MULTICOMPONENT CONVECTION

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1. INTRODUCTION

In two early reviews of this subject (Turner 1973, 1974), the phenomena described were adequately covered by the name *double-diffusive convection*. They were treated as generalizations of the process of thermal convection in a thin fluid layer, which arise when spatial variations of a second component with a different molecular diffusivity are added to the thermal gradients; the term *thermohaline convection* was introduced to describe the heat-salt system. The experimental and theoretical results then available could be described in terms of the differential diffusion of two components acting so as to release potential energy from a component that is "heavy at the top," even though the net density decreases upward. It was also shown that the form of the resulting motions depends on whether the driving energy comes from the component having the higher or lower diffusivity. Where one layer of fluid is placed above another (denser) layer having different diffusive properties, two basic types of convective instabilities arise, in the "diffusive" and "finger" configurations. These are illustrated in Figures 1 and 2, respectively. In both cases, the double-diffusive fluxes can be much larger than the vertical transport in a single-component fluid because of the coupling between diffusive and convective processes.

   The main development of these ideas since 1974 has occurred in the field of oceanography, with interactions between theoreticians, laboratory experimenters, and sea-going oceanographers playing a vital role. The field has also broadened considerably, with new applications becoming apparent in addition to those outlined by Turner (1974). A rather personal historical review of the subject was presented by Huppert & Turner (1981a), in which they emphasized the importance of transfer of information from

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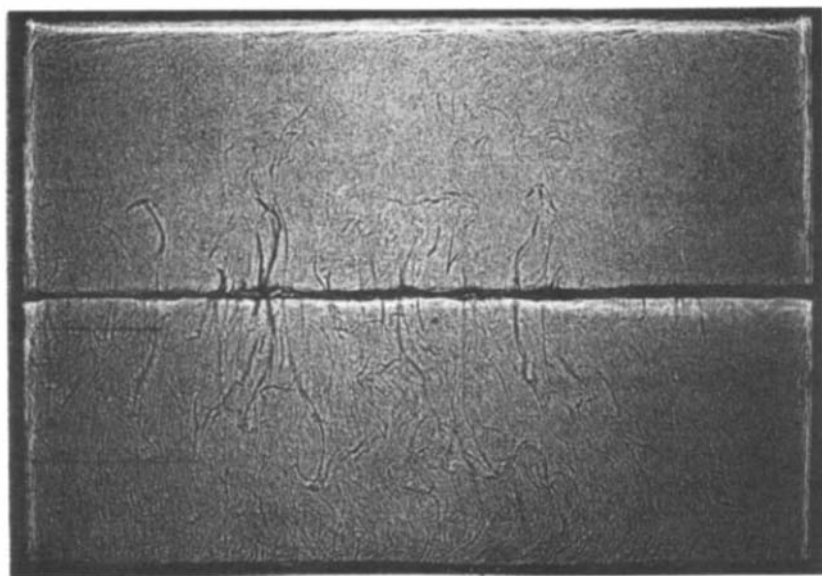
Figure 1   Shadowgraph picture of a "diffusive" interface formed by pouring a layer of NaCl solution ($\kappa = 1.5 \times 10^{-5}$ cm^2 s^{-1}) on top of a denser sucrose solution ($\kappa = 0.5 \times 10^{-5}$ cm^2 s^{-1}) in a tank 150 mm wide. Note the convective plumes on each side of the thin interface, evidence of strong interfacial transports.

one field to another. They concluded, however, that it is difficult to foster formal interactions of this kind, and that there must be a large element of chance in the spread of ideas across disciplines. In response to that review, an interdisciplinary conference was organized in March 1983 under the auspices of the Engineering Foundation, and summaries of the proceedings have been published (Huppert 1983, Chen & Johnson 1984). The present review

variety of topics to which these ideas can be applied.

It has become clear that not only can a wider range of phenomena be described in related terms, but that the previous physical descriptions, nomenclature, and even the basic definitions were unduly restrictive. There are situations (discussed below) where more than two components are significant—for example, in a star, the angular momentum, heat, magnetic field, and composition all diffuse at different rates, and they may vary spatially in dynamically important ways. Force fields other than gravity can be relevant, and there has been an increasing emphasis on two-dimensional effects due to a combination of horizontal and vertical property gradients. As disciplines that have developed relatively in-dependently have begun to communicate with one another, differences of
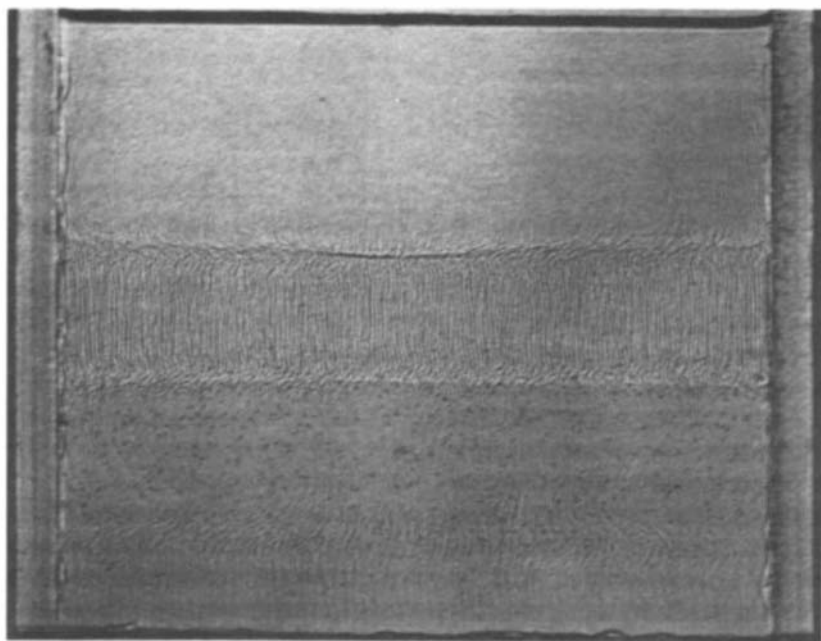
Figure 2 Shadowgraph of a thickened "finger" interface formed by placing a layer of sucrose solution on top of a denser NaCl solution and leaving it for about three hours (cf. Figure 1).

language and ways of thinking have become more apparent. A major aim of the present review is to resolve some of these differences by setting out the basic principles in a way that will be accessible not only to those whose primary interest is fluid mechanics but also to specialists in other fields.

The historical development of the subject is not pursued here (see Turner 1974). The more recent results and publications are given more attention, though there is of course some repetition of material appearing in earlier reviews. Where possible, reference is made to recent specialized reviews of particular topics. Our starting point is a broader definition of the circumstances under which multicomponent convection can occur in a fluid subject to a gravitational field:

1. The fluid must contain at least two components with different molecular diffusivities, each of which affects the density of the fluid. Chemists prefer to use the terms "isothermal ternary system," referring to a solvent and two solutes or polymers, or "nonisothermal binary system," thus distinguishing between thermal and other diffusion processes.[1]

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[1] Note that our use of the word "component" is also at variance with its technical significance in physical chemistry.

2. Differential or coupled diffusion can produce convective motions that are associated with a decrease in the gravitational potential energy of the system.

This second criterion represents an important generalization of the earlier ideas, as summarized in the first paragraph of this review. No longer is it assumed that one of the components must be distributed in a "hydrostatically unstable" manner; theoretical arguments suggest, for example, that when cross-diffusion terms are large, perturbations can grow in a ternary system even when both solutes are more concentrated at the bottom. Though the gravitational potential energy term is small compared to the free-energy change, it plays an essential role in the organization of the flow; this point is discussed in more detail in Section 3.2.

Another point, about which there is much confusion in the chemical diffusion literature, is worth making immediately. It is *not* necessary for the net vertical density gradient (across an interface, for example) to change sign and become statically unstable before convection can occur. Double-diffusive motions (and their equivalents in more complicated systems) can certainly develop under much less stringent conditions, which allow local density anomalies to develop while the horizontally averaged vertical density gradient ρ_z remains negative (i.e. the mean density decreases upward across an interface, but local anomalies cause local instabilities).

2. VERTICAL GRADIENTS OF DIFFUSING COMPONENTS

2.1 *Prototype Laboratory Experiments*

The basic phenomena and underlying physical principles are still most easily introduced by describing several one-dimensional, two-component experiments in qualitative terms. Some recent one-dimensional results are mentioned immediately, but more subtle extensions of these experiments, and the associated theory, are treated separately.

SUPERIMPOSED FLUID LAYERS   A characteristic of multicomponent convection is that well-mixed layers tend to form, separated by interfaces across which there are large gradients of the several components. A single interface can be set up directly by pouring a layer of one solution carefully on top of a denser miscible layer, which either is at a different temperature or contains a different solute. For consistency with the notation used below, we denote by T (standing for temperature in the extensively studied heat-salt system) the component with the larger molecular diffusivity D_{11}, and by S the component ("salinity") with the smaller diffusivity D_{22}. The ratio D_{22}/D_{11} is defined as τ (< 1), and cross-diffusion terms are ignored for the present.

