

Non-Linear Diffusion I. Diffusion and Flow of Mixtures of Fluids

J. E. Adkins

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NON-LINEAR DIFFUSION

I. DIFFUSION AND FLOW OF MIXTURES OF FLUIDS

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A theory for the flow and non-linear diffusion effects in mixtures of fluids is formulated based upon hydrodynamical considerations. It is assumed that each point of the mixture is occupied simultaneously by all constituents in given portions. The motion of each constituent is governed by the usual equations of motion and continuity. The mechanical properties of each component are specified by means of constitutive equations for the stresses; diffusion effects are accounted for by means of a body force acting on each constituent which depends upon the composition and relative motion of the substances in the mixture. The theory is extended to deal with the diffusion of a mixture of fluids through a rigid solid.

The theory is applied to a number of steady-state problems involving non-Newtonian fluids including the diffusion of a fluid through a rigid plate, the laminar flow of a mixture and the flow of a mixture between rotating cylinders. The propagation of plane waves through a homogeneous mixture of viscous fluids at rest is also examined.

1. INTRODUCTION

The problem of diffusion, involving the flow relative to each other of two or more quantities in space, is an important one in the study and application of the mechanics of continua. The classical treatment of the diffusion problem, which is essentially a linear one, is based upon Fick's law, which assumes that the rate of flow of a diffusing quantity is a linear function of the space derivatives of its concentration. When this constitutive law is substituted into a relation expressing the law of conservation of the diffusing substance there results the standard diffusion equation. This equation, in its more usual applications, is concerned with the transfer of heat and of mass. Modifications of Fick's law, usually linear,

are normally introduced when coupled phenomena need to be taken into account, and the conservation law is affected by chemical reactions and other phenomena. Accounts of classical diffusion theory, with references to original theoretical and experimental work have been given by Barrer (1941), Jost (1952), Bosworth (1956), Crank (1956) and others. Linear coupled phenomena, with special reference to Onsager's relations are discussed by de Groot (1951).

In a re-examination of the problem for a mixture of fluids, Truesdell & Toupin (1960, §§ 158, 215, 295) have pointed out that in this case, since the diffusion process involves the relative motion of the constituents of the mixture, a transfer of momentum between components is involved. They suggest an approach to the formulation of constitutive equations for diffusion based on this idea. In many practical cases the omission of momentum terms from the equations describing a diffusion process is unimportant since velocities at any point of the medium are small. This is usually true, for example, in the case of a heterogeneous mixture of fluids which is at rest apart from the motion arising from diffusion, for the case of gases and liquids diffusing into solids, and of solids diffusing into each other. When a mixture of fluids is in motion, we have, in effect, a combination of two different kinds of problem. The first is that of simple fluid flow in which all constituents of the mixture are moving with the same velocity at any given point and no diffusion effects are present. The second is that of simple diffusion, in which the constituents of the mixture are moving relative to each other, but in such a manner that the mean velocity of the mixture at each point is zero. When both kinds of flow are present, momentum effects may become important and a theory is required which takes into account not only such effects but also the mechanical properties of the individual constituents. Both influences may evidently affect the resultant process in a linear or a non-linear manner. The mixing of different fluids also occurs much more rapidly under conditions of turbulent flow, but it is assumed here that this situation does not arise.

In the present paper, a theory of non-linear diffusion is developed based effectively upon the approach of Truesdell & Toupin. The assumption, usual in the treatment of mixtures, is made that each point within the medium can be occupied simultaneously by the several constituents. On a macroscopic scale, this assumption probably yields a good approximation to the actual situation which exists; in any small but finite element of volume we might expect to find some or all constituents present in a given proportion, this proportion varying continuously throughout the medium. For each constituent we assume that kinematic and mechanical quantities such as velocity, acceleration, density, stress and body force per unit mass may be defined, and that with the aid of these, equations of motion and of conservation of mass may be written down. For a mixture of n components, this procedure yields a system of $4n$ partial differential equations.

It is assumed that within the mixture, the properties of any given component are defined by means of constitutive equations relating the partial stress tensor, the density (or concentration) and the kinematic quantities, such as velocity gradients, acceleration gradients and their successive time derivatives, for that component. The procedure for formulating these equations, and for satisfying the necessary invariance requirements is identical with that for single component systems (see, for example, Green & Rivlin 1957, 1960; Pipkin & Rivlin 1960; Green & Adkins 1960).

To account for diffusion phenomena, it is assumed that the body force acting on a given component, and expressed per unit mass of that component, can be subdivided into an extraneous body force, identical in character with that for single component systems, and a diffusive force. This diffusive force depends upon the composition of the mixture at the point under consideration and upon the relative velocities of its constituents. The form of this force is restricted to satisfy the necessary requirements for invariance of the properties of the mixture under rigid body motions.

The basic theory for a mixture of fluids is formulated in Cartesian co-ordinates in §§ 2 to 4 and extended to the case where the fluids are diffusing through a rigid solid in § 5. Here, the symmetry properties of the solid affect the form of the diffusive force and results for solids which are isotropic, transversely isotropic, and aeolotropic with certain crystal symmetries are given. The relation of the present formulation to Fick's law in the classical theory of diffusion is given in § 7. A more detailed examination of classical theories of diffusion based on alternative kinematical, hydrodynamical, kinetic and thermodynamic assumptions has been made by Truesdell,[†] who has also proposed a more general linear theory.

Some problems in steady-state diffusion and the steady flow of mixtures are examined on the basis of the present theory in §§ 8 to 10. Attention is restricted to non-Newtonian and classical fluids; there is no difficulty of principle in extending the analysis to more general visco-elastic mixtures. A general feature predicted by the present theory is the tendency for the components of an initially homogeneous mixture in non-uniform flow to separate out. In the cases investigated this emerges as a specifically non-Newtonian effect.

In the final sections of the paper the propagation of plane (sound) waves through a homogeneous two-component mixture is examined. As in the case of a simple fluid, longitudinal and transverse waves may be examined separately, and attenuation and dispersion effects appear. For a two-component mixture, however, the two coupled systems of equations lead to two possible wave velocities at a given frequency for each kind of wave. When the diffusive force is small, these velocities tend towards the values for each individual fluid in the absence of the other; when the diffusive force is large, the two wave numbers are replaced by a single finite value.

To avoid undue complication, attention has been restricted in the present paper to a comparatively simple non-linear theory of diffusion based purely upon hydrodynamical considerations. Within this hydrodynamical framework a number of generalizations could be made. It would be possible, for example, to postulate that the partial stress tensor for a given fluid should be dependent not only upon the kinematical quantities defined for the fluid itself, but also upon the concentrations, velocity gradients, acceleration gradients, and time derivatives of these quantities defined, at the point under consideration, for the other fluids in the mixture. This would give an explicit coupling of mechanical properties. Again attention has here been confined to the case where the diffusive forces depend only upon the densities of the components at a given point and upon their relative velocities. A possible generalization would make these diffusive forces dependent in addition upon velocity gradients and their time derivatives and also, conceivably upon density or

[†] 'Mechanical aspects of diffusion' (1962). The author is indebted to Professor Truesdell for making available a manuscript of this paper, and for a stimulating discussion on the general problem of diffusion.

concentration gradients. Further generalizations might permit the explicit appearance of time derivatives of stress or of diffusive force in the respective constitutive equations, as, for example, occurs in the theory of hypo-elasticity (Truesdell 1955), or an expression of the stress and diffusive force as functionals of the independent kinematic quantities as suggested by Green & Rivlin (1957, 1960) in considering memory effects. Invariance considerations alone are insufficient to exclude the possibility of generalizations of this kind. Finally, thermodynamic aspects and the problem of thermal diffusion, and the possibility of chemical reactions and other effects within the mixture have been excluded at this stage.

GENERAL FORMULATION

2. NOTATION AND FORMULAE

We consider a mixture of n substances \mathcal{S}_r ($r = 1, 2, \dots, n$) which are in motion relative to each other. We make the assumption customary in treating diffusion problems that each point P within the mixture is occupied simultaneously by each of the substances \mathcal{S}_r , these substances being present in the mixture at that point in specified proportions. We further assume that to each of the diffusing substances at the point P we can assign distinct kinematic quantities, such as velocity and acceleration, and mechanical quantities such as stress and body force. We may then define mean values for the velocity and acceleration of the mixture at the point P , while the total stress[†] and extraneous body force acting on the mixture at this point is the sum of the partial stresses and body forces respectively for each of the components \mathcal{S}_r .

We refer the motion to a fixed system of rectangular Cartesian co-ordinates x_i . We suppose that a particle of the substance \mathcal{S}_r which is at y_i^\ddagger at the current time t , was at the point $x_i^{(r)}$ at initial time $t = 0$. We assume that at the point y_i at time t the substance \mathcal{S}_r has velocity \mathbf{v}_r with components $v_i^{(r)}$ relative to the x_i axes. These components are given by

$$v_i^{(r)} = {}^{(r)}Dy_i/Dt, \quad y_i = y_i(x_j^{(r)}, t), \quad (2.1)$$

where ${}^{(r)}D/Dt$ denotes differentiation with respect to t holding the co-ordinates $x_j^{(r)}$ constant.

If the density of substance \mathcal{S}_r at y_i is ρ_r , the density of the mixture is

$$\rho = \sum_{r=1}^n \rho_r \quad (2.2)$$

and the mean velocity \mathbf{v} of the mixture at this point is given by

$$\rho \mathbf{v} = \sum_{r=1}^n \rho_r \mathbf{v}_r. \quad (2.3)$$

[†] This definition of total stress differs from that used by Truesdell, who includes terms involving relative velocities. Predictions of the two theories would differ when stress boundary conditions are involved. The present theory can be made formally equivalent to Truesdell's if relative velocities are included in the constitutive equations for the stresses.

[‡] For convenience in considering the situation at a fixed point within the mixture at current time t we shall refer to the reference frame y_i , it being understood that the y_i axes are fixed and coincident with the x_i system.

We observe that if $\phi = \phi(y_i, t)$ is any (scalar or tensor) function of the co-ordinates y_i and t , and $\partial/\partial t$ denotes differentiation with respect to t holding the co-ordinates y_i constant, then

$$\frac{{}^{(r)}D\phi}{Dt} = \frac{\partial\phi}{\partial t} + v_m^{(r)} \frac{\partial\phi}{\partial y_m}, \quad (2.4)$$

where, in (2.4) and subsequently, summation is carried out over repeated indices unless otherwise indicated. If we define the operator D/Dt by

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + v_m \frac{\partial}{\partial y_m}, \quad (2.5)$$

where v_m are the components of \mathbf{v} , then from (2.2) and (2.3)

$$\rho \frac{D\phi}{Dt} = \sum_{r=1}^n \rho_r \frac{{}^{(r)}D\phi}{Dt}. \quad (2.6)$$

The diffusion velocity \mathbf{U}_r of component \mathcal{S}_r may be defined by

$$\mathbf{U}_r = \mathbf{v}_r - \mathbf{v}, \quad (2.7)$$

and by virtue of (2.2) and (2.3) this relation may be rewritten

$$\rho \mathbf{U}_r = \sum_{s=1}^n \rho_s (\mathbf{v}_r - \mathbf{v}_s). \quad (2.8)$$

From (2.8) we observe that
$$\sum_{r=1}^n \rho_r \mathbf{U}_r = \mathbf{0}. \quad (2.9)$$

If we assume that none of the substance \mathcal{S}_r is removed or generated by chemical reactions, adsorption or similar processes, the equation of continuity for this constituent takes the form

$$\frac{\partial\rho_r}{\partial t} + \frac{\partial}{\partial y_i} (\rho_r v_i^{(r)}) \equiv \frac{{}^{(r)}D\rho_r}{Dt} + \rho_r \frac{\partial v_i^{(r)}}{\partial y_i} = 0 \quad (r = 1, 2, \dots, n; r \text{ not summed}). \quad (2.10)$$

By adding the n equations (2.10) and making use of (2.2) and (2.3) we obtain for the mixture as a whole

$$\frac{\partial\rho}{\partial t} + \frac{\partial}{\partial y_i} (\rho v_i) \equiv \frac{D\rho}{Dt} + \rho \frac{\partial v_i}{\partial y_i} = 0. \quad (2.11)$$

We postulate that for each constituent \mathcal{S}_r there exists a partial stress tensor $\boldsymbol{\sigma}_r$, with components $\sigma_{ij}^{(r)}$ at y_i referred to the x_i axes, which depends only upon the concentration of \mathcal{S}_r and upon its motion. We further define a total body force vector ${}^t\mathbf{F}_r$ per unit mass of \mathcal{S}_r , which is regarded as acting only upon the component \mathcal{S}_r , and a vector \mathbf{f}_r defining the acceleration of \mathcal{S}_r at y_i . We may then suppose that the movement of each constituent \mathcal{S}_r is governed by the equation of motion

$$\frac{\partial\sigma_{ik}^{(r)}}{\partial y_k} + \rho_r {}^tF_i^{(r)} = \rho_r f_i^{(r)} \equiv \rho_r \frac{{}^{(r)}Dv_i^{(r)}}{Dt} \equiv \rho_r \left[\frac{\partial v_i^{(r)}}{\partial t} + v_m^{(r)} \frac{\partial v_i^{(r)}}{\partial y_m} \right] \quad (r = 1, 2, \dots, n; r \text{ not summed}), \quad (2.12)$$

where ${}^tF_i^{(r)}, f_i^{(r)}$ are the components of ${}^t\mathbf{F}_r, \mathbf{f}_r$, respectively referred to the y_i axes.

