*J. Fluid Mech.* (1971), *vol.* 47, *part* 1, *pp.* 65–80
*Printed in Great Britain*

# Stability of circular Couette flow of binary mixtures

By WILLIAM S. SARIC
Sandia Laboratories, Albuquerque, New Mexico

AND ZALMAN LAVAN
Illinois Institute of Technology, Chicago, Illinois

(Received 12 January 1970 and in revised form 1 September 1970)

The hydrodynamic stability of an ideal mixture of two viscous, dissimilar liquids contained between two concentric rotating cylinders is analyzed. The basic flow of the mixture is determined by coupling the mass and momentum equations with an equation for the equilibrium concentration distribution. Infinitesimal, axisymmetric disturbances are assumed, and the disturbance equations are written for the limiting case of large Schmidt numbers (no diffusion).

The presence of density and viscosity variations leads to a twelfth-order eigenvalue problem with two-point boundary conditions that has the appearance of a combined Taylor and density-stratified shear flow problem. A numerical technique is devised to determine the stability boundary and to calculate Taylor numbers and oscillation frequencies for different growth rates.

It is found that very small mean density gradients alter the critical Taylor number and that oscillations occurring in both the growing and neutral solutions are the dominant mode.

## 1. Introduction

The interest in heterogeneous fluid flow problems with either a stratified density distribution or a multicomponent mixture has increased substantially in recent years. Such problems concern geophysical phenomena of the atmosphere and the ocean, mixtures of dissimilar liquids and gases, and flows with temperature gradients. In his book, Yih (1965) discusses a wide range of topics dealing with heterogeneous fluids.

This paper describes the investigation of the hydrodynamic stability of a mixture of two different fluids contained in the annulus of two concentric rotating cylinders, where a partial separation of the fluids occurs as the result of centrifugal effects. This problem may arise in journal bearings, rotating pumps, drilling operations, and other situations involving unstable circulation in heterogeneous liquids.

Essentially, the heavier component tends to migrate towards the outer cylinder and a concentration gradient is developed across the annulus. The steady-state and transient solutions of this problem have been reported by Kulinski (1966) but, to date, no quantitative analysis of the stability of such a flow has been attempted. One may say that the presence of the heavier fluid on the outside is somewhat analogous to the static problem of a lighter fluid on top of a heavier one, a situation known to be stable. On the other hand, an inviscid analysis of rotating fluids reveals that if the product of the density and the square of the circulation decreases outward, the flow is unstable (see Lin 1955). We investigate this combined effect of a stabilizing density distribution and a destabilizing circulation distribution in a viscous medium.

The hydrodynamic stability of homogeneous viscous flow in the annulus of two concentric cylinders was first successfully investigated, both theoretically and experimentally, by Taylor (1923). He showed that, at a given value of a parameter (later called the Taylor number) a secondary motion of cellular toroidal vortices appears. Chandrasekhar (1961) reports the early contributions in this area. Later, more complete solutions for the arbitrary gap size problem with axisymmetric disturbances were carried out by Sparrow, Munro & Jonsson (1964) for a large spectrum of radius ratios and angular velocity ratios. At the same time, Harris & Reid (1964) solved the small-gap problem numerically over the entire range of possible velocity ratios for axisymmetric disturbances. More recently, Krueger, Gross & DiPrima (1966) defined the regions of validity for the assumption of axisymmetric disturbances. Synder (1968) established this result experimentally and gave critical velocity ratios for several radius ratios.

The first solution of Taylor's problem with variable density and three-dimensional disturbances was obtained by Yih (1961) for the small gap. The density variation in a liquid was induced by a temperature gradient across the gap. Yih showed that the necessary and sufficient condition for inviscid stability, i.e. $d(\rho\Gamma^2)/dr > 0$ (where $\rho$ is density and $\Gamma$ circulation), is neither necessary nor sufficient when viscosity and thermal conductivity are taken into account. A more recent paper by Walowit, Tsao & DiPrima (1964) considers the effect of a temperature gradient over a large gap.