A "diffusive" interface results when the component T is heavy at the top and S is heavy at the bottom, with the lower layer being denser (see Figure 1). When T refers to temperature, this means that cold, dilute solution lies above hot, concentrated solution. As the name suggests, the vertical transports of T and S across the hydrostatically stable central core of the interface occur in this case entirely by molecular diffusion. But because $D_{11} > D_{22}$, the *edges* of the interface can become marginally unstable (cf. Linden & Shirtcliffe 1978). The resulting unstable buoyancy flux into the layers above and below drives large-scale convection that keeps the two layers well stirred and the interface sharpened.

In this case the downward flux of T [expressed in density terms as αF_T through the relation $d\rho = \rho_0(\alpha\, dT + \beta\, dS)$] is greater than the upward flux of $S(\beta F_S)$ (or in the heat-salt case, the upward flux of heat is greater than the flux of salt). It is appropriate to focus attention on the gravitational potential energy in the T field, which is continually decreasing, as is the potential energy of the system as a whole. The density difference between the two layers *increases* in time (in contrast to the slower evolution due to diffusion across an interface in a single solute system or that caused by mechanical mixing between two layers).

Over a wide range of conditions, $\beta F_S/\alpha F_T$ is observed to be nearly constant and close to $\tau^{1/2}$ for both two-solute and heat-solute systems. The reader is referred to earlier reviews for a fuller discussion of this point, and of the quantitative flux measurements in general. The experiments are described in Turner (1965) and Marmorino & Caldwell (1976).

In the opposite configuration, with a layer of S above a layer of T, the structure shown in Figure 2 is observed. Small disturbances can grow rapidly, and long, narrow, vertical convection cells called "salt fingers" are formed and extend through the interface. It is now the more rapid *horizontal* diffusion of T relative to S over the width of the fingers that makes possible the release of the potential energy stored in the S field. It can be shown [see Equation (2) below] that when τ is small (e.g. for heat-salt fingers), even a tiny fraction of salt in the warmer top layer will lead to the formation of persistent fingers. The finger mechanism of transport, in which gravity plays a vital role, is described in terms of this familiar system.

Each downward-moving finger is surrounded by upward-moving fingers, and vice versa (Shirtcliffe & Turner 1970). The downgoing fingers continually lose heat (by horizontal conduction) to the neighboring upgoing fingers; therefore the downgoing fingers become more dense and the upgoing fingers less dense. There is a slower transfer of salt sideways, which results in a small vertical salinity gradient (Figure 3). Thus the small-scale (finger) convective motions are driven by the *local* density anomalies between fingers and lead to $\beta F_S > \alpha F_T$. However, the horizontally
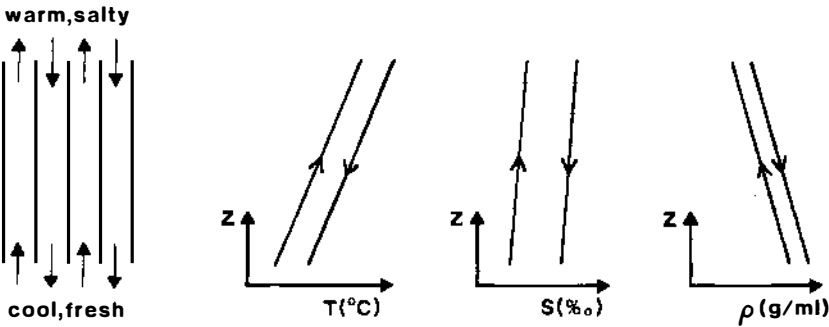
Figure 3 Upgoing and downgoing salt fingers and their temperatures, salinities, and densities. Arrows denote the direction of motion. By this mechanism, both properties are transferred down their mean vertical gradients, but much more rapidly than by simple molecular diffusion.

averaged vertical density gradient through the interface remains stable and dominated by the T gradient. There is an unstable boundary layer at the *edge* of the salt-finger interface (cf. Figure 2) that drives convection in the layers. The potential energy of the whole system is again decreasing and the density difference between the layers is increasing, but now, since $\beta F_S > \alpha F_T$, this energy can be regarded as derived from the salt field.

The schematic distributions of S and T, and the net density at the beginning and end of a two-layer "finger" experiment, are shown in Figure 4. In the initial state (state 1), S and T are homogeneous in the upper and lower layers, respectively, and ρ is the same everywhere. Rapid convective transports due to fingers produce state 2, in which S is nearly uniformly
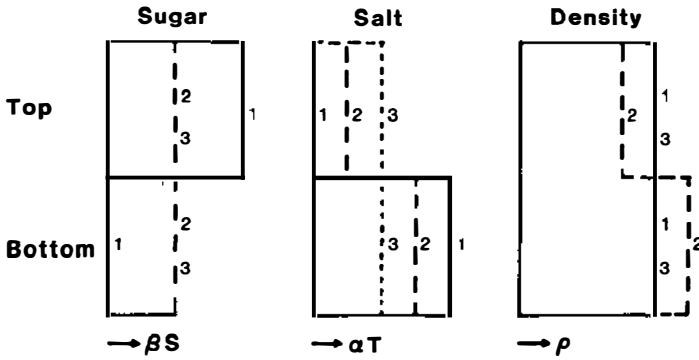

Figure 4 Distributions of S, T, and density at various stages of the "run-down" finger experiment described in the text. It has been assumed that $\alpha F_T/\beta F_S = 1/2$ for the purposes of illustration.

distributed with depth, while part of the T difference remains and there is a stable density step. The final state (state 3) of complete uniformity of both components would be achieved (by molecular diffusion) only after a much longer time.

The differences between the hydrostatic stability at the edge or the center of a "finger" interface and between static and dynamic instabilities have not always been clearly understood. Wendt (1962), for example, derived the necessary condition for *static* instability of an interface across which two solutes are diffusing as

$$R_\rho = \Delta\rho_1/\Delta\rho_2 < \tau^{-1/2}, \tag{1}$$

where $\Delta\rho_1$ and $\Delta\rho_2$ are the initial density excesses due to the faster-diffusing solute 1 in the lower layer and solute 2 in the upper layer. Huppert & Manins (1973) showed, however (see also the theoretical section below), that the center of such an interface can become *dynamically* unstable when

$$R_\rho < \tau^{-3/2}. \tag{2}$$

Since $\tau < 1$, (2) is less restrictive than (1), and dynamic instabilities in the form of the alternating finger motions sketched in Figure 3 can set in while the net density gradient remains statically stable at all times. For the heat-salt system, fingers can form when R_ρ is as large as 10^3, since $\tau \approx 10^{-2}$. Further laboratory experiments have unambiguously confirmed these results (Griffiths 1979b, Huppert & Hallworth 1984).

Fluxes have also been measured across finger interfaces. The most recent results for heat-salt fingers, as well as a critical survey of previous work, have been presented by McDougall & Taylor (1984), who extended the measurements to values of R_ρ as low as 1.2, into the range that is of most interest in the ocean.

FORMATION OF LAYERS FROM A GRADIENT   Layers can form when a smooth, statically stable gradient of one property has an opposing gradient of a second property superimposed on it, or when there is a vertical flux of the destabilizing component. The first case is the one for which most of the theory has been developed, but the formation of double-diffusive layers is most readily demonstrated in the second.

When a linear, stable salinity gradient is heated from below, for example, the fluid does not immediately convect from top to bottom. First, the heated layer immediately above the boundary breaks down to form a thin convecting layer. The depth d of this layer, and also the temperature and salinity steps ΔT and ΔS at its top, grow in time t like $t^{1/2}$, in such a way that $\alpha\Delta T = \beta\Delta S$ and there is no net density step (Turner 1968). The thickness δ of the diffusive thermal boundary layer growing ahead of the convecting

layer is also proportional to $t^{1/2}$ and therefore to d, the multiplying constant being

$$Q = \frac{\delta}{d} = D_{11}S_*/H_*. \tag{3}$$

Here $H_* = -g\alpha F_T/\bar{\rho}C$ is the imposed buoyancy flux corresponding to the heat flux F_T into a fluid of specific C, and $S_* = -g\beta dS/dz$ is a measure of the initial salinity gradient. The behavior of the system depends only on Q and on the molecular properties through the Prandtl number σ and the diffusivity ratio τ. When Q and τ are small, the criterion for instability of the thermal boundary layer is a critical Rayleigh number based on δ and ΔT; after this criterion is achieved at a certain thickness, the depth d remains constant, and a new layer grows on top of the first. Huppert & Linden (1979) have reported a combined numerical and experimental study that treats the sequential formation of new layers at the top of the region while the lowest layers merge in pairs. Linden (1976) has used the sugar-salt system to study the case where there is a destabilizing salt (T) gradient, partly compensating the stabilizing sugar (S) gradient in the interior. He found that as the two gradients become nearly equal but opposite, the layer properties depend less and less on the boundary flux, which then just acts as a trigger for an internal instability.