We assume that the effect of diffusion can be accounted for by the nature of the body force in equation (2.12). We therefore write

$${}^t\mathbf{F}_r = \mathbf{F}_r + \boldsymbol{\Psi}_r \quad (r = 1, 2, \dots, n), \quad (2.13)$$

where \mathbf{F}_r is the extraneous body force acting on the material \mathcal{S}_r and Ψ_r arises from the diffusion process, that is, from the influence of the other materials in the mixture. If we regard the quantities Ψ_r purely as interaction forces between the components of the mixture, we obtain from (2.13)

$$\left. \begin{aligned} \sum_{r=1}^n \rho_r {}^t\mathbf{F}_r &= \sum_{r=1}^n \rho_r \mathbf{F}_r = \rho \mathbf{F}, \\ \sum_{r=1}^n \rho_r \Psi_r &= \mathbf{0}, \end{aligned} \right\} \quad (2.14)$$

where \mathbf{F} is the total extraneous body force acting per unit mass of the mixture at y_i . The addition of the n equations (2.12) then yields

$$\partial \sigma_{ik} / \partial y_k + \rho F_i = \rho f_i \quad (2.15)$$

where f_i are the components of the mean acceleration vector \mathbf{f} defined by

$$\rho \mathbf{f} = \sum_{r=1}^n \rho_r \mathbf{f}_r, \quad (2.16)$$

and

$$\sigma_{ik} = \sum_{r=1}^n \sigma_{ik}^{(r)}. \quad (2.17)$$

3. CONSTITUTIVE EQUATIONS FOR FLUIDS

We assume that the mechanical properties of the fluid \mathcal{S}_r depend only upon the quantities specifying the motion of \mathcal{S}_r and its concentration in the mixture. We therefore suppose that in the constitutive equations for \mathcal{S}_r , the components of the stress tensor $\boldsymbol{\sigma}_r$ are polynomials in the velocity gradients $\partial {}^{(1)}v_i^{(r)} / \partial y_j$, acceleration gradients $\partial {}^{(2)}v_i^{(r)} / \partial y_j, \dots, (n_r - 1)$ th acceleration gradients $\partial {}^{(n_r)}v_i^{(r)} / \partial y_j$, for all possible motions, where

$$\left. \begin{aligned} {}^{(1)}v_i^{(r)} &= v_i^{(r)}, & {}^{(2)}v_i^{(r)} &= f_i^{(r)}, \\ {}^{(p)}v_i^{(r)} &= \frac{{}^{(r)}\mathbf{D}^{p-1}(v_i^{(r)})}{\mathbf{D}t^{p-1}} = \left(\frac{\partial}{\partial t} + v_m^{(r)} \frac{\partial}{\partial y_m} \right) ({}^{(p-1)}v_i^{(r)}) & (p = 2, 3, \dots, n_r). \end{aligned} \right\} \quad (3.1)$$

We further suppose that $\sigma_{ik}^{(r)}$ are continuous single-valued functions of the density ρ_r , without singularities within the range of values which needs to be considered.† We therefore write

$$\sigma_{ik}^{(r)} = f_{ik}^{(r)} \left(\rho_r; \frac{\partial v_i^{(r)}}{\partial y_k}, \frac{\partial {}^{(2)}v_i^{(r)}}{\partial y_k}, \dots, \frac{\partial {}^{(n_r)}v_i^{(r)}}{\partial y_k} \right) \quad (r = 1, 2, \dots, n). \quad (3.2)$$

It would be possible to postulate dependence of $\sigma_{ik}^{(r)}$ upon higher-order space derivatives of the velocity and accelerations ${}^{(b)}v_i^{(r)}$ and upon space derivatives of the density ρ_r . The possibility is, however, not examined here. Time derivatives ${}^{(r)}\mathbf{D}^q \rho_r / \mathbf{D}t^q$ may be excluded from (3.2) by virtue of (2.10). The velocity and acceleration components ${}^{(b)}v_i^{(r)}$ cannot occur if the stresses are to be independent of rigid body translational motions of the mixture as a whole.

If the stresses (3.2) are to be independent of rotational rigid body motions of the medium as a whole, it may be shown that $\sigma_{ij}^{(r)}$ can be expressed in the form

$$\sigma_{ij}^{(r)} = F_{ij}^{(r)}(\rho_r; {}^{(1)}A_{pq}^{(r)}, {}^{(2)}A_{pq}^{(r)}, \dots, {}^{(n_r)}A_{pq}^{(r)}) \quad (r = 1, 2, \dots, n), \quad (3.3)$$

† It is to be understood that any subsequent indication that a function is dependent upon one or more of the densities ρ_r is intended to imply, without further comment, that the function is continuous and single valued in these arguments without singularities within the relevant range of values.

where

$$\left. \begin{aligned} (1)A_{ij}^{(r)} &= A_{ij}^{(r)} = \frac{1}{2} \left(\frac{\partial v_i^{(r)}}{\partial y_j} + \frac{\partial v_j^{(r)}}{\partial y_i} \right), \\ (p)A_{ij}^{(r)} &= \frac{(r)D^{(p-1)}A_{ij}^{(r)}}{Dt} + (p-1)A_{im}^{(r)} \frac{\partial v_m^{(r)}}{\partial y_j} + (p-1)A_{mj}^{(r)} \frac{\partial v_m^{(r)}}{\partial y_i} \\ & \quad (p = 1, 2, \dots, n_r; r = 1, 2, \dots, n; r \text{ not summed}). \end{aligned} \right\} \quad (3.4)$$

In (3.3), the functions $F_{ij}^{(r)}$ are symmetric isotropic† tensor functions of the symmetric tensors $(p)A_{ij}^{(r)}$, which are polynomials in the components $(p)A_{ij}^{(r)}$ and are also functions of ρ_r . The reduction of (3.2) to the form (3.3), with references to original papers, is given in Green & Adkins (1960).

The further reduction of (3.3) to closed form follows from the work of Spencer & Rivlin (1959*a, b*, 1960) and of Spencer (1961). When $\sigma_{ij}^{(r)}$ depends only upon ρ_r and the single tensor $A_{ij}^{(r)}$, it may be expressed in closed form by the matrix equation

$$\sigma_r = \phi_0^{(r)} \mathbf{I} + \phi_1^{(r)} \mathbf{A}_r + \phi_2^{(r)} \mathbf{A}_r^2, \quad (3.5)$$

where

$$\sigma_r = \|\sigma_{ij}^{(r)}\|, \quad \mathbf{A}_r = \|A_{ij}^{(r)}\|, \quad (3.6)$$

\mathbf{I} is the unit matrix and $\phi_0^{(r)}$, $\phi_1^{(r)}$ and $\phi_2^{(r)}$ are polynomials in

$$\text{tr } \mathbf{A}_r, \quad \text{tr } \mathbf{A}_r^2, \quad \text{tr } \mathbf{A}_r^3 \quad (\text{tr } \mathbf{A} = \text{trace } \mathbf{A}), \quad (3.7)$$

with coefficients which are functions of ρ_r . For some purposes it is convenient to replace (3.5) by the form

$$\sigma_r = (-p_r + \phi_0^{(r)}) \mathbf{I} + \phi_1^{(r)} \mathbf{A}_r + \phi_2^{(r)} \mathbf{A}_r^2, \quad (3.8)$$

where p_r is a scalar function, which may depend upon ρ_r , and $\phi_0^{(r)}$, $\phi_1^{(r)}$, $\phi_2^{(r)}$ have the functional forms already indicated.

In the foregoing discussion we have assumed that the stress components $\sigma_{ij}^{(r)}$ depend only upon the quantities ρ_r , $\partial^{(p)}v_i^{(r)}/\partial y_j$ (or $(p)A_{ij}^{(r)}$) defined for the substance \mathcal{S}_r . This implies that there is no coupling, as far as the internal stresses are concerned, between the constituents of the mixture, although one might expect different functions $f_{ij}^{(r)}$ or $F_{ij}^{(r)}$ in (3.2) and (3.3) to apply to different mixtures. A more general theory could evidently be considered in which each of the stress components $\sigma_{ij}^{(r)}$ depends upon all of the arguments $\rho_1, \rho_2, \dots, \rho_n$, $\partial v_i^{(1)}/\partial y_k, \dots, \partial^{(n)}v_i^{(1)}/\partial y_k, \dots, \partial v_i^{(n)}/\partial y_k, \dots, \partial^{(n)}v_i^{(n)}/\partial y_k$. The formulation presents no difficulties of principle, and restrictions imposed by invariance considerations may be obtained by the method indicated by Green & Adkins (1960).

This kind of approach has been used by Biot (1956*a, b*) in considering the related problem of the flow of an ideal fluid through an elastic medium.

4. CONSTITUTIVE EQUATIONS FOR DIFFUSIVE FORCES

We assume that the diffusive forces Ψ_r depend upon the composition of the mixture at the point y_i and upon the relative motions of its constituents, that is, upon the densities ρ_s and upon the diffusion velocities \mathbf{U}_r .

We first observe that the differences $\mathbf{v}_r - \mathbf{v}_s$ are unaffected by a rigid body translational motion of the mixture. For if this motion has velocity \mathbf{a} , each of the velocities \mathbf{v}_r is increased

† We restrict attention to isotropic fluids. Constitutive equations for anisotropic fluids have been proposed by Ericksen (1960*a, b*).

to $\mathbf{v}_r + \mathbf{a}$ and their differences are unaltered. From (2.8) we see that this also implies that the quantities \mathbf{U}_r are unchanged.

We now consider a motion of the medium which differs from the actual motion at time t only to the extent of an arbitrary rigid body rotation. The co-ordinates \bar{y}_i of a typical particle P , which in the original motion is at y_i , are therefore given by

$$\bar{y}_i = M_{ij} y_j, \quad (4.1)$$

where $M_{ij} = M_{ij}(t)$ are a set of continuous functions of t defining the rigid body rotation and are therefore subject to the conditions

$$M_{ir} M_{jr} = M_{ri} M_{rj} = \delta_{ij}, \quad |M_{ij}| = 1. \quad (4.2)$$

For the substance \mathcal{S}_r , we may write

$$\bar{y}_i = \bar{y}_i(x_j^{(r)}, t), \quad (4.3)$$

$$\bar{v}_i^{(r)} = \frac{{}^{(r)}D\bar{y}_i}{Dt} = \frac{{}^{(r)}D}{Dt} (M_{ij} y_j) = M_{ij} v_j^{(r)} + y_j \frac{{}^{(r)}DM_{ij}}{Dt}. \quad (4.4)$$

A similar relation holds for $\bar{v}_i^{(s)}$, with s substituted for r ($r \neq s$). Moreover, since the quantities M_{ij} are functions only of t , independent of $x_i^{(r)}$, $x_i^{(s)}$ (or y_i), ${}^{(r)}DM_{ij}/Dt = {}^{(s)}DM_{ij}/Dt$ and

$$\bar{v}_i^{(r)} - \bar{v}_i^{(s)} = M_{ij} (v_j^{(r)} - v_j^{(s)}) \quad (r, s = 1, 2, \dots, n; r \neq s). \quad (4.5)$$

Similarly, for the mean velocity $\bar{\mathbf{v}}$ we have

$$\begin{aligned} \rho \bar{v}_i &= \sum_{r=1}^n \rho_r \bar{v}_i^{(r)} = \sum_{r=1}^n \rho_r \frac{{}^{(r)}D\bar{y}_i}{Dt} \\ &= \sum_{r=1}^n \rho_r \left[M_{ik} \frac{{}^{(r)}Dy_k}{Dt} + y_k \frac{{}^{(r)}DM_{ik}}{Dt} \right], \end{aligned} \quad (4.6)$$

and remembering (2.2), (2.3) and (2.6) we obtain

$$\bar{v}_i^{(r)} - \bar{v}_i = M_{ik} (v_k^{(r)} - v_k). \quad (4.7)$$

The differences $v_i^{(r)} - v_i^{(s)}$, $v_i^{(r)} - v_i$ thus behave as the components of vectors which are independent of rigid body motions of the mixture as a whole. We may therefore postulate the constitutive law

$$\Psi_r = \Psi_r(\rho_s, \mathbf{U}_t) \quad (r, s, t = 1, 2, \dots, n), \quad (4.8)$$

or

$$\Psi_r = \Psi_r(\rho_s, \mathbf{v}_p - \mathbf{v}_q) \quad (r, s, p, q = 1, 2, \dots, n), \quad (4.9)$$

for all possible motions. In (4.8) and (4.9) Ψ_r are vector functions of the arguments indicated which we suppose to be polynomials in the components of \mathbf{U}_t or of $\mathbf{v}_p - \mathbf{v}_q$. The functional forms of Ψ_r for a given system are in general different in (4.8) and (4.9).

