. In that case it is necessary to impose conditions at a wall that disturb the equilibrium in the interior of the fluid. If opposing vertical gradients of two properties are set up in a laboratory tank, the surfaces of constant concentration are normal to vertical boundaries, and so satisfy no-flux boundary conditions there. At an inclined boundary, however, diffusion will distort the surfaces of constant concentration and so produce density anomalies that tend to drive a flow along the wall. With a single component this flow can be steady and independent of distance along the slope (Phillips 1970, Wunsch 1970), but when there are two, the motions are much stronger and have some surprising features. Figure 7 shows an experiment started with linear gradients of salt overlying sugar. Counterflows develop near the inclined plate soon after it is inserted. Then fluid moves outwards to form layers, in which the vertical gradients are reversed locally and fingers form (though the original distributions were in the diffusive sense).

FLUXES ACROSS INTERFACES

Once layers and interfaces have formed in one of the ways described above, it is of interest to consider the coupled fluxes of T and S across them. Now we shall

discuss various laboratory measurements of F_T and F_S , and more detailed observations aimed at relating these to the structure of the interface. Again we emphasize that only interfaces between miscible fluids are being considered here.

Diffusive Interfaces

The overall behavior of a two-layer convecting system—with a hot salty layer below cold fresh water, for example—can be interpreted using an extension of well-known results for thermal convection. At high thermal Rayleigh numbers the Nusselt number (or nondimensional heat flux $Nu_T = F_T d / \kappa \Delta T$, where ΔT is the temperature difference between two layers of depth d) can be expressed as

$$Nu_T = f(Pr, \tau, R_\rho) Ra^{1/3} \quad (11)$$

This form is suggested by dimensional reasoning, and removes the explicit dependence on d . In a given fluid, with fixed Pr and τ , (11) is equivalent to $\alpha F_T = A_1 (\alpha \Delta T)^{4/3}$, where A_1 has the dimensions of velocity and is a function of the density ratio $R_\rho = \beta \Delta S / \alpha \Delta T$ only. The deviation of A_1 from the constant A obtained in experiments using solid boundaries (with a heat flux but no salt flux) is a measure of the effect of increasing ΔS on F_T .

Measurements using the heat-salt system (Turner 1965, Crapper 1973) have confirmed this form of relation. When R_ρ is less than about 2, the heat flux is increased above the solid-boundary value (that is, $A_1 > A$), due to the weaker constraints on both horizontal and wavelike motions. When $R_\rho > 2$ the heat flux falls progressively below the solid-boundary value as R_ρ is increased. Huppert (1971) has shown that the empirical form

$$A_1/A = 3.8(\beta \Delta S / \alpha \Delta T)^{-2} \quad (12)$$

fits the observations to the experimental accuracy over the whole of the measured range $1.3 < R_\rho < 7$.

Similar arguments can be used to express the salt flux as a function of R_ρ and R_ρ . It follows that the ratio of fluxes, expressed in density units, should be a function of R_ρ alone when Pr and τ are fixed, that is,

$$\beta F_S / \alpha F_T = f_*(\beta \Delta S / \alpha \Delta T) \quad (13)$$

The experimental results for heat and salt shown in Figure 8 support this conclusion. The most striking feature is the region of constant flux ratio $\beta F_S / \alpha F_T \approx 0.15$ for $R_\rho > 2$. Experiments by Shirtcliffe (1973), using a layer of salt solution above a sugar solution, have shown a much stronger dependence of F_T on R_ρ than (12) but again a constant flux ratio, the measured value for NaCl and sucrose being $\beta F_S / \alpha F_T \approx 0.60$.