In the present investigation of a mixture of two dissimilar fluids, the heterogeneity is produced by a component separation caused by rotation rather than by a temperature gradient. The density gradient is, therefore, governed by the Schmidt number rather than the Prandtl number. Since the Schmidt number for liquids is several orders larger than the Prandtl number, the relative lack of mass diffusion on a short time basis could produce an instability similar to that occurring in a parallel shear flow of continuously stratified fluid when mass diffusion is neglected. Yih (1965) shows that, for this case, oscillatory solutions are present when the stratification is induced by either a temperature gradient or gravitational forces.

In fact, in our present problem, one would expect a combination of the Taylor problem and the stratified shear flow problem, i.e. an unfavourable circulation distribution that tends to produce instability, but the instability may be oscillating as in the stratified problem. The solution is treated as a Taylor problem because it is the combined effect of constituent separation and the onset of the Taylor instability that is of interest.

## 2. Development of general equations

Before proceeding with the stability analysis, the equations governing the flow of a binary mixture are established in a form suitable for investigating the effects of perturbations of density and viscosity from a two-fluid standpoint. The main assumptions that govern the problem formulation are: (i) no chemical reactions, sources, or sinks are present; (ii) no temperature gradients or body forces exist; (iii) the two liquids are miscible and the mixture is ideal, i.e. we assume that the Amagat–Leduc law for liquids holds and that the fugacities of the components equal their partial pressures; (iv) the mixture is Newtonian; (v) linear irreversible thermodynamics defines the constitutive equation for the diffusion mass flux and the usual viscous stress tensor (this is certainly valid for linear instability studies where only infinitesimal perturbations of the variables are considered); and (vi) the density of the mixture is a function of concentration alone.

With these assumptions, the conservation equations of specie, mass, and momentum for multicomponent systems (see, for example, DeGroot & Mazur 1962) can be rearranged as

$$\frac{DC^*}{Dt} - \frac{1+\beta C^*}{\beta}\operatorname{div}\mathbf{V}^* = 0, \tag{1}$$

$$\operatorname{div}\mathbf{V}^* + \frac{\beta}{\rho_b}\operatorname{div}\mathbf{J}^* = 0, \tag{2}$$

$$\rho^*\frac{D\mathbf{V}^*}{Dt} = -\operatorname{grad}P^* + \operatorname{div}\boldsymbol{\tau}^*, \tag{3}$$

where $C^*$ and $\mathbf{J}^*$ are the mass fraction and the diffusion mass flow of component $a$ of a system of two components denoted by subscripts $a$ and $b$: $P^*$ is the pressure; $D/Dt$ is the material derivative with respect to time; $\mathbf{V}^*$ is the mass averaged velocity of the mixture; $V_a^*$, $\rho_a^*$ and $V_b^*$, $\rho_b^*$ are the individual velocities and local densities of components $a$ and $b$; $\boldsymbol{\tau}^*$ is the usual viscous stress tensor; $\rho_a$ and $\rho_b$ are the densities of pure components $a$ and $b$; and $\rho^*$ is the density at any point in the mixture. In these equations,

$$\left.\begin{aligned} C^* &= m_a/m, \quad \beta = (\rho_b-\rho_a)/\rho_a, \\ \rho^*\mathbf{V}^* &= \rho_b^*\mathbf{V}_b^* + \rho_a^*\mathbf{V}_a^*, \\ \mathbf{J}^* &= \rho_a^*(\mathbf{V}_a^* - \mathbf{V}^*), \\ \rho^* &= \rho_b/(1+\beta C^*), \\ \rho_a^* &= C^*\rho^*, \quad \rho_b^* = (1-C^*)\rho^*, \end{aligned}\right\} \tag{4}$$

where $m_a$ is the mass of component $a$, and $m$ is the mass of the mixture. The details of the particular arrangement of equations (1) to (3) are contained in Saric (1968).