The formulations (4.8) and (4.9) are symmetrical in the relative velocities \mathbf{U}_t or $\mathbf{v}_p - \mathbf{v}_q$, but we see from (2.8) and (2.9) that these velocities are not all linearly independent. To eliminate redundancies in the elements we may note that

$$\mathbf{v}_p - \mathbf{v}_q = (\mathbf{v}_p - \mathbf{v}_n) - (\mathbf{v}_q - \mathbf{v}_n), \quad (4.10)$$

and write

$$\Psi_r = \Psi_r(\rho_s, \mathbf{u}_t) \quad (r, s = 1, 2, \dots, n; t = 1, 2, \dots, n-1), \quad (4.11)$$

where

$$\mathbf{u}_t = \mathbf{v}_t - \mathbf{v}_n, \quad (4.12)$$

and we again regard Ψ_r as a vector polynomial function of the components $u_i^{(s)}$ of the velocities \mathbf{u}_s .

We may readily infer that the functional forms (4.8), (4.9) and (4.11) are those appropriate to mixtures of isotropic materials. We consider explicitly the form (4.11), and to deduce the isotropic property and derive constitutive equations in closed form, it is convenient to use the technique employed by Pipkin & Rivlin (1960) and Adkins (1960*a, b*). We define an arbitrary vector \mathbf{q} with components q_i referred to the system y_i and form the scalar products

$$\Phi^{(r)} = \mathbf{q} \cdot \Psi_r = q_i \Psi_i^{(r)} = \Phi^{(r)}(q_i; \rho_s, u_k^{(t)}) \quad (r, s = 1, 2, \dots, n; t = 1, 2, \dots, n-1). \quad (4.13)$$

The functions $\Phi^{(r)}$ are linear and homogeneous in the components of \mathbf{q} and the components $\Psi_i^{(r)}$ are given uniquely by

$$\Psi_i^{(r)} = \partial \Phi^{(r)} / \partial q_i. \quad (4.14)$$

If the properties of the mixture are unaffected by superposed rigid body motions, the functions $\Phi^{(r)}$ must be invariant under all transformations of the type specified by (4.1) and (4.2). This implies that

$$\Phi^{(r)}(q_i; \rho_s, u_k^{(t)}) = \Phi^{(r)}(\bar{q}_i; \rho_s, \bar{u}_k^{(t)}), \quad (4.15)$$

where

$$\bar{q}_i = M_{ik} q_k, \quad \bar{u}_i^{(t)} = M_{ik} u_k^{(t)}, \quad (4.16)$$

and M_{ik} satisfy (4.2). The conditions (4.15) imply that the functions (4.13) are isotropic functions of q_i , $u_k^{(t)}$ and that the forms (4.11) for Ψ_r are those appropriate to mixtures of isotropic materials.

If each component of the mixture is isotropic with a centre of symmetry, each of the functions $\Phi^{(r)}$ satisfies an invariance requirement of the form (4.15), but the quantities M_{ik} in (4.16) now satisfy the conditions

$$M_{ik} M_{jk} = M_{ki} M_{kj} = \delta_{ij}, \quad |M_{ik}| = \pm 1. \quad (4.17)$$

Since $\Phi^{(r)}$ is a polynomial in the elements q_i , $u_i^{(t)}$ it follows (Weyl 1946) that it can be expressed as a polynomial in the scalar products

$$\mathbf{u}_s \cdot \mathbf{u}_t \quad (\text{i}), \quad \mathbf{u}_s \cdot \mathbf{q} \quad (\text{ii}) \quad (s, t = 1, 2, \dots, n-1), \quad (4.18)$$

which is linear and homogeneous in the invariants (ii). From (4.14) we infer that

$$\Psi_i^{(r)} = \sum_{s=1}^{n-1} u_i^{(s)} \phi_{(s)}^{(r)} \quad (r = 1, 2, \dots, n), \quad (4.19)$$

where $\phi_{(s)}^{(r)}$ are polynomials in the invariants (4.18) (i) which are continuous functions of the densities ρ_s .

More generally we might suppose that Ψ_r depends upon relative accelerations of the constituents at any point and upon higher-order time derivatives of \mathbf{U}_i or \mathbf{u}_i . Yet another extension of the theory would follow from the assumption that Ψ_r may depend upon the rate at which the constituents in the mixture are deforming, that is, upon the velocity gradients $\partial v_i^{(s)} / \partial y_j$ and upon the successive acceleration gradients $\partial^{(b)} v_i^{(s)} / \partial y_j$. The manner in which relative accelerations or the velocity and acceleration gradients can occur in the functions Ψ_r must be restricted by invariance considerations and a method for obtaining permissible functional combinations of these quantities has been indicated by Green & Adkins (1960).

5. DIFFUSION OF A SYSTEM OF FLUIDS THROUGH A RIGID SOLID

In considering the diffusion of a fluid or a mixture of fluids into a solid medium, we might expect the diffusing forces Ψ_r to produce an effect in deforming the solid medium itself. This effect is often likely to be small, however, and in the classical treatment of many diffusion problems the assumption is made that the solid may be regarded as rigid (see, for example, Crank 1956). If we make this assumption in the present instance, a theory of diffusion into solids may be formulated which is based on the results of §§ 2 to 4.

For the substances $\mathcal{S}_1, \dots, \mathcal{S}_n$, equations (2.1) to (2.6) and the field equations (2.10) to (2.12), with the body forces ${}^t\mathbf{F}_r$ given by (2.13), continue to apply. The relations (2.14), however, are no longer valid, since the solid may be regarded as exerting a resultant diffusive force on the fluid mixture.

In considering the diffusive forces, we may regard the rigid solid as the $(n+1)$ th material $\mathcal{S}_{n+1} = \mathcal{S}$ of the diffusing system. For this material, when the solid is at rest, we have

$$x_i^{(n+1)} = y_i, \quad \mathbf{v}_{n+1} = \mathbf{0}. \quad (5.1)$$

The diffusive forces Ψ_r are then regarded as functions of the diffusion velocities

$$\mathbf{v}_t - \mathbf{v}_{n+1} = \mathbf{v}_t \quad (t = 1, 2, \dots, n) \quad (5.2)$$

relative to the solid \mathcal{S} .

We examine the situation in which the solid \mathcal{S} may be aeolotropic and choose three mutually orthogonal unit vectors $\mathbf{h}_1, \mathbf{h}_2, \mathbf{h}_3$, fixed in \mathcal{S} , to specify its orientation. The diffusive forces Ψ_r may then be regarded as polynomial functions of the diffusion velocities (5.2) and the vectors \mathbf{h}_k , these forces depending also upon the densities ρ_s . We therefore write

$$\Psi_r = \Psi_r(\rho_s, \mathbf{v}_t, \mathbf{h}_k) \quad (r, s, t = 1, 2, \dots, n) \quad (5.3)$$

and the scalar functions $\Phi^{(r)}$ defined by (4.13) now take the form

$$\Phi^{(r)} = \Phi^{(r)}(\mathbf{q}; \rho_s, \mathbf{v}_t, \mathbf{h}_k) \quad (r, s, t = 1, 2, \dots, n).$$

If the system is to be unaffected by the rigid body motions specified by (4.1) and (4.2), the functions $\Phi^{(r)}$ must be isotropic functions of their arguments and may be expressed as polynomials in the scalar products and scalar triple products formed from the vectors $\mathbf{q}, \mathbf{v}_t, \mathbf{h}_k$ (Weyl 1946), which are linear and homogeneous in the components of \mathbf{q} . We may without loss of generality, choose the x_i axes so that the vectors \mathbf{h}_k lie along them, and then

$$\mathbf{h}_1 = (1, 0, 0), \quad \mathbf{h}_2 = (0, 1, 0), \quad \mathbf{h}_3 = (0, 0, 1).$$

In this case

$$\mathbf{h}_k \cdot \mathbf{v}^{(t)} = v_k^{(t)}$$

and the functions $\Phi^{(r)}$ may be written

$$\Phi^{(r)} = \Phi^{(r)}(q_i; \rho_s, v_k^{(t)}) \quad (r, s, t = 1, 2, \dots, n). \quad (5.4)$$

In the present instance, the functions $\Phi^{(r)}$ are not necessarily isotropic functions of $q_i, v^{(t)}$; their form is governed by the symmetry properties of the solid \mathcal{S} .† If some or all of the constituents $\mathcal{S}_1, \mathcal{S}_2, \dots, \mathcal{S}_n$ are aeolotropic, we may expect that their properties will also affect the forms of the functions $\Phi^{(r)}$ and this poses a joint invariance problem. We

† In effect, the space through which the fluids are flowing need no longer (as in §4) be isotropic.

restrict attention here to the case where each of the diffusing substances is isotropic with a centre of symmetry and in this case only the nature of the solid \mathcal{S} need be taken into account in determining the form of $\Phi^{(r)}$. We suppose the solid \mathcal{S} to be oriented as to that the vectors \mathbf{h}_k (or the x_i axes) coincide with the principal crystal axes. The invariance condition for the functions $\Phi^{(r)}$ may therefore be written

$$\Phi^{(r)} = \Phi^{(r)}(q_i; \rho_s, v_k^{(t)}) = \Phi^{(r)}(\bar{q}_i; \rho_s, \bar{v}_k^{(t)}), \quad (5.5)$$

where, as before

$$\bar{q}_i = M_{ik} q_k, \quad \bar{v}_i^{(t)} = M_{ik} v_k^{(t)}, \quad (5.6)$$

but M_{ik} belong to the appropriate subgroup \mathcal{M} of the full orthogonal group defined by (4.17).

If the quantities I_p ($p = 1, 2, \dots, N$) form an integrity basis for the invariants of the system of vectors \mathbf{v}_r under all transformations of the subgroup \mathcal{M} and $Q_{p'}$ ($p' = 1, 2, \dots, N'$) form the corresponding system of invariants of \mathbf{v}_r and \mathbf{q} which are linear in the components q_i , then we may write

$$\Phi^{(r)} = \Phi^{(r)}(Q_{p'}, I_p) = \sum_{p'=1}^{N'} Q_{p'} \phi_{p'}^{(r)}, \quad (5.7)$$

$$\Psi_i^{(r)} = \sum_{p'=1}^{N'} \frac{\partial Q_{p'}}{\partial q_i} \phi_{p'}^{(r)}, \quad \phi_{p'}^{(r)} = \phi_{p'}^{(r)}(I_p), \quad (5.8)$$

where $\Phi^{(r)}$ and $\phi_{p'}^{(r)}$ are scalar polynomials in the arguments indicated which are dependent also upon ρ_s .

The determination of the invariants for a given subgroup \mathcal{M} may be carried out using the techniques of classical invariant theory. We list here some of the more important cases.

(i) *Hemihedral isotropic materials.* The group \mathcal{M} describing the symmetry of the solid \mathcal{S} is the proper orthogonal group and the invariants I_p in (5.7) and (5.8) are (Weyl 1956)

$$\mathbf{v}_r \cdot \mathbf{v}_s \quad (\text{i}), \quad [\mathbf{v}_r \mathbf{v}_s \mathbf{v}_t] \quad (r \neq s \neq t \neq r) \quad (\text{ii}), \quad (5.9)$$

In this case, we obtain from (5.8)

$$\Psi_i^{(r)} = \sum_{s=1}^n v_i^{(s)} \phi_{(s)}^{(r)} + \sum_{s=1}^{n-1} \sum_{t=s+1}^n \epsilon_{ijk} v_j^{(s)} v_k^{(t)} \phi_{(st)}^{(r)}, \quad (5.10)$$

where $\phi_{(s)}^{(r)}$, $\phi_{(st)}^{(r)}$ are polynomials in the invariants (5.9) and ϵ_{ijk} is the alternating tensor.