There is as yet no completely satisfactory theory that reconciles these results with the purely diffusive transport through the center of the interface (which was also documented by Shirtcliffe). Some qualitative understanding has been obtained by extending the thermal-burst model of Howard (1964) to this situation. Boundary layers of both T and S are supposed to grow by diffusion to thicknesses proportional to $\kappa_T^{1/2}$ and $\kappa_S^{1/2}$, and then break away intermittently. If only the

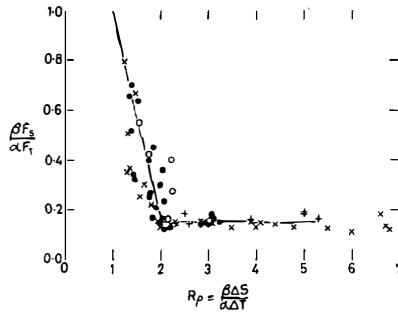


Figure 8 The measured ratio of salt to heat fluxes (in density units), across an interface between a hot salty layer below a cooler fresher layer, plotted as a function of the density ratio R_p (from Turner 1965).

statically unstable part at the edge is removed (such that $\alpha \Delta T = \beta \Delta S$), then the fluxes will be in the ratio $\tau^{1/2}$, in agreement with (6). Linden (1971b) has given a mechanistic argument to explain the increase of flux ratio at lower values of R_p , which he attributes to the direct entrainment of both properties across the interface, and Crapper (1973) has made detailed measurements of fluctuations near the interface, which support this picture. There is visual evidence in the sugar-salt experiments for a systematic interfacial wavemotoin, rather like the interfacial solitary wave described by Davis & Acrivos (1967), coupled to the large-scale convection in the layers.

Some measurements have also been made in the case where several solutes with different diffusivities κ_x are driven across an interface by heating from below (Turner, Shirtcliffe & Brewer 1970). The individual eddy-transport coefficients of the several solutes are different and approximately proportional to $\kappa_x^{1/2}$, but this result too awaits a detailed explanation.

Finger Interfaces

Because of the symmetry between the two systems remarked on in the previous section, many of the dimensional arguments developed for the diffusive case can be applied also to finger interfaces. Relations like (11) and (13) again hold, and both the salt flux and the ratio $\alpha F_T / \beta F_S$ are systematic functions of the density ratio $R_p^* = \alpha \Delta T / \beta \Delta S$ (it is convenient now to define it in this inverse sense, so that $R_p^* > 1$).

Turner (1967) has shown in the heat-salt case that the salt flux is about 50 times as large at $R_p^* \rightarrow 1$ as it would be if the same salinity difference were maintained at solid boundaries, and falls slowly with increasing R_p^* . He also obtained the value $\alpha F_T / \beta F_S = 0.56$ for heat-salt fingers over the range $2 < R_p^* < 10$. Linden (1973a) has since made direct observations of the structure of salt fingers and the velocity in them that support the earlier estimates of salt flux. He has suggested, however, that the steady-state flux ratio (though still a constant, independent of R_p^*) may

be much smaller, and that the conditions in Turner's experiments were not steady. Similar measurements using a layer of sugar solution over a salt solution (Stern & Turner 1969) give a flux ratio of 0.91 in the range $1.05 < R_\rho^* < 1.6$; fingers are clearly a very effective means of transporting *both* components in the vertical.

Visual observations of the kind described earlier suggest that there is a self-determined interface thickness, such that the fluxes carried by the fingers and by the larger-scale convection in the bounding layers are equal. In the course of time the fluxes between two layers are reduced, and the interface thickens; Stern & Turner (1969) showed how this behavior is consistent with the 4/3-power flux law for large-Rayleigh-number convection. Stern (1969) predicted the same kind of behavior using the collective-instability model referred to above. He showed that an established field of salt fingers ($\tau \ll 1$) will become unstable to long internal waves if

$$\beta F_S \approx \alpha \frac{dT}{dz} \geq \text{constant of order 1} \quad (14)$$

Thus if the temperature gradient is too weak (or the interface too thick) for a given salt flux, the fingers will be unstable over part of their length and the interface must thin till the critical state (14) is attained. When extra mechanical stirring is imposed on each side of a finger interface the fingers are disrupted, the equilibrium is upset, and there can actually be a decrease in salt flux (Linden 1971a). More recent work by Linden (1973b) has shown, however, that a *steady* shear does not have a dramatic effect on the fluxes, but just changes the fingers into two-dimensional sheets aligned down shear.