Convecting layers can also be produced in the "finger" situation, with a flux of sugar (S) imposed at the top of a salt (T) gradient (Stern & Turner 1969). Fingers form, grow, and break up because of a collective instability having the form of an internal wave (Stern 1969, Holyer 1981); this process produces a convecting layer that deepens, bounded below by an interface containing shorter, stable fingers. These fingers in turn grow and become unstable, thus producing a second convecting layer. Viewed on the scale of the convecting layers, there is a strong similarity between finger and diffusive systems, since they are both driven by unstable buoyancy fluxes through the bounding interfaces.

2.2 Stability Analyses

Some of the statements made above in describing the experiments can only be properly justified by referring to the corresponding theoretical results. Only a few examples can be treated here, and these only in brief, but even the simplest stability arguments go some way toward describing and explaining what is observed, especially in the finger case.

LINEAR THEORIES WITH SIMPLE DIFFUSION OF TWO COMPONENTS   The early developments, initiated by Stern (1960), were reviewed by Turner (1974). Stern was the first to consider the stability of temperature and salinity

profiles, both of which increase linearly with height between two free horizontal boundaries separated by a distance d and held at fixed temperatures and salinities. He thus concentrated on the "finger" case, though he mentioned the "diffusive" case in a footnote. His results are applicable to any pair of independently diffusing components, and they have since been extended (e.g. see Baines & Gill 1969) explicitly to the diffusive case and to other boundary conditions. Here we consider ΔT and ΔS to be the differences in solute concentrations between the boundaries; positive values imply that these are greater at the lower plate. Four nondimensional parameters are required to specify such systems. Using the notation already introduced, these can be defined as $R = g\alpha\Delta T d^3/D_{11}v$, the Prandtl number $\sigma = v/D_{11}$, $\tau = D_{22}/D_{11}$, and $R_\rho = |\alpha\Delta T/\beta\Delta S| = |\alpha T_z/\beta S_z|$. A frequently used combination is $Rs = R/R_\rho$.

Assuming small disturbances of the form $T' = e^{pt} \cos \pi ax \cdot \sin \pi bz$, for example, from the state of constant vertical property gradients T_z and S_z and no initial motion, we obtain a cubic characteristic equation in p, with coefficients that depend on R, Rs, σ, and τ. In a given fluid (fixed σ and τ) the stability boundaries may be calculated as lines in the R, Rs plane, and the character of the initial motion determined in each region. When $R > 0$ and $Rs < 0$ (the finger regime), the motion sets in as a direct, growing mode when

$$-R - Rs/\tau > \frac{27}{4}\pi^4. \tag{4}$$

When R and Rs are large (e.g. because the diffusivities are small), fingers can form provided

$$R_\rho < \tau^{-1}. \tag{5}$$

When τ is small, instability sets in at values of $Rs \ll -R$, i.e. while the system is still statically stable. In this limit, the fastest-growing motions have a horizontal length scale $\ell \sim (g\alpha\Delta T/dD_{11}v)^{-1/4}$, which is much smaller than d and represents a balance between the destabilizing diffusive effects and the dissipative viscous effects. This is in agreement with observations, and though the linear stability theory is not strictly applicable to fully developed fingers, a later finite-amplitude calculation by Huppert & Manins (1973) using the same force balance has verified that this is the relevant scale. We recall that these authors also derived the condition for dynamical instability at an interface, quoted in Equation (2) above. It can now be seen that this condition differs from the static condition (1) because of the factor τ^{-1} appearing in Equation (5).

Corresponding results have been obtained for the "diffusive" case ($R < 0$, $Rs > 0$), where it is found that instability to small perturbations occurs in

an overstable or oscillatory mode and (at large $|Rs|$) when the net density distribution is statically stable. Many of the recent developments in this area relate to finite-amplitude calculations, which are discussed separately.

The calculations for the finger case have been extended by Schmitt (1979a, 1983) to cover general values of σ and τ. He has computed the maximum growth rates, the flux ratios of the two components (at particular values of R_ρ), the wave numbers of the fastest-growing fingers, and a "bandwidth," or measure of the spread of scales evolving from a white-noise spectrum. The fastest-growing fingers occur at low values of σ and τ. The flux ratios $\gamma = \alpha F_T/\beta F_S$ calculated by Schmitt (1979a) using similarity solutions and linear gradients are in good agreement with laboratory values for two-layer heat/salt and salt/sugar systems (Turner 1967, Schmitt 1979b, Lambert & Demenkow 1972, Griffiths & Ruddick 1980). With this encouragement, Schmitt (1983) produced the more general results shown in Figure 5. Note that for $\sigma > 1$ and $\tau < 10^{-1}$ the flux ratio lies between 0.5 and 0.7 at $R_\rho = 2$, and it is predicted (but not yet tested experimentally) that systems, such as liquid metals, with low σ and τ should have low γ (0.1–0.2).
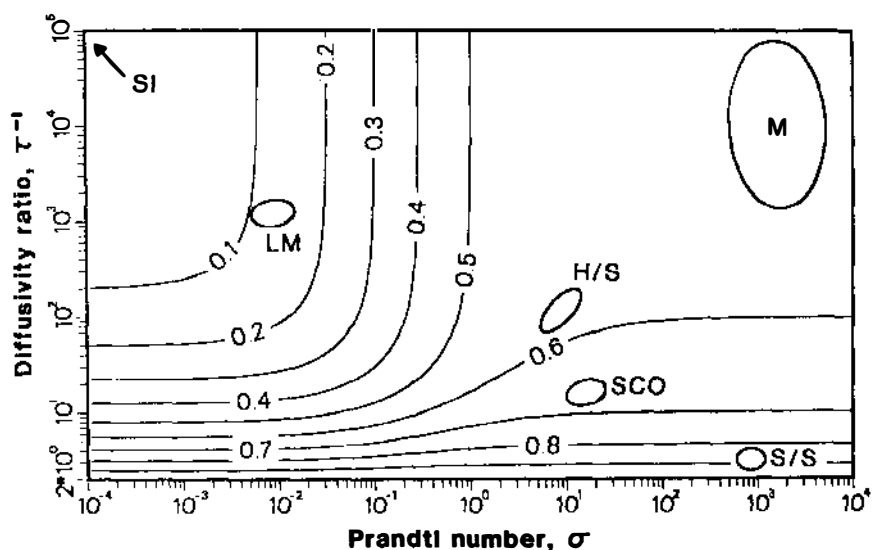

Figure 5   Contours of the flux ratio $\gamma = \alpha F_T/\beta F_S$ in fingers, calculated for general values of σ and τ with $R_\rho = 2$ by Schmitt (1983). The regions corresponding to a number of natural double-diffusive systems are denoted by the following abbreviations: LM = liquid metals and metallic semiconductors, SCO = semiconductor oxides, H/S = heat/salt, S/S = salt/sugar, M = magmas. The region for stellar interiors (SI) lies off the diagram at about $\sigma = 2 \times 10^{-6}$, $\tau = 10^{-7}$.

The growth-rate calculations in the relevant σ, τ ranges indicate that stellar interiors, liquid metals, and warm heat/salt systems should develop vigorous fingers, while salt/sugar and magma systems will be more sluggish.

COUPLED MOLECULAR DIFFUSION   When cross-diffusion terms are significant, further kinds of instabilities become possible. If the fluxes are parameterized in terms of the gradients by

$$-\text{Flux of } T = D_{11}\nabla T + D_{12}\nabla S, \tag{6}$$

$$-\text{Flux of } S = D_{22}\nabla S + D_{21}\nabla T, \tag{7}$$

with the density ρ of the fluid given by $\rho = \rho_0(1 + \alpha T + \beta S)$, the cases already discussed correspond to $D_{12} = D_{21} = 0$. These relations apply strictly only to isothermal ternary systems, but when T and S represent heat and solute, D_{21} is proportional to the Soret coefficient. Hurle & Jakeman (1971) have shown that a large value of D_{21} produces a concentration gradient near a heated boundary, which can influence the form of the instability. For example, a homogeneous layer of water-methanol mixture (with a low concentration of methanol) can break down when heated from below in an oscillatory rather than direct mode because of the stabilizing concentration gradient set up in this way. Schechter et al. (1972) and Verlande & Schechter (1972) have investigated theoretically the effect of heating above and cooling below a thin layer of solution, and they have shown that double-diffusive instabilities can and do occur when the density gradient is statically stable (see also Platten & Legros 1983).