(ii) *Holohedral isotropic materials.* Here the group \mathcal{M} is the full orthogonal group and the invariants I_p are given by (5.9) (i). In this case the diffusive forces $\Psi_i^{(r)}$ take the form

$$\Psi_i^{(r)} = \sum_{s=1}^n v_i^{(s)} \phi_{(s)}^{(r)}, \quad (5.11)$$

$\phi_{(s)}^{(r)}$ being polynomials in the invariants (5.9) (i).

(iii) *Transversely isotropic bodies.* If the solid \mathcal{S} is transversely isotropic with respect to the y_1 (or x_1) direction, the functions $\Phi^{(r)}$ are form invariant under the transformation

$$\bar{y}_1 = y_1, \quad \bar{y}_\alpha = M_{\alpha\beta} y_\beta \quad (\alpha, \beta = 2, 3), \quad (5.12)$$

$$\text{where} \quad M_{\alpha\gamma} M_{\beta\gamma} = M_{\gamma\alpha} M_{\gamma\beta} = \delta_{\alpha\beta}, \quad |M_{\alpha\beta}| = 1 \quad (\alpha, \beta, \gamma = 2, 3), \quad (5.13)$$

and also under the transformation

$$(\bar{y}_1, \bar{y}_2, \bar{y}_3) = (y_1, y_2, -y_3). \quad (5.14)$$

The invariants I_p are then (Adkins 1960*b*)

$$v_1^{(r)}, \quad \mathbf{v}_r \cdot \mathbf{v}_s, \quad (5.15)$$

and the functions $\Psi_i^{(r)}$ take the form

$$\Psi_i^{(r)} = \sum_{s=1}^n v_i^{(s)} \phi_{(s)}^{(r)} + \delta_{i1} \phi^{(r)}, \quad (5.16)$$

where $\phi_{(s)}^{(r)}, \phi^{(r)}$ are polynomials in the invariants (5.15).

(iv) *Triclinic system*. The triclinic system contains two crystal classes. In the *pedial* class the functions $\Phi^{(r)}$ are form-invariant only under the identity transformation, and the diffusive forces Ψ_r are then general vector polynomial functions of the components $v_i^{(s)}$. In the *pinacoidal* class, the functions $\Phi^{(r)}$ are form invariant under the central inversion $\bar{y}_i = -y_i$, the invariants I_p are the products

$$v_j^{(s)} v_k^{(t)} \quad (j, k = 1, 2, 3; s, t = 1, 2, \dots, n), \quad (5.17)$$

and the components $\Psi_i^{(r)}$ take the forms

$$\Psi_i^{(r)} = \sum_{s=1}^n v_k^{(s)} \phi_{(s)ik}^{(r)}, \quad (5.18)$$

where $\phi_{(s)ik}^{(r)}$ are polynomial tensor functions of the invariants (5.17).

(v) *Rhombic (orthotropic) system*. This system contains three classes. To examine these, we define the transformations

$$\left. \begin{aligned} \mathbf{R}_1: (\bar{y}_1, \bar{y}_2, \bar{y}_3) &= (-y_1, y_2, y_3), \\ \mathbf{D}_1: (\bar{y}_1, \bar{y}_2, \bar{y}_3) &= (y_1, -y_2, -y_3), \end{aligned} \right\} \quad (5.19)$$

and denote by $(\mathbf{R}_2, \mathbf{D}_2)$, $(\mathbf{R}_3, \mathbf{D}_3)$ the transformations obtained by replacing $(1, 2, 3)$ by $(2, 3, 1)$ and $(3, 1, 2)$ respectively in (5.19).

The transformations characterizing the *rhombic-pyramidal* class are $\mathbf{R}_2, \mathbf{R}_3, \mathbf{D}_1$. For this class, the invariants I_p are

$$v_1^{(s)}, \quad v_2^{(s)} v_2^{(t)}, \quad v_3^{(s)} v_3^{(t)} \quad (s, t = 1, 2, \dots, n), \quad (5.20)$$

and the diffusive force components $\Psi_i^{(r)}$ take the form

$$\Psi_i^{(r)} = \delta_{i1} \phi^{(r)} + \sum_{s=1}^n \delta_{i2} v_2^{(s)} \phi_{(s)}^{(r)} + \sum_{s=1}^n \delta_{i3} v_3^{(s)} \Phi_{(s)}^{(r)}, \quad (5.21)$$

where the functions $\phi^{(r)}, \phi_{(s)}^{(r)}$ and $\Phi_{(s)}^{(r)}$ are polynomials in the invariants (5.20).

For the *rhombic-disphenoidal* system, the functions $\Phi^{(r)}$ are form invariant under the transformations $\mathbf{D}_1, \mathbf{D}_2, \mathbf{D}_3$. The invariants I_p are then

$$v_1^{(s)} v_1^{(t)}, \quad v_2^{(s)} v_2^{(t)}, \quad v_3^{(s)} v_3^{(t)}, \quad v_1^{(s)} v_2^{(t)} v_3^{(u)} \quad (s, t, u = 1, 2, \dots, n) \quad (5.22)$$

and the components $\Psi_i^{(r)}$ become

$$\begin{aligned} \Psi_i^{(r)} &= \sum_{s=1}^n \{ \delta_{i1} v_1^{(s)} \phi_{(1s)}^{(r)} + \delta_{i2} v_2^{(s)} \phi_{(2s)}^{(r)} + \delta_{i3} v_3^{(s)} \phi_{(3s)}^{(r)} \} \\ &\quad + \sum_{s=1}^n \sum_{t=1}^n \{ \delta_{i1} v_2^{(s)} v_3^{(t)} \phi_{(1st)}^{(r)} + \delta_{i2} v_3^{(s)} v_1^{(t)} \phi_{(2st)}^{(r)} + \delta_{i3} v_1^{(s)} v_2^{(t)} \phi_{(3st)}^{(r)} \} \quad (r = 1, 2, \dots, n), \end{aligned} \quad (5.23)$$

where the functions $\phi_{(ks)}^{(r)}, \phi_{(kst)}^{(r)}$ are polynomials in the invariants (5.22).

In the case of the *rhombic-dipyramidal* system, the transformations characterizing the symmetry properties are $\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \mathbf{D}_1, \mathbf{D}_2, \mathbf{D}_3$, together with the central inversion $\bar{y}_i = -y_i$; the invariants are

$$v_1^{(s)}v_1^{(t)}, \quad v_2^{(s)}v_2^{(t)}, \quad v_3^{(s)}v_3^{(t)} \quad (s, t = 1, 2, \dots, n), \quad (5.24)$$

and for the components $\Psi_i^{(r)}$ we have

$$\Psi_i^{(r)} = \sum_{s=1}^n \{ \delta_{i1} v_1^{(s)} \phi_{(1s)}^{(r)} + \delta_{i2} v_2^{(s)} \phi_{(2s)}^{(r)} + \delta_{i3} v_3^{(s)} \phi_{(3s)}^{(r)} \} \quad (r = 1, 2, \dots, n), \quad (5.25)$$

where $\phi_{(ks)}^{(r)}$ are now polynomials in the quantities (5.24).

These results for the triclinic and rhombic crystal classes may be obtained by the methods developed by Smith & Rivlin (1958) and given in Green & Adkins (1960). Other crystal classes may be examined in a similar manner. The results of an extensive investigation of the invariants for a vector and symmetric second-order tensor under the transformations characterizing the crystal classes are given by Smith, Smith & Rivlin (1963).

In the present section attention has been confined to the forms for the diffusive forces Ψ_r . For inviscid fluids (including gases) it is reasonable to suppose that the isotropic forms for the stresses are unaffected by the presence of an aeolotropic solid \mathcal{S} . In the case of viscous fluids, however, it is conceivable that the stress-deformation relations would become aeolotropic during diffusion through the solid \mathcal{S} . This implies a coupling between the properties of \mathcal{S} and the fluids \mathcal{S}_r which requires further investigation and is not considered here.

6. CURVILINEAR CO-ORDINATES

Formulae in curvilinear co-ordinates are readily derived by tensor transformations. We quote here the main results for reference later.

We choose a fixed system of curvilinear co-ordinates ξ^i defined by

$$\xi^i = \xi^i(y_k), \quad \xi^i = \xi^i(x_k^{(r)}, t). \quad (6.1)$$

The operators ${}^{(r)}\mathbf{D}/\mathbf{D}t$, $\mathbf{D}/\mathbf{D}t$ given in (2.4) and (2.5) then become

$$\frac{{}^{(r)}\mathbf{D}\phi}{\mathbf{D}t} = \frac{\partial\phi}{\partial t} + w^{(r)m}\phi_{,m}, \quad \frac{\mathbf{D}\phi}{\mathbf{D}t} = \frac{\partial\phi}{\partial t} + w^m\phi_{,m}, \quad (6.2)$$

where $w^{(r)m}$, w^m are the contravariant components of the vectors \mathbf{v}_r , \mathbf{v} , respectively, referred to the system ξ^i , and the comma denotes covariant differentiation with respect to ξ^i .

The contravariant components $\tau^{(r)ij}$ of the stress tensor σ_r referred to the system ξ^i are obtained by the usual tensor transformation

$$\tau^{(r)ij} = \frac{\partial\xi^i}{\partial y_k} \frac{\partial\xi^j}{\partial y_l} \sigma_{kl}^{(r)}, \quad (6.3)$$

and the equations of continuity (2.10) and (2.11) and of motion (2.12) are replaced by

$$\left. \begin{aligned} \partial\rho_r/\partial t + (\rho_r w^{(r)m})_{,m} &= 0, \\ \partial\rho/\partial t + (\rho w^m)_{,m} &= 0, \end{aligned} \right\} \quad (6.4)$$

and $\tau^{(r)ik}_{,k} + \rho_r({}^{(r)}F^i + {}^{(r)}\Psi^i) = \rho_r(\partial w^{(r)i}/\partial t + w^{(r)m}w^{(r)i}_{,m})$ (r not summed), (6.5)

respectively. In (6.5) ${}^{(r)}F^i$, ${}^{(r)}\Psi^i$ are the contravariant components of the corresponding extraneous and diffusion body forces \mathbf{F}_r , Ψ_r acting on the component \mathcal{S}_r .

The relations (3·3), (3·4) for $\sigma_{ij}^{(r)}$ remain unchanged, and $\tau^{(r)ij}$ is given by (6·3). For an isotropic non-Newtonian fluid, however, (3·8) may be replaced by

$$\tau^{(r)ij} = (-p_r + \phi_0^{(r)}) G^{ij} + \phi_1^{(r)} d^{(r)ij} + \phi_2^{(r)} d^{(r)i}_k d^{(r)kj} \quad (r \text{ not summed}), \quad (6\cdot6)$$

where G^{ij} is the contravariant metric tensor for the system ξ^i ,

$$d^{(r)i}_j = G^{ik} d^{(r)}_{kj}, \quad d^{(r)ij} = G^{ik} d^{(r)j}_k, \quad d^{(r)}_{ij} = \frac{1}{2}(w^{(r)}_{i,j} + w^{(r)}_{j,i}) \quad (w^{(r)i} = G^{ik} w^{(r)}_k), \quad (6\cdot7)$$

and $\phi_0^{(r)}$, $\phi_1^{(r)}$, $\phi_2^{(r)}$ are functions of ρ_r which are polynomials in the invariants

$$d^{(r)i}_i, \quad d^{(r)i}_k d^{(r)k}_i, \quad d^{(r)i}_k d^{(r)k}_l d^{(r)l}_i \quad (r \text{ not summed}). \quad (6\cdot8)$$

For a mixture of fluids, the contravariant components of the diffusion forces $\Psi_{,i}$, may, from (4·19) and (4·12) be written

$${}^{(r)}\Psi_i = \sum_{s=1}^{n-1} (w^{(s)i} - w^{(n)i}) \phi_{(s)}^{(r)}, \quad (6\cdot9)$$

where $\phi_{(s)}^{(r)}$ are scalar polynomials in the invariants (4·18) (i).