There is as yet no satisfactory quantitative theory of the dependence of F_S and F_T on the molecular properties, though Lambert & Demenkow (1972) have generalized Stern's (1969) collective-instability model to include $\tau \approx 1$. They also reported experiments that confirm the previous value of $\alpha F_T / \beta F_S$ in the sugar-salt case, but there is still some disagreement about the magnitude of the individual fluxes.

The Time-History of Several Convecting Layers

Let us suppose first that two layers, say of hot salty water below a cold fresh lighter layer, are confined between impermeable boundaries, so that their properties change only because of transfers across the interface. The density difference between them must clearly increase in time, since for energetic reasons the flux ratio [f_* in equation (13)] must always be less than unity. When f_* is small, the density difference increases most rapidly, because $\beta \Delta S$ remains little changed while the destabilizing $\alpha \Delta T$ is decreasing. The convective transports of both T and S fall to zero as $\Delta T \rightarrow 0$, when the *driving* component is nearly uniformly distributed in the vertical, but in this final state $\Delta \rho / \rho = \beta \Delta S$ is nonzero and stable.

The behavior of a three-layer system has been investigated by Huppert (1971). He showed that a finite layer with intermediate properties between two infinite layers in which S and T are fixed will be stable when f_* is constant (e.g., when $R_\rho > 2$ in the heat-salt experiments, see Figure 8) and unstable otherwise. When f_*

is constant, there is a range of combinations of properties of the middle layer such that the fluxes of both T and S across the two interfaces will be equal, and layers started with other values will tend to some point on the equilibrium curve. When f_* is not constant no stable equilibrium is possible, and the central layer will in time approach the density of one of the others and merge with it. The results can immediately be generalized to apply to any number of intermediate interfaces and layers.

No quantitative tests of this theory have yet been made, though Lindberg & Haberstroh (1972) have set up a multilayer version of Turner's (1965) experiment that suggests that a steady state can be attained only at large R_ρ . Turner & Chen (1973), using the salt-over-sugar technique to produce many convecting layers, have observed the successive merging of neighboring layers at values of R_ρ close to unity. The same principles will apply to a series of layers separated by finger interfaces, but in that case there are very few quantitative results on which to base definite predictions. In particular there have been no measurements of the value of R_ρ^* below which the flux ratio $\alpha F_T/\beta F_S$ begins to increase.

APPLICATIONS

The above outline of the principles underlying double-diffusive phenomena has been given using simple laboratory examples to illustrate the fundamental concepts. Applications of the ideas have been made in diverse fields, ranging from oceanography and astrophysics to various engineering contexts, and some of these will now be sketched briefly as an indication of the many ways in which these processes can become significant.

The Ocean

The term *thermohaline circulation* is used in oceanography to describe large-scale motions driven by density anomalies (e.g., Stommel 1965). This is of course not the kind of motion of concern to us here, nor are convective motions driven by evaporation or freezing (Foster 1969), which do not essentially involve the *coupled* fluxes of the two properties.

Layering that can confidently be associated with both kinds of double-diffusive processes has now been observed in the ocean by using modern continuous sampling techniques. The regularity of the steps and the systematic association of T and S serve to distinguish these from layers produced in other ways (by internal wave breaking, for instance). Neal, Neshyba & Denner (1969, 1971) have found layers about 5 m thick, in a situation where colder fresher water overlies warm salty water underneath a drifting ice island in the Arctic ocean. Other striking examples are the layers of hot salty water, bounded by diffusive interfaces, found at the bottom of various Deeps in the Red Sea (Degens & Ross 1969). These are nearly saturated with salts of geothermal origin, including a high proportion of heavy metals, and are of special interest because of the potential commercial value of the associated thick sediments (Backer & Schoell 1973).