To complete the formulation of the governing equations, the constitutive relations for viscous stress, mass diffusion, and viscosity are required. The viscous stress tensor retains its usual form by virtue of Curie's theorem. In tensor notation,

$$\tau_{ij}^* = \lambda V_{k,k}^*\delta_{ij} + \mu(V_{i,j}^* + V_{j,i}^*), \tag{5}$$

where $\mu$ is the dynamic viscosity of the mixture and $\lambda$ is the bulk viscosity. Mass diffusion is due only to pressure and concentration gradients. With the use of the Onsager reciprocal relations and the Gibbs–Duhem relation, the mass diffusion equation may be written

$$\mathbf{J}^* = -\rho^* D_{ab}\left[\operatorname{grad} C^* + \frac{\beta M_a}{\rho_b R_g \theta}(1+MC^*)(1-C^*)C^* \operatorname{grad} P^*\right], \tag{6}$$

where $D_{ab}$ is the binary diffusion coefficient, considered constant; $R_g$ is the universal gas constant; $\theta$ is the temperature; $M_a$ is the molecular weight of component $a$; and $M = (M_b - M_a)/M_a$. The viscosity, assumed to be a function of concentration alone, is given by an Eyring formula (see, for example, Hirschfelder, Curtiss & Bird 1954):

$$\log \mu = \xi_a \log \mu_a + \xi_b \log \mu_b, \tag{7}$$

where $\mu_a$ and $\mu_b$ are the viscosities of the two pure components and $\xi_a$ and $\xi_b$ are their mole fractions. In terms of mass fractions, we have

$$\log \mu = \log \mu_b + \left[\frac{M_b}{M_a} C^* \Big/ (1+MC^*)\right] \log (\mu_a/\mu_b). \tag{8}$$

For specific mixtures, a polynomial correction in powers of $C^*$ can be added to (8) to give a better fit. Kulinski (1966) gave an example of this for a water–glycerol mixture.

The non-dimensionalization of the governing equations is accomplished by reference to a circular Couette flow in the annulus of two rotating cylinders with inner and outer radii of $R_1$ and $R_2$, respectively, and inner and outer angular velocities $\Omega_1$ and $\Omega_2$. The properties of the mixture are referenced to the homogeneous (well-mixed) values of concentration, density, and viscosity. We define $\hat{C}$ as the mass fraction of component $a$ in the thoroughly mixed solution with $\hat{\mu} = \hat{\mu}(\hat{C})$ and $\hat{\rho} = \hat{\rho}(\hat{C})$. Then

$$\left.\begin{array}{l} C = C^*, \quad \mathbf{J} = \mathbf{J}^* R_2/D_{ab}\hat{\rho}, \quad P = P^*/\hat{\rho} R_2^2 \Omega_1^2, \\ \mathbf{V} = \mathbf{V}^*/R_2 \Omega_1, \quad r = r^*/R_2, \quad t = t^* \Omega_1, \\ \mu = \mu^*/\hat{\mu}, \quad \rho = \rho^*/\hat{\rho}, \quad \boldsymbol{\tau} = \boldsymbol{\tau}^* \hat{\mu} \Omega_1, \end{array}\right\} \tag{9}$$

where asterisks denote dimensional quantities.

Upon substitution, (1) to (4) and (6) become

$$\frac{\beta \hat{\rho}}{\rho_b} \rho \frac{DC}{Dt} - \operatorname{div} \mathbf{V} = 0, \tag{10}$$

$$\operatorname{div}(\mathbf{V} + \epsilon \mathbf{J}) = 0, \tag{11}$$

$$\rho \frac{D\mathbf{V}}{Dt} = -\operatorname{grad} P + \frac{1}{T^{\frac{1}{2}}} \operatorname{div} \boldsymbol{\tau}, \tag{12}$$

$$\mathbf{J} = -\rho(\operatorname{grad} C + \Phi T f \operatorname{grad} P), \tag{13}$$

$$\rho = \frac{1+\beta \hat{C}}{1+\beta C}. \tag{14}$$

Two non-dimensional groups are recognized as

$$S = \hat{\nu}/D_{ab}\text{: Schmidt number}$$

and

$$T = R_2^4 \Omega_1^2/\hat{\nu}^2\text{: Taylor number,}$$

where the definition of $T$ follows that of Sparrow *et al.* (1964). In addition, we introduce the notation

$$\epsilon = \frac{\beta \hat{\rho} D_{ab}}{\rho_b R_2^2 \Omega_1} = \frac{\beta \hat{\rho}}{\rho_b S \sqrt{T}}\text{: inverse diffusion Reynolds number,}$$

$$\Phi = \frac{\beta \hat{\rho} M_a \hat{\nu}^2}{\rho_b R_g \theta R_2^2}\text{: separation potential,}$$

$$f = (1+MC)(1-C)C.$$

The roles of $\Phi$ and $\epsilon$ are discussed in § 3 and 4.