Corresponding results for a system of fluids diffusing through a rigid solid are derived from those of §5 by tensor transformations. When the solid is isotropic and holohedral, equations (5·11) are replaced by

$${}^{(r)}\Psi_i = \sum_{s=1}^n w^{(s)i} \phi_{(s)}^{(r)}, \quad (6\cdot10)$$

where $\phi_{(s)}^{(r)}$ are polynomials in the invariants (5·9) (i).

7. RELATION TO CLASSICAL THEORY: FICK'S LAW

For two-component mixtures, and for a single fluid diffusing into a rigid solid, we may, with special assumptions, derive from the foregoing theory the classical diffusion equations based on Fick's law.

In the case of a two-component mixture we make the following assumptions:

(i) The diffusing substances are perfect fluids with a similar linear pressure-density relation.

(ii) The velocity and acceleration components are sufficiently small for the inertia terms in the equations of motion and for products of velocities in the expressions for the diffusive forces to be neglected.

(iii) The total density $\rho = \rho_1 + \rho_2$ of the mixture is constant.

(iv) Extraneous body forces are absent.

From the assumptions (ii) and (iv), the equations of motion (2·12) become

$$\partial \sigma_{ik}^{(\mu)} / \partial y_k + \rho_\mu \Psi_i^{(\mu)} = 0 \quad (\mu = 1, 2; \mu \text{ not summed}). \quad (7\cdot1)$$

The assumption (i) implies that

$$\sigma_{ik}^{(\mu)} = -p_\mu \delta_{ik}, \quad p_\mu = k \rho_\mu \quad (\mu = 1, 2), \quad (7\cdot2)$$

where k is a constant and p_μ are the fluid pressures for the two components.

If we take $\mathbf{v}_n = \mathbf{v}_2$ in (4·12) so that

$$\mathbf{u}_1 = \mathbf{v}_1 - \mathbf{v}_2 = \mathbf{u} \quad (\text{say}), \quad \mathbf{u}_2 = \mathbf{0}, \quad (7\cdot3)$$

and make use of (2.14), we have

$$\rho_1 \Psi_1 = -\rho_2 \Psi_2 = \mathbf{u}\alpha, \quad (7.4)$$

where α is a scalar function of ρ_1 and ρ_2 . Also, from (2.3), (2.7) and (7.3), the diffusion velocities \mathbf{U}_1 and \mathbf{U}_2 are given by

$$\mathbf{U}_1/\rho_2 = -\mathbf{U}_2/\rho_1 = \mathbf{u}/\rho. \quad (7.5)$$

If we now choose

$$\alpha = -k\rho_1\rho_2/(D\rho), \quad (7.6)$$

where D is a function of ρ_1, ρ_2 , and write

$$c_v = \rho_v/\rho, \quad (v = 1, 2), \quad (7.7)$$

we obtain, by combining (7.4) to (7.7) with (7.1) and (7.2) and using assumption (iii), Fick's law

$$c_1 \mathbf{U}_1 = -D \text{grad } c_1, \quad c_2 \mathbf{U}_2 = -D \text{grad } c_2, \quad (7.8)$$

connecting the concentrations c_μ and the diffusion velocities \mathbf{U}_μ .

If we made use of the equations of continuity (2.10) and (2.11) we find that

$$\begin{aligned} \frac{Dc_1}{Dt} &= \frac{\partial}{\partial t} \left(\frac{\rho_1}{\rho} \right) + v_k \frac{\partial}{\partial y_k} \left(\frac{\rho_1}{\rho} \right) = \frac{1}{\rho} \left[\frac{\partial \rho_1}{\partial t} + \frac{\partial}{\partial y_k} (\rho_1 v_k) \right] \\ &= -\frac{1}{\rho} \frac{\partial}{\partial y_k} [\rho_1 (v_k^{(1)} - v_k)] = -\frac{1}{\rho} \text{div} (\rho_1 \mathbf{U}_1), \end{aligned} \quad (7.9)$$

and this relation, with the first of (7.8) yields the standard diffusion equation

$$Dc_1/Dt = \text{div} (D \text{grad } c_1). \quad (7.10)$$

A corresponding equation holds for c_2 .

In the case of a single fluid of density ρ diffusing into an anisotropic solid with velocity \mathbf{v} , the components of the diffusive force Ψ referred to Cartesian co-ordinates y_i , may be written as

$$\rho \Psi_i = \alpha_{ij} v_j, \quad (7.11)$$

where α_{ij} is a tensor function of ρ and v_j . Equations analogous to (7.1), (7.2) then yield

$$-k(\partial\rho/\partial y_i) + \alpha_{ij} v_j = 0, \quad (7.12)$$

where k is a constant. As a special case of (7.12) we have the anisotropic form of Fick's law

$$\rho v_i = -D_{il}(\partial\rho/\partial y_l), \quad (7.13)$$

where D_{il} is a tensor function of ρ , and α_{ij} is chosen in (7.12) so that

$$\alpha_{ij} D_{il} = -k\rho \delta_{jl}, \quad (7.14)$$

δ_{jl} being the Kronecker delta. From (7.13) and the equation of continuity (2.11) we obtain immediately the anisotropic diffusion equation

$$\frac{\partial\rho}{\partial t} = \frac{\partial}{\partial y_l} \left(D_{il} \frac{\partial\rho}{\partial y_l} \right). \quad (7.15)$$

In (7.10) and (7.15) all velocity components are regarded as being small so that to the first order of small quantities, the operators D/Dt and $\partial/\partial t$ are equivalent.

In the classical case, we observe from (7.6) to (7.8) that a large value of α corresponds to a small value of D , and therefore to a slow rate of diffusion. Conversely, a small value of α corresponds to a rapid rate of diffusion. In the non-linear theory this corresponds to the situation where the diffusive forces Ψ_r are small and the equations of motion for the individual components are loosely coupled. As we might expect, each constituent then moves freely with comparatively little interference from the others. We note further that in (7.8) D is positive so that from (7.6) α is negative. In the general theory, we should expect the diffusive force Ψ_r to oppose the motion of the constituent \mathcal{S}_r . In this case, the quantities $\Psi_i^{(r)}$ become negative definite functions of their arguments.

STEADY-STATE SOLUTIONS

8. STEADY-STATE DIFFUSION THROUGH A RIGID PLATE

We consider the steady-state diffusion of a fluid through a holohedral isotropic rigid plate or slab, bounded in the Cartesian system x_i (or y_i) by the faces $y_1 = a_1$, $y_1 = a_2$ ($a_1 > a_2 \geq 0$). For the fluid we assume a constitutive equation of the form (3.5) and we suppose the flow to take place in the y_1 direction. We therefore write

$$(v_1, v_2, v_3) = (v, 0, 0), \quad v = v(y_1) \quad (v_i^{(1)} = v_i), \quad (8.1)$$

and in this case, writing $A_{ij}^{(1)} = A_{ij}$ we have

$$A_{11} = v', \quad A_{22} = A_{33} = A_{12} = A_{23} = A_{31} = 0, \quad (8.2)$$

where a prime denotes differentiation with respect to y_1 .

From (5.9) and (5.11) the components Ψ_i of the body force vector $\Psi_1 = \Psi$ take the form

$$(\Psi_1, \Psi_2, \Psi_3) = (v\alpha, 0, 0), \quad (8.3)$$

where α is a scalar polynomial in v^2 which is also a function of the density ρ at the point y_i .

The stress components σ_{ij} for the fluid are given from (3.5) and (8.2) by

$$\left. \begin{aligned} \sigma_{11} = \phi_0 + \phi_1 v' + \phi_2 v'^2, \quad \sigma_{22} = \sigma_{33} = \phi_0, \\ \sigma_{12} = \sigma_{23} = \sigma_{31} = 0. \end{aligned} \right\} \quad (8.4)$$

where ϕ_0, ϕ_1, ϕ_2 are functions of ρ and polynomials in v' .

In the absence of extraneous body forces the equations of motion (2.12) therefore yield

$$\partial(\phi_0 + \phi_1 v' + \phi_2 v'^2)/\partial y_1 + \rho v \alpha = \rho v v', \quad (8.5)$$

the second and third equations being satisfied identically if we assume ρ to be a function only of y_1 . The equation of continuity (2.10) yields

$$\partial(\rho v)/\partial y_1 = 0 \quad \text{or} \quad \rho v = k, \quad (8.6)$$

where k is a constant. Combining this with (8.5) we obtain

$$\partial(\phi_0 + \phi_1 v' + \phi_2 v'^2)/\partial y_1 + k \alpha = k v'. \quad (8.7)$$

When ρ is eliminated from ϕ_0, ϕ_1 and ϕ_2 by means of (8.6), (8.7) becomes a second order differential equation for the determination of v . If α is a constant, a first integral of this equation may be obtained immediately in the form

$$\phi_0 + \phi_1 v' + \phi_2 v'^2 + k \alpha y_1 = k v + k', \quad (8.8)$$

where k' is a further constant of integration.

To make further progress we require a knowledge of the functional forms of ϕ_0 , ϕ_1 and ϕ_2 in terms of ρ and v' . In the classical case (see, for example, Crank 1956), ϕ_1 and ϕ_2 are taken as zero, the inertia term kv' is neglected in (8.7) and

$$\phi_0 = A\rho = kA/v, \quad (8.9)$$

where A is a constant. This classical approximation assumes that k is small in comparison with unity and with α and that $v = O(k)$. The term kv is then small in comparison with the remaining terms in (8.8) and for a first approximation to ρ we have the linear form

$$\rho = (k' - k\alpha y_1)/A \quad (k' - k\alpha y_1 \neq 0). \quad (8.10)$$

The small velocity v is given, to a first approximation, by

$$v = kA/(k' - k\alpha y_1) \quad (k' - k\alpha y_1 \neq 0). \quad (8.11)$$

If the small term kv in (8.8) is taken into account a second approximation to ρ and v based upon (8.10) and (8.11) is readily obtained in the form

$$\left. \begin{aligned} \rho &= \frac{k' - k\alpha y_1}{A} \left[1 + \frac{k^2 A}{(k' - k\alpha y_1)^2} \right] + O(k^4), \\ v &= \frac{kA}{k' - k\alpha y_1} \left[1 - \frac{k^2 A}{(k' - k\alpha y_1)^2} \right] + O(k^5). \end{aligned} \right\} \quad (8.12)$$

The constants k and k' in (8.10) to (8.12) may be determined from a knowledge of the density ρ at the faces $y_1 = a_1$, $y_1 = a_2$.

9. STEADY DIFFUSION THROUGH FLUID IN LAMINAR FLOW

The theory of §§ 2 to 4 may be used to investigate either the interdiffusion of several fluids or the flow of a mixture of fluids. We examine first a steady state problem which may be regarded as a combination of the diffusion and mixed flow problems.

We suppose that fluid 1 is in steady state laminar flow such that particles in y_1 , y_3 planes at the current time t are moving in the y_1 -direction. A second fluid is diffusing steadily into fluid 1 so that in addition to the motion in the y_1 direction characterizing the laminar flow of fluid 1, the particles of fluid 2 also have a velocity component in the y_2 direction. The velocity components at the point y_i for the constituents of the mixture may therefore be taken as

$$\left. \begin{aligned} (v_1^{(1)}, v_2^{(1)}, v_3^{(1)}) &= (U, 0, 0), & U &= U(y_2) & \text{(fluid 1),} \\ (v_1^{(2)}, v_2^{(2)}, v_3^{(2)}) &= (u, v, 0), & u &= u(y_2), & v &= v(y_2) & \text{(fluid 2),} \end{aligned} \right\} \quad (9.1)$$

respectively, and the corresponding densities are ρ_1 , ρ_2 .

From (9.1) and (3.4) we see that

$$A_{ij}^{(1)} = \begin{bmatrix} 0 & \frac{1}{2}U' & 0 \\ \frac{1}{2}U' & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad A_{ij}^{(2)} = \begin{bmatrix} 0 & \frac{1}{2}u' & 0 \\ \frac{1}{2}u' & v' & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (9.2)$$

the prime denoting differentiation with respect to y_2 .