Good examples of layering consistent with the finger process have also been

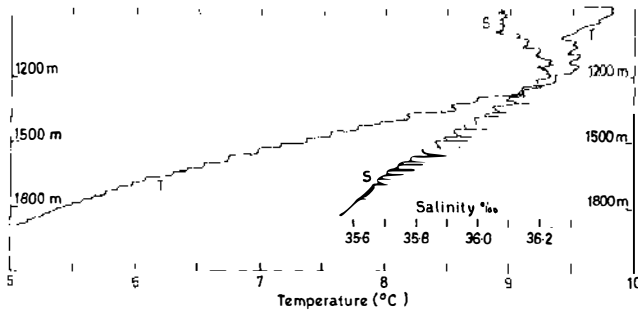


Figure 9 A series of layers in T and S observed under the Mediterranean outflow into the Atlantic, and attributed to the finger mechanism. The spikes on the right of the salinity trace are instrumental and can be ignored. (From Tait & Howe 1971.)

found by many oceanographers. Figure 9 shows a series of layers about 20 m deep recorded by Tait & Howe (1968, 1971) in the Atlantic underneath the Mediterranean outflow, which is an intruding tongue of warm salty water. (Layers stratified in the opposite sense have also been found above it.) Other cases have been reported by Cooper & Stommel (1968), Wunsch (1970), and Zenk (1970). Microstructure measurements extending into the millimeter range have been made by Gregg & Cox (1972), which contain examples of both mechanical and double-diffusive mixing processes. Although their reports have not yet been published, Magnell (1972), using a conductivity probe, and Williams (1972), using an optical device, seem to have found direct evidence for salt fingers in the ocean, considerably increasing our confidence in this explanation of the layering.

Conditions are particularly favorable for double-diffusive convection when a layer with compensating T and S differences intrudes at its own density level into an environment with different properties (as the Mediterranean outflow does), since relations like (12) show that the fluxes then have their maximum values. Fischer (1971) has suggested that this can be important in the context of sewage disposal in the sea, where a rising buoyant plume of relatively cold fresh water spreads out in the thermocline below warmer saltier water. The salt-finger mechanism can cause this layer to thicken vertically and even spread to the surface.

It is clear from the observations of Stommel & Fedorov (1967) and Pingree (1971) that horizontal gradients and advection will often play a significant part in the formation of layers, and the same must be true of many of the measurements reported above. Though Fedorov (1970) has attempted to use one-dimensional laboratory experiments to explain some of these observations, it seems premature to do so before the effects of horizontal gradients are more thoroughly understood (Huppert & Turner 1972). Turner & Chen (1973) have begun a laboratory study of such two-dimensional effects, and of the formation and horizontal propagation of layers in a region with opposing vertical gradients of two properties.

Lakes

There are many fresh-water lakes in various parts of the world that have become stratified at some time in the past by the intrusion of sea water. Some of these are observed to be hotter at the bottom than the top, and convectively mixed layers separated by diffusive interfaces are formed. A well-documented example is Lake Vanda in the Antarctic (Hoare 1966, 1968; Shirtcliffe & Calhaem 1968). Since these lakes are not complicated by advection processes, Huppert & Turner (1972) have been able to use the Lake Vanda data to confirm that the one-dimensional laboratory results (11) and (12) can be transferred quantitatively to comparable large-scale motions.

Artificially stratified lakes or solar ponds also rely on opposing density gradients for their operation (Tabor 1966, Weinberger 1964). Radiant heat can be stored in a salty layer that is protected from the air above by fresh water, and the efficiency is determined by the rates of transport of heat and salt due to double-diffusive convection.

The Atmosphere

Because the diffusivities of heat and water vapor in air are so close (the latter being about 20% greater), it is harder to attribute any observations in the atmosphere unequivocally to double-diffusive processes. In situations where the two contributions to the density gradient are opposing, however, it would be worth bearing the possibility in mind (remembering the laboratory results with two solutes of comparable diffusivity). For instance, the interface between a cold, moist layer of air underlying warmer drier air (at a cold front) is often observed to be very sharp. Since moist air is lighter than dry at the same temperature, there is potential energy in the moisture distribution, and double-diffusive convection could help to maintain the sharpness of the interface.