## 3. Basic flow

The basic flow of the system is steady and in thermodynamic equilibrium. If variations in tangential and axial position are also neglected in (10) to (13) and the condition of $\mathbf{J} = 0$ for equilibrium is applied, the general flow equations reduce to

$$dC_0/dr = -\Phi T f_0 \rho_0 r \Omega_0^2, \tag{15}$$

$$\frac{d}{dr}\left[\mu_0 r^3 \frac{d\Omega_0}{dr}\right] = 0, \tag{16}$$

where subscript 0 denotes basic flow quantities, $r$ is the radial co-ordinate, and $\rho_0$ and $\mu_0$ are given by (14) and (8) with $C = C_0$. Equation (15), which results from the condition of thermodynamic equilibrium, states that, when steady state has been reached, the diffusion due to a concentration gradient just balances the baro-diffusion term. A comparison with equations and relationships that govern specie separation in a centrifuge suggest that $\Phi$ is proportional to the ratio of the sedimentation coefficient to diffusivity, hence the net separation potential. Consequently, we refer to $\Phi$ as the separation potential.

The non-dimensional boundary conditions are

$$\Omega_0 = 1 \quad \text{at} \quad r = R_1/R_2,$$

$$\Omega_0 = \Omega_2 \quad \text{at} \quad r = 1,$$

$$\int_{R_1/R_2}^{1} C_0 r \, dr = \tfrac{1}{2}\hat{C}[1-(R_1/R_2)^2].$$

The first two conditions are apparent from the geometry, while the third expresses a macroscopic conservation of species. It should be noted that, in the usual case of constant viscosity, (16) can be integrated immediately to give $\Omega_0 = A + B/r^2$. However, since viscosity is dependent on concentration, the flow field will be concentration-dependent.

Equations (15) and (16) were integrated numerically, and the results compare favourably with Kulinski (1966) when the notation is adjusted. We have chosen the average concentration of the mixture, $\hat{C}$, rather than the concentration at the inner cylinder, as the controlling parameter. In addition, Kulinski chose the case of the stationary inner cylinder, which is stable to the Taylor type of instability. All the basic flow calculations were conducted with the velocity ratio $\Omega_2/\Omega_1$ equal to 0·53 and a radius ratio $R_1/R_2$ equal to 0·75. These are typical values used by Sparrow *et al.* (1964) for the homogeneous case. In addition, they offer a fair chance for significant specie separation. Figure 1 illustrates the effect of $\Phi T$ on the concentration distribution with all other parameters held constant. The relatively small amount of centrifuging is due to our choice of liquids (e.g. a mixture of 85 % glycerine and 15 % tetrabromoethane has a separation potential $\Phi = 0{\cdot}7 \times 10^{-8}$ at $T = 0{\cdot}2 \times 10^7$). As subsequent results show, very little separation is needed to demonstrate the effects of density stratification on the hydrodynamic stability.