We consider non-Newtonian fluids with constitutive equations of the form (3.8); a corresponding analysis for more general visco-elastic liquids presents no difficulty of principle. From (9.2) and (3.8) we obtain for fluid 1

$$\left. \begin{aligned} \sigma_{11}^{(1)} = \sigma_{22}^{(1)} = -p_1 + \phi_0 + \phi_2 U'^2/4, \quad \sigma_{33}^{(1)} = -p_1 + \phi_0, \\ \sigma_{12}^{(1)} = \frac{1}{2}\phi_1 U', \quad \sigma_{23}^{(1)} = \sigma_{31}^{(1)} = 0, \end{aligned} \right\} \quad (9.3)$$

and for fluid 2

$$\left. \begin{aligned} \sigma_{11}^{(2)} = -p_2 + \Phi_0 + \frac{1}{4}u'^2\Phi_2, \\ \sigma_{22}^{(2)} = -p_2 + \Phi_0 + v'\Phi_1 + (\frac{1}{4}u'^2 + v'^2)\Phi_2, \\ \sigma_{33}^{(2)} = -p_2 + \Phi_0, \\ \sigma_{12}^{(2)} = \frac{1}{2}u'(\Phi_1 + v'\Phi_2), \quad \sigma_{13}^{(2)} = \sigma_{23}^{(2)} = 0. \end{aligned} \right\} \quad (9.4)$$

Here and subsequently, to simplify the notation when considering a mixture of two fluids we write, in (3.5), (3.8) and (6.6) $\phi_\nu^{(1)} = \phi_\nu$ (fluid 1) and $\phi_\nu^{(2)} = \Phi_\nu$ (fluid 2) ($\nu = 0, 1, 2$). In (9.3), p_1, ϕ_0, ϕ_1 and ϕ_2 are functions of ρ_1 and ϕ_0, ϕ_1, ϕ_2 are also polynomials in the invariants (3.7). From (9.2) we see that this implies that ϕ_0, ϕ_1 and ϕ_2 are polynomials in U'^2 . Similarly p_2, Φ_0, Φ_1 and Φ_2 in (9.4) are functions of ρ_2 and from (9.2) and (3.7) we infer that Φ_0, Φ_1, Φ_2 may be regarded as polynomials in v' and u'^2 .

Writing in (4.12) $\mathbf{v}_n = \mathbf{v}_2, \mathbf{u}_1 = \mathbf{v}_1 - \mathbf{v}_2, \mathbf{u}_2 = \mathbf{0}$, we see from (9.1) and (4.19) that the diffusive force components $\Psi_i^{(1)}$ for fluid 1 are given by

$$(\Psi_1^{(1)}, \Psi_2^{(1)}, \Psi_3^{(1)}) = [(U-u)\alpha, -v\alpha, 0], \quad (9.5)$$

where α is a polynomial in the invariants (4.18) (i), or in the present instance of

$$(U-u)^2 + v^2, \quad (9.6)$$

with coefficients which are functions of ρ_1, ρ_2 . The corresponding components $\Psi_i^{(2)}$ for fluid 2 are given, from the second of (2.14) by

$$\rho_2 \Psi_i^{(2)} = -\rho_1 \Psi_i^{(1)}. \quad (9.7)$$

We examine the possibility of flow in which ρ_1, ρ_2, p_1 and p_2 are functions only of y_2 . In the absence of extraneous body forces, the equations of motion (2.12) combined with (9.3) to (9.7), yield

$$\left. \begin{aligned} \frac{d}{dy_2}(\phi_1 U') + 2\rho_1(U-u)\alpha = 0, \\ \frac{d}{dy_2}(-p_1 + \phi_0 + \frac{1}{4}\phi_2 U'^2) - \rho_1 v\alpha = 0 \quad (\text{fluid 1}), \end{aligned} \right\} \quad (9.8)$$

$$\left. \begin{aligned} \frac{d}{dy_2}[u'(\Phi_1 + v'\Phi_2)] - 2\rho_1(U-u)\alpha = 2\rho_2 v u', \\ \frac{d}{dy_2}[-p_2 + \Phi_0 + v'\Phi_1 + (\frac{1}{4}u'^2 + v'^2)\Phi_2] + \rho_1 v\alpha = \rho_2 v v' \quad (\text{fluid 2}). \end{aligned} \right\} \quad (9.9)$$

In the steady flow considered, $\partial\rho_1/\partial t = \partial\rho_2/\partial t = 0$. The equation of continuity (2.10) is thus satisfied identically for fluid 1 but for fluid 2, we have

$$d(\rho_2 v)/dy_2 = 0 \quad \text{or} \quad \rho_2 v = k, \quad (9.10)$$

k being a constant.

If p_1 and p_2 can be regarded as known functions of ρ_1, ρ_2 then (9·8) to (9·10) represent a system of five equations for the determination of U, u, v, ρ_1, ρ_2 as functions of y_2 .

By making use of (9·10) in the first of (9·9) and combining the resulting equation with the first of (9·8) we obtain

$$\phi_1 U' + u'(\Phi_1 + v' \Phi_2) = 2ku + K_1; \quad (9\cdot11)$$

similarly from the second equations of (9·8) and (9·9) we obtain

$$-(p_1 + p_2) + \phi_0 + \Phi_0 + v' \Phi_1 + \frac{1}{4} U'^2 \phi_2 + (\frac{1}{4} u'^2 + v'^2) \Phi_2 = kv + K_2, \quad (9\cdot12)$$

K_1 and K_2 being constants of integration. Equations (9·11) and (9·12) are first order differential equations involving U, u and v and may be regarded as first integrals of the equations of motion.

When both fluids are in laminar flow, so that $v = 0$, equations (9·8) and (9·9) reduce to

$$\left. \begin{aligned} d(\phi_1 U')/dy_2 + 2\rho_1(U-u)\alpha &= 0, \\ d(-p_1 + \phi_0 + \frac{1}{4}\phi_2 U'^2)/dy_2 &= 0 \quad (\text{fluid 1}), \end{aligned} \right\} \quad (9\cdot13)$$

$$\left. \begin{aligned} d(\Phi_1 u')/dy_2 - 2\rho_1(U-u)\alpha &= 0, \\ d(-p_2 + \Phi_0 + \frac{1}{4}\Phi_2 u'^2)/dy_2 &= 0 \quad (\text{fluid 2}), \end{aligned} \right\} \quad (9\cdot14)$$

respectively, and (9·10) is satisfied identically.

In this case the second of equations (9·13) and (9·14) integrate to yield

$$\left. \begin{aligned} 4p_1 &= 4\phi_0 + \phi_2 U'^2 + K'_1, \\ 4p_2 &= 4\Phi_0 + \Phi_2 u'^2 + K'_2, \end{aligned} \right\} \quad (9\cdot15)$$

and from the first of each pair of equations we obtain

$$\phi_1 U' + \Phi_1 u' = K'_3, \quad (9\cdot16)$$

where K'_1, K'_2, K'_3 are constants.

The system of equations (9·13) and (9·14) is evidently satisfied if ρ_1, ρ_2, p_1 and p_2 are constants and U and u have the linear forms

$$U = u = ay_2 + b, \quad (9\cdot17)$$

where a and b are constants. In this case, the mixture is homogeneous and remains so throughout the motion.

More generally, (9·13) and (9·14) form a system of four differential equations involving $U, u, \rho_1, \rho_2, p_1$ and p_2 . These may be solved, in principle, if we postulate, for compressible fluids, relationships of the form

$$p_1 = f_1(\rho_1), \quad p_2 = f_2(\rho_2) \quad (9\cdot18)$$

between the partial pressures p_1, p_2 and the densities ρ_1, ρ_2 . These relationships assume that the pressure-density relationship for either constituent is unaffected by the presence of the other. More generally, we might assume coupled relationships of the form

$$p_1 = f_1(\rho_1, \rho_2), \quad p_2 = f_2(\rho_1, \rho_2). \quad (9\cdot19)$$

For an incompressible mixture, $p = p_1 + p_2$ is indeterminate and is, in general, a function of position which is determined from the field equations and the boundary conditions. The total density $\rho = \rho_1 + \rho_2$ is independent of p . If the density of the mixture is also independent of its composition then

$$\rho = \rho_1 + \rho_2 = \text{constant}. \quad (9\cdot20)$$

More generally, we might suppose that although a mixture of given composition is incompressible, the density ρ varies as we vary ρ_1, ρ_2 . In this case we have a functional relation of the form

$$\rho \equiv \rho_1 + \rho_2 = f(\rho_1, \rho_2). \quad (9.21)$$

Either of the relations (9.20) or (9.21) implies a known functional relationship between ρ_1 and ρ_2 .

To complete the system of field equations for an incompressible mixture, we require a further relationship involving the partial pressures p_1, p_2 and the densities ρ_1, ρ_2 . If

$$F(\rho_1, \rho_2, p_1, p_2) = 0, \quad (9.22)$$

then this relation, together with (9.20) or (9.21) may replace (9.18) or (9.19). As a particular case of (9.22) it is plausible to suppose that the ratio of the partial pressures p_1, p_2 depends upon the composition of the mixture. This will evidently be the case for a homogeneous mixture at rest which is subjected to a hydrostatic pressure. We may therefore write

$$p_1/p_2 = F_1(\rho_1, \rho_2), \quad (9.23)$$

or, rather less generally,
$$p_1/p_2 = F_2(\rho_1/\rho_2), \quad (9.24)$$

where F_1 is a monotonic increasing function of ρ_1 and a monotonic decreasing function of ρ_2 such that $F_1(0, \rho_2) = 0, F_1(\rho_1, 0) = \infty$ and $F_2(x)$ is a monotonic increasing function of x such that $F_2(0) = 0$ and $F_2(x) \rightarrow \infty$ as $x \rightarrow \infty$. Dimensional considerations support a relationship of the form (9.24).

If a solution of the system (9.13), (9.14) can be found in which U and u do not have the linear form (9.17) then in general, from (9.15), the partial pressures p_1, p_2 and the densities ρ_1, ρ_2 will be constant throughout the fluid only if ϕ_0 and Φ_0 are independent of u'^2 and U'^2 and ϕ_2 and Φ_2 are both zero. This suggests that if a homogeneous mixture of non-Newtonian fluids is subjected to a non-uniform laminar flow, the constituents will tend to separate out, this separation occurring as a specifically non-Newtonian effect.

10. FLOW OF MIXTURE BETWEEN ROTATING CYLINDERS

We consider a mixture of two non-Newtonian fluids contained in the annular space between two infinitely long concentric circular cylinders of radii a_1, a_2 ($a_1 > a_2$). We suppose the cylinders to rotate about their axis with different uniform angular velocities Ω_1, Ω_2 respectively and examine the possibility of steady two-dimensional flow in which each particle of fluid moves uniformly in a circular path around the axis. This problem has been examined theoretically by Rivlin (1948, 1956), Oldroyd (1950, 1951) and others for single-component non-Newtonian and visco-elastic fluids, and experimental investigations of anomalous effects have been carried out by Weissenberg (1947), Greensmith & Rivlin (1953) and others. In the present instance we neglect gravitational effects.

In a system of cylindrical polar co-ordinates we denote the current position of a particle P of fluid by (r, θ, y_3) where

$$y_1 = r \cos \theta, \quad y_2 = r \sin \theta, \quad (10.1)$$

and the y_3 direction coincides with the axis of the cylinders. We suppose that at P , the particles of fluids 1 and 2 have angular velocities ω, Ω respectively about the axis of the cylinders, ω and Ω being functions only of r .

We choose the curvilinear co-ordinates ξ^i of § 6 so that

$$(\xi^1, \xi^2, \xi^3) = (r, \theta, y_3), \quad (10\cdot2)$$

and the covariant and contravariant metric tensors G_{ij} , G^{ij} are given by

$$\left. \begin{aligned} (G_{11}, G_{22}, G_{33}) &= (1, r^2, 1), & G^{ii} &= 1/G_{ii} \quad (i \text{ not summed}), \\ G_{ij} &= G^{ij} = 0 \quad (i \neq j). \end{aligned} \right\} \quad (10\cdot3)$$

For fluid 1, the contravariant velocity components $w^{(1)i}$ are given by

$$(w^{(1)1}, w^{(1)2}, w^{(1)3}) = (0, \omega, 0), \quad (10\cdot4)$$

and the mixed and contravariant components $d^{(1)i}_j$, $d^{(1)ij}$ of the rate of deformation tensor are

$$d^{(1)i}_j = \begin{bmatrix} 0 & \frac{1}{2}r^2\omega' & 0 \\ \frac{1}{2}\omega' & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad d^{(1)ij} = \begin{bmatrix} 0 & \frac{1}{2}\omega' & 0 \\ \frac{1}{2}\omega' & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (10\cdot5)$$

the prime denoting differentiation with respect to r .