McDougall & Turner (1982) and McDougall (1983a) have taken a further step and investigated the conditions under which fingers can form in a ternary system where both D_{12} and D_{21} can be significant. On following through the linear stability analysis for small perturbations from linear gradients and no initial motion, the condition for finger instability with $\alpha T_z < 0$ can be written as

$$\left(\frac{\beta}{\alpha}\frac{D_{21}}{D_{22}} - 1\right) + \frac{1}{\tau}\frac{\beta}{\alpha}\frac{S_z}{T_z}\left(\frac{\alpha}{\beta}\frac{D_{12}}{D_{11}} - 1\right) > \frac{27}{4}\frac{\pi^4}{R}\left(1 - \frac{D_{12}D_{21}}{D_{11}D_{22}}\right). \tag{8}$$

This reduces to (4) when $D_{12} = D_{21} = 0$, but clearly considerable extra complexity is introduced by adding these two coefficients. McDougall (1983a) has sketched the stability boundaries as functions of $\beta D_{21}/\alpha D_{22}$ and $\alpha D_{12}/\beta D_{11}$ for specified ranges of αT_z and βS_z. He has also shown that essentially the same expression as (8), with the right-hand side zero, is obtained for finite-amplitude fingers if T_z and S_z are now taken to be the vertical gradients of the horizontal average of T and S through the fingers (cf. Huppert & Manins 1973).

Of special interest is the case where *both* components are hydrostatically stably stratified. From (8) it is seen that instability is assisted by large positive values of both D_{12} and D_{21}, and that at least one of $\beta D_{21}/\alpha D_{22}$ and $\alpha D_{12}/\beta D_{11}$ must exceed unity. Figure 6 illustrates the variations of properties in the upgoing and downgoing fingers when $D_{12} = 0$ and Equation (8) is satisfied; this should be compared with Figure 3, which shows the variations in "ordinary" fingers (when $D_{12} = D_{21} = 0$). In Figure 6, the T concentration of the downgoing fingers increases as a result of the diffusion of T from the upgoing into the downgoing fingers, in a manner that is unaffected by coupled diffusion. The flux of S between the fingers is, however, due mainly to the cross-diffusion flux driven by the spatial gradients of T; even though the concentration of S in a downgoing finger is already greater than that in the upgoing finger, it continues to increase because of the D_{21} term. Note that the horizontally averaged density gradient is statically stable, and that the downgoing fingers are more dense than the surrounding upgoing fingers. Thus, again denser fluid will be transported downward and less dense fluid upward, and the total gravitational potential energy of the fluid will decrease, in accordance with our extended definition of multicomponent convection.

McDougall (1983a) has generalized the above results to include nonzero values of both D_{12} and D_{21} and has also examined the instability of initially sharp interfaces with cross-diffusion included. When both components are stably stratified initially, or even when they are distributed in the "diffusive" sense, a large cross-diffusion term can lead to the formation of fingers before the density gradient at the center of the interface becomes statically unstable. Comparisons of this theory with experiments by polymer chemists are made in Section 3.2.

INFLUENCE OF A THIRD DIFFUSING COMPONENT   Griffiths (1979a,b) extended the linear stability analyses to include three solutes (without cross-diffusion effects). In this case, the characteristic equation is fourth order, rather than a
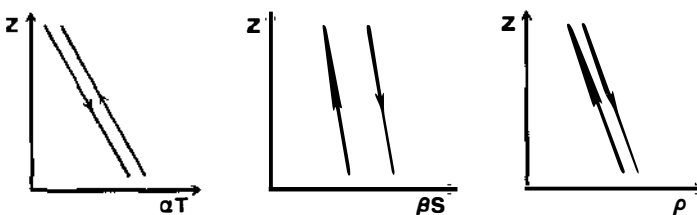

Figure 6   Sketch of the vertical profiles of αT, βS, and ρ in the centers of up- and downgoing fingers with both T and S stably stratified. Here $D_{12} = 0$ and D_{21} satisfies Equation (8), so that the cross-diffusive flux of S due to the T gradient sustains the density anomalies that drive the motion.

cubic, and the stability boundaries are planes in the space defined by three Rayleigh numbers. He gave special attention to systems with one diffusivity (say D_{11}) much larger than the other two (D_{22} and D_{33}) and with large Prandtl number, i.e. to the heat/two-solute system. Small concentrations of a third property with a smaller diffusivity can profoundly affect the nature of diffusive instabilities. For example, oscillatory and direct finger modes can be unstable simultaneously when the density gradients due to components with the largest D_{11} and smallest D_{33} diffusivities are of the same sign. What is actually seen experimentally depends on the relative growth rates of the two modes, and for three components of comparable diffusivities, fingers will be dominant.

Griffiths (1979b) has extended this analysis, using the technique that led to (2) in the two-solute case, to determine the type of interface formed between well-mixed layers containing three diffusing components with different relative concentrations. He tested his theory using three solutes (1: KCl, 2: NaCl, and 3: sucrose in water, listed in order of decreasing diffusivity), first with component 2 above 1 and 3, and then with 2 and 3 above 1. His experimental results are in good agreement with the theoretical criterion, which predicts the marginal concentration ratio $\beta_3 \Delta C_3 / \beta_1 \Delta C_1$ for fingers to form as a function of the overall density ratio R_ρ. Fingers forming away from the center, with a "diffusive" core, were also observed in a certain parameter range.

A final step in this series of related studies is mentioned now. Griffiths (1979c) measured the transport of multiple components through heat-solute diffusive interfaces, following some exploratory experiments by Turner et al. (1970). He also predicted theoretically that the ratio of transport coefficients K_i, defined as the mass flux divided by the corresponding concentration step, should be proportional to $\tau^{1/2} = (D_{33}/D_{22})^{1/2}$ at low total solute-heat density ratios R_ρ^T, and to τ at higher R_ρ^T, where a steady diffusive core dominates. The experiments with R_ρ^T between 2 and 4 were consistent with $K_3/K_2 = \tau$, but there was an even greater separation of components at higher R_ρ^T. This discrepancy has not been explained, but it is unlikely to result from the neglect of cross-diffusion terms. The separation of different salts during transport through a finger interface, on the other hand, is negligible when heat is the stabilizing property and the solutes are destabilizing.

NONLINEAR THEORIES   Experiments such as the heating of a salinity gradient from below make it clear that a series of diffusive layers and interfaces can only be properly explained theoretically using nonlinear theories. The oscillatory instability predicted by linear theory is rapidly overtaken by monotonic motion and convective mixing within the layers,

and there is not a close correspondence between the form of the growing motions and the final steady state, as there is in the finger case.

Huppert & Moore (1976) tackled the "diffusive" system numerically and found two different solution branches, one oscillatory and one steady. For $\tau = 0.1$, the smallest value for which it was convenient to run the numerical procedure, they found that nonlinear steady convection can occur at a lower value of the thermal Rayleigh number R than the linear prediction for the oscillatory mode. Recent numerical experiments by Toomre et al. (1982) have extended Huppert & Moore's (1976) calculations to $\tau = 0.01$ and $R \approx 10^6$. The solutions exhibit time-dependent behavior, including a sequence of period doublings, leading eventually to chaotic behavior. Such results have more general implications for the behavior of systems of differential equations describing other physical systems, and this aspect of the theoretical work has taken on a life of its own (e.g. see Marzec & Spiegel 1980).

Proctor (1981) has described the steady solutions near the minimum value of R, using an analysis valid as $\tau \to 0$. For small enough τ, salt can be swept to the boundaries of the convection cells, so that the S field is uniform in the interior and has large gradients only at the edges. A value of R sufficient to initiate purely thermal convection is thus able to drive steady motion, no matter how large the stabilizing salt field is initially.

There have been various attempts at steady and unsteady numerical models of a series of diffusive layers and interfaces, but none of these are really satisfactory yet. Piacsek & Toomre (1980) have produced a numerical model of salt fingers growing across an initially sharp interface. Though the ends of the fingers became unstable in this calculation, they did not evolve into convecting layers developing independently of the fingers, possibly because only a few fingers were included in the calculation, thereby restricting the horizontal scale of the convecting motions.

A totally different theoretical approach has been used by Stern (1982) to study fingers between two deep layers. He has applied a variational principle to place an upper bound on the salt flux, keeping the heat and momentum fluxes constant.

2.3   Direct Applications of One-Dimensional Results

Before we add extra physical effects, in other geometries, there are several examples of practical importance that can be discussed immediately.

LAKES AND SOLAR PONDS   Many lakes have now been described that are more saline at the bottom, where they are heated by the Sun or by geothermal energy. A good example is Lake Kivu (Newman 1976), which contains many well-mixed layers separated by diffusive interfaces. Griffiths (1979c) has shown that laboratory results on the transport of different

components through these interfaces give a satisfactory explanation of the observed fluxes.