Radar observations of ascending moist parcels of air by Konrad (1970) have shown that the tops are much more clearly defined when the parcels are cold than when they are warmer than the environment, and this too is in the right sense for a double-diffusive explanation to be feasible.

Stars

Spiegel (1969, 1972) has shown that gradients of chemical composition can produce a density gradient in stars that plays the role of the salinity gradient in the laboratory experiments. Some large stars have a helium-rich core, which is heated from below and therefore convecting. Outside the core, lighter hydrogen predominates, but helium is transported up by double-diffusive convection. The name *semiconvection* has been given to the process whereby the motion outside the core can regulate itself to remain near a condition of convective neutrality. This must clearly be a state of uniform potential temperature (i.e., uniformity of the *driving* component) as suggested by Schwarzschild & Härm (1958), not one of uniform potential density.

It has also been suggested (see Goldreich & Schubert 1967, James & Kahn

1971, Spiegel 1972) that in differentially rotating stars, small-scale instabilities comparable to salt fingering may occur, the two properties with different diffusivities being matter and angular momentum. Such effects are, however, likely to be overshadowed by larger-scale baroclinic instabilities.

Liquid Metals

As metals solidify, undesirable inhomogeneities on the microscopic scale can be produced by several mechanisms, among which is double-diffusive convection in both the diffusive and finger senses. The oscillatory behavior induced by the Soret effect has already been mentioned, and Jakeman & Hurler (1972) have recently given a comprehensive review of double-diffusive and other ways in which thermal oscillations can arise in liquid metals. These can produce fluctuations in growth rate of crystals, which result in a banded distribution of solute or impurity concentration.

Another form of impurity distribution arising in some alloys solidified from below is called *freckling*. This consists of long narrow segregated regions that in horizontal cross section appear as small randomly distributed spots. Copley et al (1970) have shown that they arise when the heavier components solidify first, so that the concentration gradient ahead of the advancing front is unstable, opposing the stabilizing temperature gradient. The motions that result are complicated by the presence of a "mushy" zone of crystals at the solidifying front, but there are close analogies with the salt-finger mechanism, and freckle formation is favored by high thermal diffusivity and low solute diffusivity.

Chemical Effects

The detailed experimental proof that the Soret effect can lead to overstable oscillations was given by Hurler & Jakeman (1971), using water-methanol mixtures heated from below. Oscillations probably due to the same cause were reported by Caldwell (1970) in the course of measuring the thermal expansion of sea water using the onset of convection. Earlier, Agar & Turner (1960), in experiments designed to measure Soret coefficients, had reported direct convective instability due to increased concentrations at the top boundary of their cells.

Related problems are currently receiving attention in the chemical physics literature. For example, Bdzil & Frisch (1971) have studied the effect on stability of temperature and concentration gradients created by surface catalyzed reactions, and Velarde & Schechter (1971) have made a critical survey of Soret coefficient measurements. Much of the work in this area that has appeared recently (in the same journals as the last two papers cited) seems to be rediscovering, with minor variations and surprisingly little cross reference, many of the results already developed elsewhere.

Storage of Liquid Gas

Another phenomenon that depends on the presence of several components with different diffusivities and opposing contributions to the vertical density gradient is that of *rollover* in liquified natural gas tanks (see Sarsten 1972, and the two

papers that follow this). If a storage tank is refilled from below with heavier but warmer liquid natural gas than that already present, two layers separated by a diffusive interface can result. The pressure and the rate at which gas is withdrawn are adjusted to the conditions in the top layer. As the lighter fractions boil off, the upper fluid increases in density more rapidly than the lower layer can by cooling through the interface. When the densities become equal, rollover occurs; warmer liquid natural gas is suddenly convected to the surface and there is an uncontrollable increase in the rate of release of gas.

ACKNOWLEDGMENTS

The author is grateful for the support of a grant from the British Admiralty. H. E. Huppert, P. F. Linden, and P. C. Manins made helpful comments on an earlier draft of the manuscript.

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