If we assume constitutive equations of the form (6·6), the components $\tau^{(1)ij}$ of the stress tensor for fluid 1 take the form

$$\left. \begin{aligned} \tau^{(1)11} &= r^2\tau^{(1)22} = -p_1 + \phi_0 + \frac{1}{4}r^2\omega'^2\phi_2, \\ \tau^{(1)33} &= -p_1 + \phi_0, \\ \tau^{(1)12} &= \frac{1}{2}\omega'\phi_1, \quad \tau^{(1)13} = \tau^{(1)23} = 0, \end{aligned} \right\} \quad (10\cdot6)$$

where, from (6·8) and (10·5), ϕ_0 , ϕ_1 and ϕ_2 are polynomials in $r^2\omega'^2$. The quantities p , ϕ_0 , ϕ_1 , ϕ_2 are also functions of the density ρ_1 .

Equations corresponding to (10·4) to (10·6) hold for fluid 2, with the index 1 replaced by 2 throughout and ω , ϕ_0 , ϕ_1 , ϕ_2 replaced by Ω , Φ_0 , Φ_1 , Φ_2 , respectively.

From (6·9), the contravariant components ${}^{(1)}\Psi^i$ of the diffusive body force for fluid 1 are given by

$$({}^{(1)}\Psi^1, {}^{(1)}\Psi^2, {}^{(1)}\Psi^3) = [0, (\omega - \Omega)\alpha, 0], \quad (10\cdot7)$$

where α is a function of the densities ρ_1 , ρ_2 and, from (4·18) (i), is a polynomial in $r^2(\omega - \Omega)^2$.

For fluid 2, we have, from (2·14) $\rho_2{}^{(2)}\Psi^i = -\rho_1{}^{(1)}\Psi^i$.

$$(10\cdot8)$$

For steady-state motion in which there is cylindrical symmetry, the equations of motion (6·5), with (10·3), (10·4), (10·6) and (10·7) yield, for fluid 1

$$\left. \begin{aligned} \frac{d}{dr}(-p_1 + \phi_0 + \frac{1}{4}r^2\omega'^2\phi_2) &= -\rho_1\omega^2r, \\ \frac{1}{2r^3} \frac{d}{dr}(r^3\omega'\phi_1) + \rho_1(\omega - \Omega)\alpha &= 0, \end{aligned} \right\} \quad (10\cdot9)$$

the third equation being satisfied identically if we assume p_1 and ρ_1 to be functions only of r . Similarly for fluid 2 we have

$$\left. \begin{aligned} \frac{d}{dr}(-p_2 + \Phi_0 + \frac{1}{4}r^2\Omega'^2\Phi_2) &= -\rho_2\Omega^2r, \\ \frac{1}{2r^3} \frac{d}{dr}(r^3\Omega'\Phi_1) - \rho_1(\omega - \Omega)\alpha &= 0. \end{aligned} \right\} \quad (10\cdot10)$$

The equations of continuity (2·10) are satisfied identically.

If p_1 and p_2 are regarded as known functions of ρ_1 and ρ_2 through relations of the form (9·18) or (9·19), then (10·9) and (10·10) may be regarded as four differential equations for the determination of ω , Ω , ρ_1 and ρ_2 as functions of r . In the case of an incompressible fluid, we may eliminate ρ_1 and p_1 from (10·9) and (10·10) by means of (9·20) or (9·21) and (9·22), (9·23) or (9·24), and these equations then serve to determine ω , Ω , ρ_2 and p_2 . We observe that ρ_1 and ρ_2 are not, in general, constants. The components of an initially homogeneous mixture will therefore tend to separate out during the rotation, the variation with radius of the composition of the mixture in any final steady-state motion depending upon the angular speeds Ω_1 , Ω_2 of the outer and inner cylinders and upon the constitutive equations for the fluids and for the diffusive force. This effect is analogous to that for plane laminar flow discussed in § 9.

From the second of (10·9) and (10·10) we readily obtain a first integral in the form

$$r^3(\omega'\phi_1 + \Omega'\Phi_1) = K, \quad (10\cdot11)$$

where K is a constant. If centrifugal forces can be neglected, the first of equations (10·9) and (10·10) can be integrated to yield

$$\left. \begin{aligned} 4(p_1 - \phi_0) &= r^2\omega'^2\phi_2 + k_1, \\ 4(p_2 - \Phi_0) &= r^2\Omega'^2\Phi_2 + k_2, \end{aligned} \right\} \quad (10\cdot12)$$

k_1 and k_2 being constants of integration. The resultant normal stress across planes $y_3 = \text{constant}$ is therefore, from (10·6) and the analogous equation for $\tau^{(2)33}$,

$$\tau^{(1)33} + \tau^{(2)33} = -\frac{1}{4}\{r^2(\omega'^2\phi_2 + \Omega'^2\Phi_2) + k_1 + k_2\}. \quad (10\cdot13)$$

If fluid 2 is an ideal fluid, for which $\Phi_0 = \Phi_1 = \Phi_2 = 0$, then from (10·10) provided $\alpha \neq 0$,

$$\Omega = \omega, \quad dp_2/dr = \rho_2\omega^2r, \quad (10\cdot14)$$

and from (10·9)

$$r^3\omega'\phi_1 = \text{constant}. \quad (10\cdot15)$$

If p_1 , ϕ_0 , ϕ_1 and ϕ_2 are known functions of ρ_1 and ω' , equations (10·15) and the first of (10·9) serve to determine ρ_1 and ω . The second of (10·14) may then be regarded as an equation for ρ_2 . Although the angular speeds of the two fluids are now identical, we cannot conclude that the radial distribution of densities is uniform.

WAVE PROPAGATION

11. PROPAGATION OF PLANE WAVES THROUGH A MIXTURE

The equations of §§ 2 to 4 may be expected to imply anomalous effects in the propagation of waves in mixtures. We examine here the propagation of plane progressive waves through a homogeneous two-component mixture which is infinite in extent and is at rest apart from the infinitesimal disturbance produced by the wave motion.

We consider a wave of angular frequency ω propagated in the x_1 direction of the rectangular Cartesian reference frame x_i . The velocity components $v_j^{(1)}$, $v_j^{(2)}$ for fluids 1 and 2, respectively are therefore given by

$$v_j^{(1)} = a_j \exp [i(kx_1 - \omega t)], \quad v_j^{(2)} = b_j \exp [i(kx_1 - \omega t)], \quad (11\cdot1)$$

where a_j , b_j and k are constants. The densities ρ_1 , ρ_2 of the two fluids may be written

$$\rho_1 = \bar{\rho}_1 + \rho'_1 \exp [i(kx_1 - \omega t)], \quad \rho_2 = \bar{\rho}_2 + \rho'_2 \exp [i(kx_1 - \omega t)], \quad (11.2)$$

where $\bar{\rho}_1$, $\bar{\rho}_2$ are the constant densities in the initial rest state and ρ'_1 , ρ'_2 are constants such that $\rho'_1 \ll \bar{\rho}_1$, $\rho'_2 \ll \bar{\rho}_2$.

We assume constitutive equations of the form (3.8) and make the additional assumption that the partial pressures p_1 , p_2 can be related to the densities by relations of the form (9.18). To the first order in ρ'_1 we may therefore write

$$p_1 = \bar{p}_1 + p'_1 \exp [i(kx_1 - \omega t)], \quad (11.3)$$

$$\text{where} \quad \bar{p}_1 = f_1(\bar{\rho}_1), \quad p'_1 = c_1 \rho'_1, \quad c_1 = [df_1(\rho_1)/d\rho_1]_{\rho_1=\bar{\rho}_1}, \quad (11.4)$$

and corresponding relations hold for p_2 .

For the small disturbance represented by (11.1) we neglect squares and products of small quantities and take the constitutive equations (3.8) in the classical form

$$\sigma_\nu = (-p_\nu + \lambda_\nu \text{tr } \mathbf{A}_\nu) \mathbf{I} + 2\mu_\nu \mathbf{A}_\nu \quad (\nu = 1, 2; \nu \text{ not summed}), \quad (11.5)$$

where λ_ν and μ_ν are functions of the corresponding density ρ_ν and to a sufficient degree of approximation may be regarded as constants. From (11.1) to (11.5), the stress components $\sigma_{ij}^{(1)}$ for fluid 1 become

$$\left. \begin{aligned} \sigma_{11}^{(1)} &= -\bar{p}_1 + [ika_1(\lambda_1 + 2\mu_1) - p'_1] \exp [i(kx_1 - \omega t)], \\ \sigma_{22}^{(1)} &= \sigma_{33}^{(1)} = -\bar{p}_1 + [ika_1\lambda_1 - p'_1] \exp [i(kx_1 - \omega t)], \\ \sigma_{1\gamma}^{(1)} &= ik a_\gamma \mu_1 \exp [i(kx_1 - \omega t)] \quad (\gamma = 2, 3), \quad \sigma_{23}^{(1)} = 0. \end{aligned} \right\} \quad (11.6)$$

The components $\Psi_j^{(1)}$ of the diffusive force acting on constituent 1, may, from (4.19) and (11.1) be written in the form

$$\Psi_j^{(1)} = (v_j^{(1)} - v_j^{(2)}) \alpha = (a_j - b_j) \alpha \exp [i(kx_1 - \omega t)], \quad (11.7)$$

where, to the first order in a_i , b_i , ρ'_1 and ρ'_2 , α is a constant and as before, we have taken $\mathbf{u}_1 = \mathbf{v}_1 - \mathbf{v}_2$, $\mathbf{u}_2 = \mathbf{0}$. For fluid 2, the diffusive force components are given by

$$\rho_2 \Psi_j^{(2)} = -\rho_1 \Psi_j^{(1)}. \quad (11.8)$$

In the absence of extraneous body forces, the equations of motion (2.12), with (11.4), (11.6), (11.7) and $y_j = x_j$, yield

$$\left. \begin{aligned} [\bar{\rho}_1(\alpha + i\omega) - k^2(\lambda_1 + 2\mu_1)] a_1 - \bar{\rho}_1 \alpha b_1 - ikc_1 \rho'_1 &= 0, \\ [\bar{\rho}_1(\alpha + i\omega) - k^2\mu_1] a_\gamma - \bar{\rho}_1 \alpha b_\gamma &= 0 \quad (\gamma = 2, 3), \end{aligned} \right\} \quad (11.9)$$

Similarly, for the component 2, we obtain

$$\left. \begin{aligned} [\bar{\rho}_1 \alpha + i\omega \bar{\rho}_2 - k^2(\lambda_2 + 2\mu_2)] b_1 - \bar{\rho}_1 \alpha a_1 - ikc_2 \rho'_2 &= 0, \\ [\bar{\rho}_1 \alpha + i\omega \bar{\rho}_2 - k^2\mu_2] b_\gamma - \bar{\rho}_1 \alpha a_\gamma &= 0 \quad (\gamma = 2, 3). \end{aligned} \right\} \quad (11.10)$$

With (11.9) and (11.10) must be coupled the equations of continuity which, from (2.10), (11.1) and (11.2) yield

$$\left. \begin{aligned} k\bar{\rho}_1 a_1 - \omega \rho'_1 &= 0, \\ k\bar{\rho}_2 b_1 - \omega \rho'_2 &= 0. \end{aligned} \right\} \quad (11.11)$$

As in the case of single component solids or fluids we may consider separately transverse waves and longitudinal waves. Here, however, the presence of the diffusive force has the effect of coupling the waves which would be present in either of the media alone.