The main large-scale engineering application of double-diffusive concepts is to solar ponds, shallow artificial lakes that are density stratified. Radiant heat is trapped and stored in a bottom layer, with an insulating gradient layer above; the aim is to prevent convection that would increase the heat losses (Poplawsky et al. 1981). The salinity gradient is not constant with depth, and this has prompted theoretical studies (Walton 1982) of the breakdown, which is found to occur preferentially (in agreement with observations) in a thin layer where the salinity gradient is a minimum. The behavior of interfaces at the large values of R_ρ observed in solar ponds has been examined theoretically and experimentally (Newall 1984). The convective heat flux persists up to $R_\rho = 28$, well beyond the value given by theoretical models based on the stability of the interface, which predict a cutoff around $R_\rho = \tau^{-1/2} = 9$ (Linden & Shirtcliffe 1978). Allowance for extra entrainment due to the turbulence in the convecting layers extends the range over which the transports remain nonzero (McDougall 1984).

There are still many unsolved practical problems relating to long-term unsteady behavior of interfaces (for example, the erosion of thick interfaces) and to replenishment methods. Layering caused by sloping sidewalls may also be significant for the operation of solar ponds.

LIQUID NATURAL GAS TANKS   The phenomenon of *rollover* in LNG tanks has continued to receive attention. In storage tanks refilled from below with denser but warmer LNG than that already present, one or more layers, separated by diffusive interfaces, will tend to form. As the lighter fractions boil off at the top, the upper layer can increase in density more rapidly than the lower layer can by cooling through the interface, so that overturning occurs. This leads to an uncontrollable increase in the rate of release as warmer LNG is convected to the surface.

The earlier consideration of two-layer systems has been followed by calculations of the mixing between the new and resident LNG (Germeles 1975), but the influence of the multiple layering that can develop during this process has not yet been properly evaluated. Multi-diffusive convection is also likely to occur because of the different chemical compositions of LNG from various sources. The latter aspect is complicated by the fact that the diffusivities are not well known in this system.

3.   IMMEDIATE EXTENSIONS AND APPLICATIONS

A wide range of related phenomena are now described briefly in the context of their most common application (though some cross-references and overlap are inevitable).

3.1 *Oceanography*

The literature in this field, which has provided the main motivation for the development of the subject of double-diffusion over the past 25 years, is now very large. There is no doubt that double-diffusive transport mechanisms are significant in particular regions of the ocean, but their overall importance relative to other mixing mechanisms still awaits a systematic assessment (Turner 1981, Caldwell 1983). A selection of the more recent observational, laboratory, and theoretical developments of particular relevance to this field are presented in the remainder of this section.

SIGNATURES OF DOUBLE-DIFFUSIVE PROCESSES   New instruments with high resolution have produced many profiles of temperature and salinity showing a "finestructure" of well-mixed layers separated by sharp, nearly horizontal interfaces. When temperature and salinity both increase or both decrease with depth across these interfaces, in such a way that their density effects are opposing and nearly compensating, it is reasonable to presume that double-diffusive convection will be important. Two further generalizations have also emerged. First, conditions are especially favorable for double diffusion to occur when a layer with compensating T and S differences intrudes at its own density level into an environment with different properties. Layers are thus most prominent near regions of strong horizontal property contrasts, i.e. across fronts, and this has led to the studies reported in the next section. Second, the smallest scale fluctuations or microstructure should always be examined in relation to the finestructure. There are now many measurements (see, for example, Gargett 1976, Schmitt & Georgi 1982) that show that the regions of most intense activity are the upper and lower boundaries of intrusions. Once diffusive or finger interfaces have formed, by whatever mechanism, there is reason to believe that the fluxes through them are adequately treated using one-dimensional models.

The existence of salt fingers in the ocean has been demonstrated in several ways. Gargett & Schmitt (1982) have reported direct measurements of temperature fluctuations attributable to fingers, which have a quasi-periodic structure and limited amplitude. The observed spectral shape compares well with theoretical predictions, and the buoyancy flux estimated directly from the variance of temperature gradient is in fair agreement with predictions using laboratory flux laws. Fingers have also been observed directly, using an optical method (Williams 1975, Schmitt & Georgi 1982). The images can be quantified, and different structures can be related to fingering or mechanical mixing activity. The latter authors have shown that finger activity is strongest at $R_\rho \approx 1$, and shear instabilities associated with low Richardson numbers (due to shears created by internal

wave motions) dominate near $R_\rho = 2$. Their correlations indicate that double diffusion is more common than shear instabilities in frontal interleaving zones, but they do not identify the mechanism responsible for creating the interleaving structures.

It has also been plausibly argued (Schmitt 1981) that the characteristic curvature of the T/S relation in central waters is consistent with R_ρ = constant, a condition that can be maintained by (unequal) double-diffusive transports of T and S but that is inconsistent with equaly eddy diffusivities (which would produce a linear T/S relation). Gordon (1981) has also attributed observed large midthermocline mixing rates in the South Atlantic to salt-finger activity.

Two recent studies related to "diffusive" interfaces are worth reporting. McDougall (1981) has shown that the nonlinearity of the equation of state (e.g. in seawater near $0°C$) modifies normal double-diffusive convection by causing asymmetric entrainment across an interface. This is more important than the previously discussed "cabbeling" process, which becomes possible where mixtures of two water masses are more dense than either parent mass. These results can be used to predict the signature imposed on a series of migrating interfaces by the asymmetrical entrainment. McDougall (1983b) has also developed a model for the formation of Greenland Sea bottom water, in which there is a balance between the advection of Atlantic water toward the center of the Sea and subsurface modification of its properties by double-diffusive convection.

HORIZONTAL GRADIENTS AND FRONTS   There have been many theoretical and laboratory studies following up early work on the effect of horizontal property gradients. Paliwal & Chen (1980) have generalized the problem of convection in a slot by considering stratified fluid contained in a small gap set at any angle to the vertical. Linear stability theory predicts the observed critical Rayleigh number and the scale of the steady convection cells. The case of a wide gap, with layers forming at a single heated or cooled sidewall, has been studied experimentally by Huppert & Turner (1980). They also examined vertical ice blocks melting into a salinity gradient (with an application to icebergs in mind) and showed that at Rayleigh numbers above about 10^5 the layer scale was

$$h = 0.65 \lfloor \rho(T_w, S_\infty) - \rho(T_\infty, S_\infty) \rfloor \left/ \left| \frac{d\rho}{dz} \right. \right., \tag{9}$$

where the subscripts w and ∞ denote T and S values at the wall and in the far field, resepctively (i.e. the layer scale depends only on the temperature difference and not on the presence of a melt layer near the wall). Huppert et al. (1984a) have extended these experiments to include crystallization at the

wall, or other solutes, and have shown that this conclusion appears also to be independent of σ and τ, i.e. of the molecular properties.

Tsinober et al. (1983) have described experiments in which layers were produced by heating at a small source in a linearly stratified salt solution. A plume penetrated to an increasing height above the source, with layers forming above and around it; in time, the evolving pattern shown in Figure 7 was set up, with features related to both bottom- and side-heating experiments. The convecting layers sloped downward as they moved radially away from a central plume into cooler surroundings, setting up systematic shearing motions with an outflow at the top and inflow at the bottom of each extending layer.

Intrusions of one fluid injected at its own density level into a gradient of another have been studied in the laboratory by Turner (1978), and the intrusive motions produced at a front (separating salt and sugar solutions with the same vertical density gradient) have been examined by Ruddick & Turner (1979). In the latter case a series of regular interleaving layers
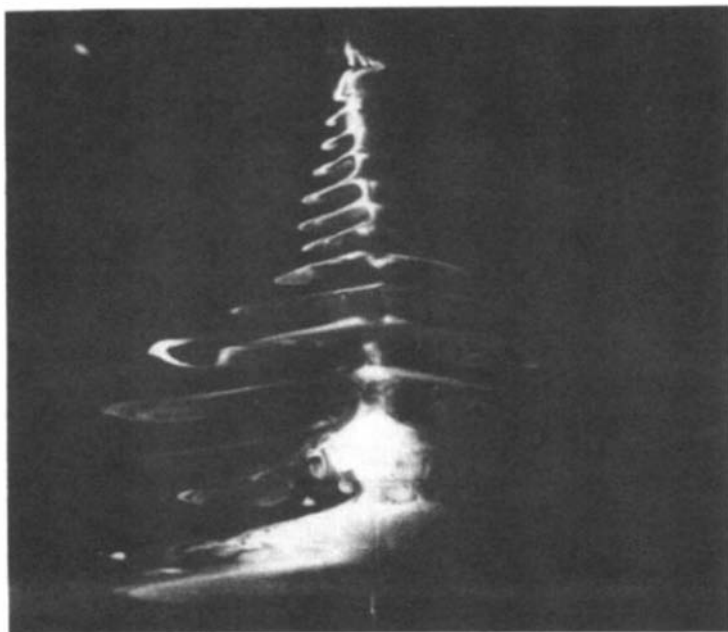
Figure 7   A series of double-diffusive layers produced after 35 min by heating a small region at the bottom of a salt-stratified tank 30 cm deep. Fluorescent dye placed near the bottom was lifted into convecting layers, which formed from the bottom up, and was also spread sideways because of the horizontal property gradients. (From Tsinober et al. 1983.)

develops, with both the vertical spacing and speed of advance approximately proportional to the horizontal property difference. There is a characteristic tilt of the layers across isopycnals as they advance that results from the net buoyancy flux through the finger interfaces. This tilt has now been documented in many ocean observations and supports fingering as the most probable driving mechanism; a particularly well-documented example has been described by Gregg (1980). Larson & Gregg (1983) have compared measured dissipation rates in profiles through intrusions with the production of turbulent energy by double-diffusive buoyancy fluxes. Their measurements are consistent with simple salt fingering in some regions, but they suggest some production of energy by shear in others; in addition, they point to the need for more accurate laboratory measurements of fluxes, especially at values of R_ρ near unity.

Note the remarkable general implication of the measurements that indicate that salt fingers are important in intrusions. Not only can molecular diffusion affect the motions on the layer scale, i.e. tens of meters in the vertical, but it can also, by driving the intrusions across fronts, influence large-scale mixing between water masses.

There is as yet no completely satisfactory theoretical description of the height scale or velocity of double-diffusive intrusions. Ruddick & Turner (1979) used a simple energy argument that is consistent with the vertical scale observed in their experiments with a sharp vertical front. For the oceanographically more realistic case of smooth, compensating horizontal gradients of T and S, Stern (1967) used a linear stability analysis to show that with vertical gradients in the finger sense, variations of T and S along isopycnals can be dissipated by interleaving motions that are driven by salt fingers. Toole & Georgi (1981) have extended Stern's results in a series of calculations (some of them including rotation) that give the scale and tilt of the fastest-growing mode. Their model is based on the assumption of eddy fluxes of heat and salt due to fingers, which depend only on the vertical gradient of S. Holyer (1983) has considered a similar problem (in the nonrotating case), but she parameterizes the flux divergences in a manner that uses molecular diffusivities and also includes horizontal derivatives of T and S. Her analysis is therefore indicative of the effect of horizontal gradients on the fingers themselves, rather than descriptive of the larger-scale horizontal interleaving that is caused by the fingers. It is still difficult to reconcile any of these theories with the laboratory results or with the limited field observations.

LANGMUIR CIRCULATIONS IN THE SURFACE LAYER   All the phenomena considered so far have depended on the presence of different thermal and solute diffusivities. Now we discuss a process that involves the diffusion of

momentum, the two relevant transport parameters being a turbulent or eddy diffusivity K_T and an eddy viscosity v_T. Leibovich (1983) has recently reviewed the theories related to Langmuir cells and the formation of windrows. Their generation depends on an instability mechanism in which there is an interaction between a wind-driven current shear $U(z)$ and a Stokes drift produced by surface waves. Physically, this implies that the vertical vorticity associated with small lateral variations of the shear flow is twisted by the presence of the Stokes drift into that of the Langmuir circulations, with growing motions in a plane transverse to the wind direction.

When a stabilizing temperature gradient is added to this flow, the linearized equations governing the stability to rolls aligned with U are precisely those describing double-diffusive instabilities in a salinity gradient heated from below. In this case we have $v_T > K_T$, and the velocity plays the role previously taken by temperature, and temperature that of the stabilizing salt gradient in the heat/salt problem. The results have been extended to finite amplitude, including regions of monotonic growth in which the Langmuir cells distort the initial constant temperature gradient. These calculations produce a nearly uniform layer (in the horizontally averaged temperature) with a strong thermocline below it, but it is not clear if they properly describe vertical mixing, since the dynamical effects of the temperature field have not been considered.

Another instability involving angular momentum and horizontal density gradients has been applied in the oceanic context (Lambert 1982); this is treated in the astrophysics section (Section 4.1), since it was first discussed in that field.

3.2   Chemical Studies

Chemists measuring diffusion coefficients, and trying to understand the variations with concentration and temperature, have usually sought to avoid convective effects because they introduce unwanted complications. In some cases, however, they can be exploited [e.g. by Caldwell (1973) in his determination of the Soret coefficient for NaCl]. Furthermore, the deliberate exclusion of multicomponent convection from experiments might in fact make the measurements irrelevant to practical cases in which convection does occur. The influence of cross-diffusion effects in particular has already been discussed in general terms. Now we refer specifically to several observational studies carried out in a chemical or biochemical context that are consistent with this mechanism, but that have previously been described in a very different way.

OBSERVATIONS OF FINGERS IN POLYMER SOLUTIONS   In a series of papers, Preston and coworkers (Preston et al. 1980, Comper et al. 1983) have

investigated transports between two layers containing various polymers in aqueous solution. An extensively studied case is a ternary system containing dextran and a small amount of a high-molecular-weight, long-chain polymer PVP in the bottom layer, and a less-concentrated dextran solution in the layer above it; in some experiments they added a trace of sorbitol to monitor the water in the bottom layer. The transport of either radioactively labeled dextran or PVP in this experiment is extremely rapid compared with ordinary diffusion in binary systems.

The PVP concentration in the upper layer increases linearly in time, rather than as $t^{1/2}$ (which is characteristic of a diffusion process), while the sorbitol (and water) do obey the $t^{1/2}$ relation. This rapid polymer transport is a consequence of the formation of fingers and the associated vertical convective motions. The phenomena continue to be observed when both components are more concentrated at the bottom, or even when the lower solution is supplemented with either low molecular weight solutes (such as NaCl) or dense solvents.

The theory of McDougall (1983a) has been applied to this case. He has argued (as described earlier) that cross-diffusion terms, if they are large enough for (8) to be satisfied, can allow fingers to form because of the property variations set up in the counterflowing fingers (see Figure 6). An extra (favorable) factor of $\tau^{-1/2}$ appears in the criterion for cross-diffusion fingers to form from an initially sharp interface [cf. the difference between Equations (2) and (5)]. There are as yet few quantitative experiments, and definitive comparisons between theory and experiment are difficult because of the lack of measurements of D_{12} and D_{21}. It would be especially valuable to know all four coefficients for a ternary system containing low-molecular-weight solutes with globular molecules so that the importance of coupled diffusion could be assessed and compared with possible effects due to networks of long-chain molecules in the polymer systems.

PREVIOUS DESCRIPTIONS AND INTERPRETATIONS   The motivation for much of the work just described has been biological, with the eventual aim of relating the observations to transport phenomena in connective tissues and membranes. In the laboratory model system, however, it seems clear that gravity has always been an important factor, since all the ordered structures (which we have identified with fingers) grow vertically. The weak dependence of fluxes on g observed in centrifuge experiments (Preston et al. 1980) is also consistent with this view (McDougall 1983a).

Explanations of the structure in terms of chemical potential gradients and other concepts familiar to physical chemists tend to obscure this dependence on gravity, and they give what I believe to be false hope that similar arguments might be extended to nonhorizontal interfaces. Consider again the simple two-layer system sketched in Figure 4. The major energy

change (and increase in entropy) between states 1 and 2 are certainly associated with the free energy. Because the solute S is more uniformly distributed in state 2, there has been a decrease in the Gibbs free energy of $RT \ln 2$ per mole (where R is the gas constant and T the absolute temperature), which for 10-cm-deep layers of molar concentration amounts to approximately 17 J cm^{-2}. The corresponding change in gravitational potential energy is only about 10^{-4} times as large, and if the second component is included, the contrast between the gravitational potential energy and the free-energy changes is even greater. But the fact that the free-energy change is so much larger does *not* mean that it is the most important kinetically. It is the gravitational term that appears in the dynamical equations (i.e. the equations of motion) and provides the mechanism for organizing the flow into the efficient form shown in Figure 3. The coupling between vertical motions driven by gravity and diffusion acting on small scales produces much more rapid transports of properties than is possible with molecular diffusion alone.

Note that the final state 3 in Figure 4 represents a further large decrease in free energy (as the second component also becomes uniformly distributed), while there is an *increase* in gravitational potential energy (back to the original value in the specific example described). The process is very slow because only molecular diffusion is now acting, and there is no release of gravitational potential energy to organize a convective flow. Arguments based on fluxes driven by osmotic pressure, or more detailed thermodynamic considerations of the effect of one component on the chemical potential gradient of another, can go further than the free-energy calculations sketched above in describing the molecular processes that produce coupled transports. They do, however, all suffer from the same drawback: They cannot in themselves explain the origin of the regular vertically oriented structures and the very much larger fluxes. These organized flow structures depend essentially on gravity and cannot be produced by diffusion alone.