12. TRANSVERSE WAVES

From the second of (11·9) and (11·10) we obtain by eliminating a_ν, b_ν the secular equation

$$k^4\mu_1\mu_2 - k^2[\bar{\rho}_1\alpha(\mu_1 + \mu_2) + i\omega(\bar{\rho}_2\mu_1 + \bar{\rho}_1\mu_2)] + [i\omega\bar{\rho}\bar{\rho}_1\alpha - \omega^2\bar{\rho}_1\bar{\rho}_2] = 0 \quad (\bar{\rho} = \bar{\rho}_1 + \bar{\rho}_2). \quad (12\cdot1)$$

If we assume that ω is real and positive, then from (12·1) by a straightforward formal calculation, we obtain for k^2 the complex values

$$k^2 = K_\nu + iL_\nu \quad (\nu = 1, 2), \quad (12\cdot2)$$

where

$$\left. \begin{aligned} K_1, K_2 &= [\bar{\rho}_1\alpha(\mu_1 + \mu_2) \pm \beta \cos \chi] / (2\mu_1\mu_2), \\ L_1, L_2 &= [\omega(\bar{\rho}_2\mu_1 + \bar{\rho}_1\mu_2) \pm \beta \sin \chi] / (2\mu_1\mu_2), \end{aligned} \right\} \quad (12\cdot3)$$

with

$$\left. \begin{aligned} \beta^4 &= \bar{\rho}_1^4\alpha^4(\mu_1 + \mu_2)^4 + \omega^4(\bar{\rho}_2\mu_1 - \bar{\rho}_1\mu_2)^4 + 2\bar{\rho}_1^2\omega^2\alpha^2(\bar{\rho}_2\mu_1 - \bar{\rho}_1\mu_2)^2(\mu_1^2 + \mu_2^2 - 6\mu_1\mu_2), \\ \cot 2\chi &= \frac{\bar{\rho}_1^2\alpha^2(\mu_1 + \mu_2)^2 - \omega^2(\bar{\rho}_2\mu_1 - \bar{\rho}_1\mu_2)^2}{2\omega\bar{\rho}_1\alpha(\mu_1 - \mu_2)(\bar{\rho}_2\mu_1 - \bar{\rho}_1\mu_2)}. \end{aligned} \right\} \quad (12\cdot4)$$

From (12·2) it follows that

$$k = \pm \gamma_\nu(\cos \psi_\nu + i \sin \psi_\nu) = \pm(k_\nu + il_\nu) \quad (\text{say}) \quad (\nu = 1, 2), \quad (12\cdot5)$$

where k_ν, l_ν are real and

$$\gamma_\nu^4 = K_\nu^2 + L_\nu^2, \quad \tan 2\psi_\nu = L_\nu/K_\nu \quad (\nu = 1, 2). \quad (12\cdot6)$$

Of the four solutions (12·5), two yield values for k with positive real parts. From (11·1) we see that these represent waves travelling in the positive x_1 direction; the remaining solutions represent waves travelling in the opposite direction. In either case, from (11·1) it follows that the wave is attenuated during propagation if k_ν and l_ν have the same sign, that is if $\tan \psi_\nu = l_\nu/k_\nu$ is positive. If k_ν and l_ν have opposite signs, that is, if $\tan \psi_\nu$ is negative, the wave builds up during propagation and we shall assume that this situation cannot occur. The condition for stability may evidently impose a restriction upon the constants occurring in the constitutive equations for the fluids and the diffusive force.

If both $\tan \psi_1$ and $\tan \psi_2$ are positive, we may thus obtain from (12·1) two different solutions for k for any given angular frequency ω . These correspond to two different waves travelling with different speeds and suffering different amounts of attenuation and dispersion. When the diffusive force is large, so that the terms in α predominate in (12·1) one value of k^2 becomes very large, and for the other we have, approximately, the finite value

$$k^2 = \frac{i\omega\bar{\rho}}{\mu_1 + \mu_2} \quad \text{or} \quad k = \pm(1+i) \sqrt{\left(\frac{\omega\bar{\rho}}{2(\mu_1 + \mu_2)}\right)}. \quad (12\cdot7)$$

This corresponds to the case in which the two liquids diffuse only slowly into each other.

If the diffusive force, and therefore α , is small, equations (11·9) and (11·10) may to a first approximation be regarded as uncoupled. In this case we obtain the solutions

$$k = \pm(1+i) \sqrt{\{\bar{\rho}_\nu\omega/(2\mu_\nu)\}} \quad (\nu = 1, 2), \quad (12\cdot8)$$

corresponding to wave propagation in each of the fluids in the absence of the other. We observe that in each of these two extreme cases the situation is stable and an attenuated wave is obtained.

It is of some interest to examine the situation corresponding to a real wave number k and a complex angular frequency ω . In this case, by writing $\omega = \omega_1 + i\omega_2$, where ω_1 and ω_2 are real, and separating real and imaginary parts in (12.1) we obtain

$$\left. \begin{aligned} \omega_1 &= \pm [\Lambda/(\bar{\rho}_1 \bar{\rho}_2) - \omega_2^2]^{\frac{1}{2}}, \\ \omega_2 &= \Gamma/(2\bar{\rho}_1 \bar{\rho}_2), \end{aligned} \right\} \quad (12.9)$$

where

$$\left. \begin{aligned} \Gamma &= \bar{\rho} \bar{\rho}_1 \alpha - k^2(\bar{\rho}_2 \mu_1 + \bar{\rho}_1 \mu_2), \\ \Lambda &= k^2[k^2 \mu_1 \mu_2 - \bar{\rho}_1 \alpha(\mu_1 + \mu_2)]. \end{aligned} \right\} \quad (12.10)$$

We assume that the expression in brackets in the first of (12.9) is positive, so that ω_1 is real. Equations (11.1) then represent a disturbance which is periodic in x_1 and which decays exponentially with time provided ω_2 is negative. It is readily seen from (12.9) and (12.10) that this is, in general true, for in these equations $\bar{\rho}_1$, $\bar{\rho}_2$, μ_1 and μ_2 are positive quantities, and as indicated in § 7, it is physically reasonable to assume that α is negative.

From (12.9) we see that ω_1 is real if

$$\Gamma^2 - 4\bar{\rho}_1 \bar{\rho}_2 \Lambda < 0.$$

If $\Gamma^2 - 4\bar{\rho}_1 \bar{\rho}_2 \Lambda > 0$ then we may have $\omega_1 = 0$ and

$$\omega_2 = [\Gamma \pm (\Gamma^2 - 4\bar{\rho}_1 \bar{\rho}_2 \Lambda)^{\frac{1}{2}}]/(2\bar{\rho}_1 \bar{\rho}_2). \quad (12.11)$$

These values of ω_2 are both negative, giving a disturbance which is non-periodic in time and decays exponentially with time provided Γ is negative and $\Lambda > 0$.

13. LONGITUDINAL WAVES

If we eliminate a_1 , b_1 , ρ'_1 and ρ'_2 from (11.10) and the first equations of (11.8) and (11.9) we obtain

$$\begin{vmatrix} \bar{\rho}_1(\alpha + i\omega) - k^2\kappa_1 & -\bar{\rho}_1\alpha & -i\kappa c_1 & 0 \\ -\bar{\rho}_1\alpha & \bar{\rho}_1\alpha + i\omega\bar{\rho}_2 - k^2\kappa_2 & 0 & -i\kappa c_2 \\ k\bar{\rho}_1 & 0 & -\omega & 0 \\ 0 & k\bar{\rho}_2 & 0 & -\omega \end{vmatrix} = 0,$$

or

$$\begin{aligned} &k^4\{\omega^2\kappa_1\kappa_2 - c_1c_2\bar{\rho}_1\bar{\rho}_2 + i\omega(c_1\bar{\rho}_1\kappa_2 + c_2\bar{\rho}_2\kappa_1)\} \\ &+ k^2\{\omega^2\bar{\rho}_1[\bar{\rho}_2c - \alpha(\kappa_1 + \kappa_2)] - i\omega[\bar{\rho}_1\alpha(\bar{\rho}_1c_1 + \bar{\rho}_2c_2) + \omega^2(\bar{\rho}_1\kappa_2 + \bar{\rho}_2\kappa_1)]\} \\ &+ \omega^3\bar{\rho}_1(i\bar{\rho}\alpha - \omega\bar{\rho}_2) = 0, \end{aligned} \quad (13.1)$$

where

$$\kappa_1 = \lambda_1 + 2\mu_1, \quad \kappa_2 = \lambda_2 + 2\mu_2, \quad c = c_1 + c_2. \quad (13.2)$$

The relation (13.1), regarded as an equation in k^2 , is similar in structure to (12.1), with coefficients of k^3 and k zero and the remaining coefficients complex. We may therefore obtain solutions for k analogous to (12.5), two of which yield real waves, suffering attenuation and dispersion during propagation provided the appropriate values of k_p and l_p have the same sign.

As in the case of transverse waves, when the diffusive force, and hence α , is small the two values of k^2 tend towards the values which would be obtained in either fluid in the absence

of the other. If the diffusive force is sufficiently large for the terms in α to predominate in (13.1) we obtain, for the finite values of the wave number

$$k^2 = \frac{i\omega^2\bar{\rho}}{\omega(\kappa_1 + \kappa_2) + i(\bar{\rho}_1 c_1 + \bar{\rho}_2 c_2)}. \quad (13.3)$$

When α is large compared with unity, the finite values (12.7) and (13.3) for k^2 may be derived directly from (11.9) to (11.11) if we observe that in this case $a_i \approx b_i$. Writing $b_i = a_i + a'_i/\alpha$ where a'_i are finite constants, and discarding terms of order $1/\alpha$ in the resulting equations, we obtain a system of equations linear in $a_i, a'_i, \rho'_1, \rho'_2$. The elimination of these constants leads to the values (12.7), (13.3) for k^2 .

It is of some interest to consider the propagation of longitudinal waves through a homogeneous mixture of gases whose densities ρ_1, ρ_2 and pressure density relationships do not differ greatly from each other. In this case we write

$$\left. \begin{aligned} \lambda_\nu = \mu_\nu = \kappa_\nu = 0 \quad (\nu = 1, 2), \\ \bar{\rho}_2 = \bar{\rho}_1 + \epsilon\bar{\rho}'_1, \quad c_2 = c_1 + \epsilon c'_1, \end{aligned} \right\} \quad (13.4)$$

where ϵ is a small (constant) parameter and $\bar{\rho}'_1$ and c'_1 are constants. When the gases are identical in physical characteristics, so that $\bar{\rho}_2 = \bar{\rho}_1, c_2 = c_1$ we may choose $b_1 = a_1$ and $\rho'_2 = \rho'_1$. The equations derived from the first of (11.9) and (11.10) then become identical and equations (11.11) also yield only one distinct equation. In this case, as might be expected, the mixture behaves as a single-constituent gas.

When $\bar{\rho}_2$ and c_2 are given by (13.4) we write

$$b_1 = a_1 + \epsilon a'_1, \quad \rho'_2 = \rho'_1 + \epsilon \rho''_1, \quad (13.5)$$

where a'_1 and ρ''_1 are constants. By introducing (13.4) and (13.5) into (11.11) and the first equations of (11.9) and (11.10) and eliminating $a_1, a'_1, \rho'_1, \rho''_1$ from the resulting equations we derive in place of (13.1) the secular equation

$$\begin{vmatrix} i\omega\bar{\rho}_1 & 0 & -ikc_1 & 0 \\ i\omega\bar{\rho}'_1 & \bar{\rho}_1(2\alpha + i\omega) & -ikc'_1 & -ikc_1 \\ k\bar{\rho}_1 & 0 & -\omega & 0 \\ k\bar{\rho}'_1 & k\bar{\rho}_1 & 0 & -\omega \end{vmatrix} + O(\epsilon) = 0, \quad (13.6)$$

and neglecting terms of order ϵ this yields

$$\text{or } \left. \begin{aligned} k^2 = \omega^2/c_1 & \quad (\text{i}), \\ k^2 = \omega(\omega - 2i\alpha)/c_1 & \quad (\text{ii}). \end{aligned} \right\} \quad (13.7)$$

The first of these values of k^2 corresponds to the unattenuated wave which would be propagated in the single-constituent gas characterized by the constants $\bar{\rho}_1, c_1$. For the second value we may write from (13.7) (ii)

$$k^2 = R^2(\cos 2\psi + i \sin 2\psi), \quad (13.8)$$

where

$$\left. \begin{aligned} R^2 = \omega(\omega^2 + 4\alpha^2)^{\frac{1}{2}}/c_1, \\ \cos 2\psi = \omega/(\omega^2 + 4\alpha^2)^{\frac{1}{2}}, \quad \sin 2\psi = -2\alpha/(\omega^2 + 4\alpha^2)^{\frac{1}{2}}. \end{aligned} \right\} \quad (13.9)$$

If, as discussed in §7, α is negative, $\cos 2\psi$ and $\sin 2\psi$ are both positive, and we may choose $0 \leq 2\psi \leq \frac{1}{2}\pi$. In this case, k takes the form

$$k = \pm(A + iB), \quad (13\cdot10)$$

where, remembering (13·9)

$$\begin{aligned} A &= R \cos \psi = \{\omega[(\omega^2 + 4\alpha^2)^{\frac{1}{2}} + \omega]/(2c_1)\}^{\frac{1}{2}}, \\ B &= R \sin \psi = \{\omega[(\omega^2 + 4\alpha^2)^{\frac{1}{2}} - \omega]/(2c_1)\}^{\frac{1}{2}} \end{aligned} \quad (13\cdot11)$$

are both positive. From (11·1) we infer that the values (13·10) for k imply waves which are attenuated; the attenuation is rapid for large values of B , that is, for large values of α .

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