4.   EXTENSIONS TO OTHER DIFFUSIVE SYSTEMS

4.1   *Astrophysics*

As noted in Section 1, there are many diffusing properties in stars and, therefore, a wide variety of multicomponent convection effects that can be included in the broader definition. A direct analogue of heat/salt diffusive convection has been used to explain the properties of large stars with a helium-rich core, which is heated from below and thus convecting. Spiegel (1972) has shown that variations in the helium/hydrogen ratio can produce a density gradient that limits the helium transport by double-diffusive

convection, though whether this may be in layers is still unclear. The process is called *semiconvection*, since it allows the motion outside the core to regulate itself and remain nearly convectively neutral. The potential temperature will be almost uniform (since this is the driving component), though there can still be a stabilizing gradient of He. The possibility of "helium fingers" in stellar atmospheres has been suggested too, following accretion of He-rich material onto a star.

ROTATION   An early example of an instability that is akin to salt fingers, but that is due to differential rotation in the radiation zone of a star (where heat transport is rapid), was discussed by Goldreich & Schubert (1967). If the angular momentum decreases radially outward, this provides a destabilizing distribution of the more slowly diffusing component (angular momentum) that can be superimposed on a stable temperature gradient. This could lead to the formation of "angular-momentum fingers," drawing on the energy of the differential rotation. McIntyre (1970) developed this theme further and showed both that a baroclinic circular vortex can be destabilized by diffusive effects in various ways and that oscillatory modes can also occur.

In the oceanic application where eddy viscosity and diffusivity may be appropriate and $\sigma > 1$, viscosity (now the faster transport process) can again be destabilizing and produce an overturning that leads to the formation of layers in a density stratification [see Lambert (1982) and the section on Langmuir circulations]. This type of instability occurs at Richardson numbers much greater than the critical value for nonrotating shear instability. Baker (1971) and Calman (1977) have described related laboratory experiments in which layers were produced by differential rotation of a disk in a rotating cylinder of water.

Worthem et al. (1983) have summarized relevant earlier work and carried out a linear stability analysis of a doubly diffusive (heat-salt) system on which rotation and shear have been superimposed. Their calculations reduce to the Baines & Gill (1969) results in one limit and the McIntyre (1970) rotating instability in another, and they indicate that shear and rotation in combination could cause instability and the formation of layers under oceanic conditions that would otherwise be stable. Recent theoretical work on rotating stars has added a compositional gradient (e.g. a H-He gradient) to temperature and angular momentum, where now the diffusivities have the relative magnitudes $\kappa_{He} \leq \nu \ll \kappa_T$.

MAGNETIC FIELDS   In the outer layers of stars like the Sun, thermal convection is affected by the presence of magnetic fields. The nature of magnetoconvection depends crucially on the ratio $\zeta = \eta/\kappa_T$ of the magnetic diffusivity η to the thermal diffusivity κ_T. In the astrophysical context ζ is

small (because of radiative heat transport), and in this regime a rich variety of both steady and oscillatory solutions are possible. Proctor & Weiss (1982) have recently reviewed this subject, and they assert that "convection in a magnetic field is now the best studied example of double-diffusive convection and serves as a guide to the behavior of related systems." Magnetic fields can be stabilizing or destabilizing; they are also vector functions that can be concentrated by convective motions, so there are phenomena that are peculiar to this system. For example, theory can now shed light on the existence and properties of sunspots, which are maintained by strongly nonlinear processes.

The opposite case of ζ large is relevant to laboratory experiments and to studies of the Earth's core, and this work too is the subject of recent reviews (Busse 1983, Loper & Roberts 1983). A further reference to this case is made later, though neither of these vast subjects can be pursued much further here. Though particular combinations of effects have been isolated to make the studies manageable, all these systems are in principle quadruply diffusive and can involve angular momentum, magnetic field, temperature, and composition simultaneously. There are few finite-amplitude calculations, and even fewer that include horizontal gradients or spatial nonuniformities.

4.2 *Metallurgy and Materials Science*

The phenomenon of crystal growth has taken on a new practical importance with the increasing needs of the electronics industry for larger and more chemically homogeneous crystals. Crystal growth from a solution or melt involves both heat and mass transfer, and these usually lead to convection in the fluid, often double-diffusive in character. In common practice, convection was regarded as always deleterious and thus to be avoided (perhaps by mechanical stirring). Detailed fluid-dynamical studies have only recently begun to contribute to the understanding of the processes that lead to fluctuations in growth rate and consequent nonuniformities in crystals.

STABILITY OF A HORIZONTAL PLANAR INTERFACE   As a crystal grows upward from the cooled horizontal boundary of a region containing a solution or melt, an initially planar interface may become unstable in two ways: morphologically or hydrodynamically. Morphological instability pertains to the spontaneous change in shape of a two-phase interface during a phase change. The consequent influence on surface boundary conditions couples this to hydrodynamic instabilities in the fluid above, which depend on both temperature gradients and compositional gradients set up by the depletion or concentration in the melt of the crystallizing components [see Langer (1980), Coriell & Sekerka (1981), and Azouni (1981) for reviews of recent

work]. The nature of the stability depends on the sign of the boundary concentration gradient, which decays exponentially away from the interface with length scale D/V, where D is the liquid diffusion coefficient and V the velocity of advance of the interface.

Linear stability analyses of the coupled problem (which have recently been extended numerically into the nonlinear regime) show that for large V and stabilizing boundary gradients of T and composition, the growing interface becomes morphologically unstable to short wavelength disturbances, which lead to the formation of dendrites. The mechanism has been understood qualitatively for many years; it is called "constitutional supercooling," and it depends on a local increase of solute concentration near the boundary. On the other hand, when a less dense component is rejected during crystallization and the T and compositional gradients are opposing, a truly double-diffusive instability is possible. The resulting fluid motions are described in the next section. The intermediate range, where there is close coupling between fluid and morphological effects, leads to more complicated behavior, including oscillatory modes. A noteworthy new numerical method of attack on this and related problems has been described by Ungar & Brown (1983).

Surface tension can also be important in this context, especially in a low-gravity environment. Convection phenomena produced, or partly driven, by gradients of interfacial surface tension are called "Marangoni effects." When these are combined with other driving forces, the problem becomes one involving multiple diffusivities, and the stability must be assessed accordingly. [Castillo & Velarde (1982) have used an energy argument to study this case.]

SOLIDIFICATION AND FLUID FLOW   The motions that develop above a cooled, rapidly crystallizing boundary at which lighter residual fluid is being released are closely related to salt fingers, though they are complicated by a "mushy zone" of crystals through which the convecting motions penetrate. These produce long, narrow segregated regions (called "freckles" because of their appearance in horizontal cross-sections), which can seriously weaken alloy castings. The phenomenon has recently attracted increased attention because of its relevance to solidification at the Earth's inner core (Roberts & Loper 1983).

It is still not possible to predict in detail the rate of growth of a dendritic interface: The presence of dendrites can retard the volume growth by a large factor compared with the theory for a planar interface. Associated with this growth is the rejection of solute laterally between the dendrite arms, and the behavior of even a single growing tip will depend strongly on the direction of the "compositional convection" set up by this process.

The large-scale effects of fluid flow in a solidifying ingot are better

understood (Fisher 1981), at least in a qualitative way. Macrosegregation, or the gross separation of different components by the relative motion of crystals and remaining melt under gravity, is accomplished by mechanisms related to those described in the next section. They are complicated by the large fraction of a casting that contains dendrites (so that much of the convection is interdendritic) and by flows due to the shrinkage accompanying solidification. Existing theories give good predictions when the latter effect is dominant, but the convective case remains a major unsolved double-diffusive problem.

4.3   *Geology*

The rate of transfer of ideas from oceanographic to geological contexts has accelerated in recent years, following the review by Turner & Gustafson (1978). They considered the various flow phenomena that can result from the efflux of hot, salty water from vents in the seafloor, in relation to the genesis of massive sulfide ore deposits. This application subsequently has been pursued further (McDougall 1984, Campbell et al. 1984). It was soon realized, however, that double-diffusive effects can also be important during the solidification of large magma chambers (i.e. in magmas as well as aqueous solutions). It can now be confidently asserted that they probably play a role in all major igneous processes, and much of the recent work [reviewed by Huppert & Turner (1981a) and more particularly by Huppert & Sparks (1984)] has been carried out with these processes in mind. Aqueous solutions have also been used for the laboratory modeling of magma chambers, with crystallization added to the double-diffusive systems previously studied; these laboratory experiments are our main concern here.

CRYSTALLIZATION AND COMPOSITIONAL CONVECTION   Once thermal and compositional gradients have become established in a chamber, multicomponent convection will inevitably lead to layering. The starting point of the recent fluid-dynamical work was the idea that extensive quasi-horizontal layers are more likely to form in the fluid state, and that the crystallization and final mineral layering can subsequently be affected by these preexisting layers. This is very different from the long-held idea that crystal settling dominates layer formation in igneous rocks.

Most of the convective phenomena studied, including those responsible for setting up the compositional gradients from which layering can develop, depend on density changes produced by crystallization. In magmas (and many common solutions that can be used as laboratory analogues), crystallization causes much larger changes in melt (or solution) density than the associated temperature changes. In Na_2CO_3 solution, for example, the

residual fluid is cold but less dense, and upward "compositional convection" can occur; its effects have now been studied in many different geometries.

Sidewall crystallization   Turner (1980) and Turner & Gustafson (1981) have shown how the cooling and crystallization of a homogeneous solution at a vertical wall generates a boundary-layer flow, which rises to the top to form a growing region that is thermally and compositionally stratified. Worster & Leitch (1984) have made a detailed theoretical and experimental study of the stratification produced by a laminar boundary layer rising along a plane vertical wall. Smaller-scale double-diffusive layering develops in the upper stratified region because of the cooling from the top and the side. When there are several components in the solution or melt, the same process leads to differentiation or fractionation, i.e. a vertical zonation of the several components.

McBirney (1980) has shown how this model can account for the changes with time in the composition of lava erupted from the same volcano, or the contrasting compositions from neighboring vents that tap different levels in a single stratified magma chamber. The striking trace-element gradients in ash-flow deposits derived from some high-silica rhyolite magma chambers (Hildreth 1981) can also be explained by convective fractionation. Highly fractionated fluids, with element concentrations depending systematically on their distribution coefficients, can be generated in boundary-layer flows that involve only a small proportion of the total chamber volume [see Sparks et al. (1984) for a fuller discussion]. In the future, the decisive tests of the dynamical ideas are likely to depend on a critical reevaluation of trace-element and isotopic data.

Top cooling   Chen & Turner (1980) also cooled and crystallized concentrated solutions from the top, with the roof of the container held below the eutectic temperature. These experiments have been followed up by Huppert et al. (1984b) with an application to komatiite (very high temperature) lavas in mind. Dendritic crystals were observed to grow down nearly vertically from the roof in a static fluid layer generated by the light residual fluid. The eutectic crystals advanced more slowly, with a horizontal front, and filled the interdendritic spaces (see Figure 8). The morphology of these crystals resembled the "spinifex texture" observed in komatiites, while the crystals growing on the bottom in a convecting layer were more solid and randomly oriented.

REPLENISHED MAGMA CHAMBERS   Experiments of a different kind have also been used to model chambers replenished from below with magma of different composition and temperature. Denser and less dense inputs, rapid

and slower injection rates, and homogeneous or already stratified tanks have all been investigated with particular geological processes in mind. Three cases, all with hot, denser inputs, are given here as examples.

Huppert & Turner (1981b) placed a hot layer of KNO_3 solution under a cold, lighter $NaNO_3$ layer, taking care to minimize the mixing and to produce a sharp (triple-diffusive) interface. The rapid convective heat transfer through the interface led to the growth of crystals on the bottom, so that the density of the residual lower solution decreased. When it became equal to that in the upper layer, the interface broke down and the layers mixed thoroughly together. This is the behavior to be expected in a basaltic magma chamber, with nearly equal viscosities of the two layers. When the viscosity of the upper layer was much greater (as it would be with rhyolitic magma), then Huppert et al. (1983) showed, using glycerine as the upper fluid, that crystallization along the interface led to light residual KNO_3 fluid being released continuously into the viscous layer. There was little mixing, and a layer of light KNO_3 was deposited above the glycerine (with, incidentally, fingers forming on the interface with the glycerine below).

With a rapid input of either lighter or denser fluid, there can be extensive turbulent mixing with the originally homogeneous fluid already in the chamber. Campbell & Turner (1984) have examined the case of a dense, hot, salty fountain that is ejected upward from a source in the base of a tank of freshwater and falls back to produce a stratified region that breaks up into
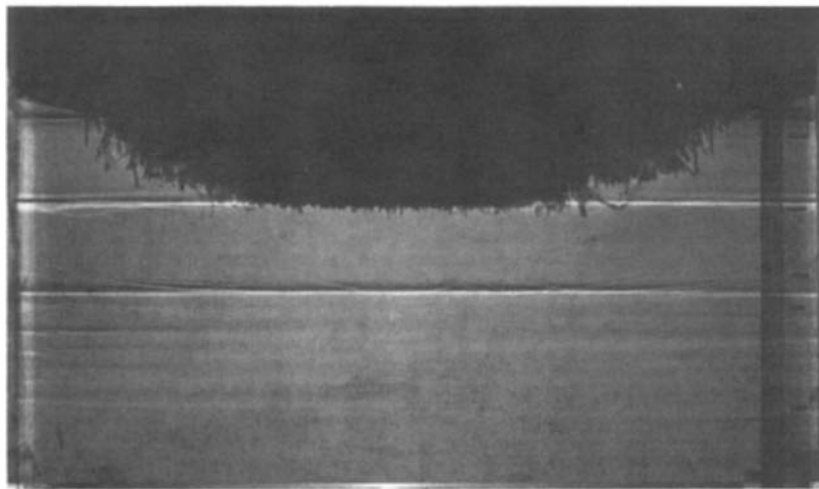
Figure 8 Shadowgraph picture (taken 18 hr after the start) showing the effect of strong cooling at the top of a tank containing a stable gradient of Na_2CO_3 solution. Convecting double-diffusive layers formed first, and these influenced the subsequent growth of the crystals and the eutectic layer above. (From Chen & Turner 1980.)

thinner double-diffusive layers. Campbell & Murck (1984) have used this model to explain the properties and distribution of chromite seams in large igneous intrusions such as the Bushveld. This experiment is an extension of the "filling box" model of Baines & Turner (1969), which is also the basis for the work of McDougall (1984) on double-diffusive plumes in confined environments and their application to massive sulfide ore deposits.

4.4   Geophysics

There are many large-scale examples of processes in the Earth that can be influenced by multicomponent or compositional convection. The modification of the composition of midoceanic ridge basalts by convective processes in a magma chamber below, and the more general problem of differentiation of the crust from the mantle have been discussed in these terms. Two further examples are now described.

MANTLE CONVECTION   It is now believed that mantle convection must be the major driving mechanism for plate tectonics. There is still a debate about whether the convection occurs throughout the mantle or is confined to the upper 650 km (Spohn & Schubert 1982). A possible alternative, discussed by Richter (1979) and Stevenson & Turner (1979), for example, is that the mantle is chemically layered, with independent convective circulations in the upper and lower mantle and heat transfer (but little transfer of matter) across a double-diffusive interface between them. McKenzie & Richter (1981) concluded that if convection does occur in two layers, then the present surface heat flux is determined mainly by the initial thermal conditions of the Earth and not by the radioactive generation of heat.

   Subducting plates are compositionally less dense than the mantle; they are made denser and plunge downward only because they are colder. As they sink, they equilibrate in temperature, and a small fraction of the lithosphere melts to form a water-rich component that is less dense than the surrounding mantle (see Ringwood 1982). This lighter material rises, and further melting occurs at shallower depths, to form andesitic volcanoes at an island arc or continental margin. This process too can be regarded as double-diffusive in character.

THE EARTH'S CORE   Several of the topics discussed in other contexts have implications for the dynamics of the Earth's core. Gubbins et al. (1979) and Loper & Roberts (1983) have convincingly argued that compositional convection, due to the solidification of dense metallic iron-nickel crystals on the inner core and the rejection of light components such as silicon, will be much more important than thermal effects in driving convection in the outer core. The latter authors further suggest that the boundary is likely to be dendritic, as observed for solidifying metal ingots; the evidence for this, however, is inconclusive.

An examination of the energetics of the core shows that compositional convection is in principle capable of sustaining a dynamo with a large toroidal magnetic field, whereas thermal driving is inefficient because of the heat conducted down the adiabatic gradient. The study of the magneto-hydrodynamic effects themselves still involves at least the magnetic field, rotation, and composition, all with different diffusion rates; this too has been recognized as a multicomponent convection problem.

5. CONCLUDING REMARKS

The first purpose of this article has been to show how the presence of several components in a fluid, each influencing the density and having different diffusive properties, can lead to dynamic instabilities, often well before a fluid system would become statically unstable. The convective phenomena described depend essentially on gravity or some other external force field. It is the coupling between this force field and diffusion that organizes the flow into a form that produces much more rapid transports than diffusion alone could achieve. Linear stability theories have now been extended to increasingly complicated multicomponent systems, and these give a first indication of the fully developed behavior; however, more nonlinear and numerical studies will certainly be needed in the future. Laboratory studies of idealized systems have been, and will continue to be, important in the development of this subject.

The second aim of this review has been to emphasize the very broad range of disciplines in which the effects of multicomponent convection should be taken into account. If these effects are ignored, a whole range of qualitatively different phenomena can be missed entirely. In particular, mixing rates will be seriously underestimated if only molecular diffusion is considered without taking into account the coupled convection that can occur in a gravitational field.

This review has been necessarily very selective, with space for only a few examples in each field. Other possible applications have not been discussed at all, and there are no doubt many more areas that readers will now be able to relate to the ideas presented here. The recent recognition that multicomponent convection is a fundamental phenomenon of importance in many disciplines should certainly lead to more rapid progress over the next few years.

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