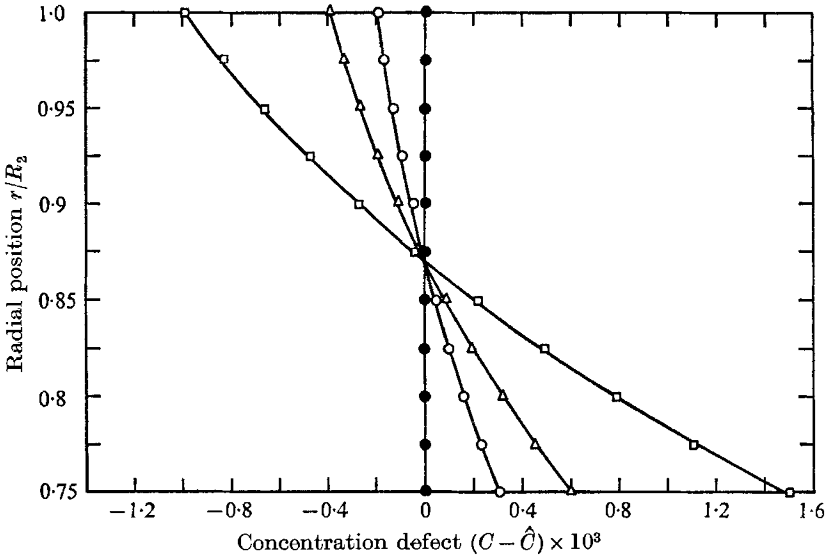

FIGURE 1. Change in concentration as a function of radius for different values of $\Phi T$ where $\mu_a/\mu_b = 50$, $\beta = 1{\cdot}35$, $M = 2{\cdot}75$, $\hat{C} = 0{\cdot}85$, $\rho_a = 1{\cdot}26$, and $\rho_b = 2{\cdot}96$. $\Phi T$: ●, 0; ○, 0·01; △, 0·02; □, 0·05.

Substantial concentration dependence of the velocity profile can be expected for large viscosity ratios. Figure 2 illustrates the effects of viscosity changes on the velocity profile when the Eyring formula, (7), is used. Component $a$ is always the lighter fluid and the angular velocity $\Omega_s$ is the standard constant viscosity profile $\Omega_s = A + B/r^2$. When $\mu_a/\mu_b > 1$, the mixture is more viscous toward the inner cylinder, giving rise to a larger velocity gradient in the radial direction, thus producing the 'positive' bulge. For this case, the velocity distribution shifts toward solid body rotation. If the reversal is assumed, i.e. $\mu_a/\mu_b < 1$ (conditions are unchanged except that the heavy fluid is more viscous), the mixture has a higher viscosity toward the outer cylinder and a lower velocity gradient. Figure 3 illustrates this result on the actual velocity profile with a large value of $\Phi T$.

Finally, the variation in density is of the same order as the variation in concentration, except in the opposite sense. When the quantity $C-\hat{C}$ is small, the density equation (14) can be rewritten

$$\rho - 1 \simeq -\frac{\beta}{1+\beta\hat{C}}(C-\hat{C}). \tag{17}$$

Actually, it is the small density change that produces the effects outlined in the following sections.

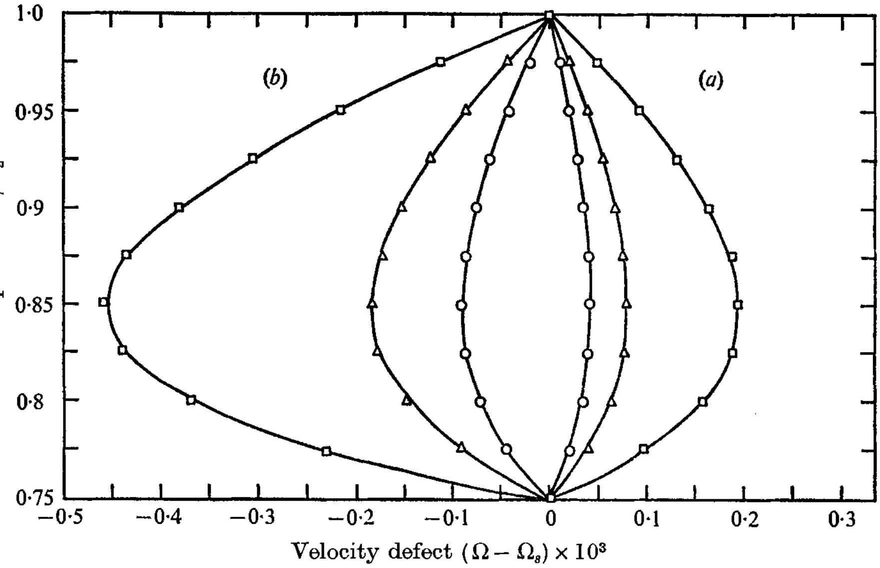

FIGURE 2. Change in angular velocity as a function of radius for different values of $\Phi T$: ○, 0·01; △, 0·02; □, 0·05. Side (*a*): $\mu_a/\mu_b = 50$, $\beta = 1{\cdot}35$, $M = 2{\cdot}75$, $\hat{C} = 0{\cdot}85$. Side (*b*): $\mu_a/\mu_b = 0{\cdot}02$, $\beta = 1{\cdot}35$, $M = 2.75$, $\hat{C} = 0{\cdot}15$.

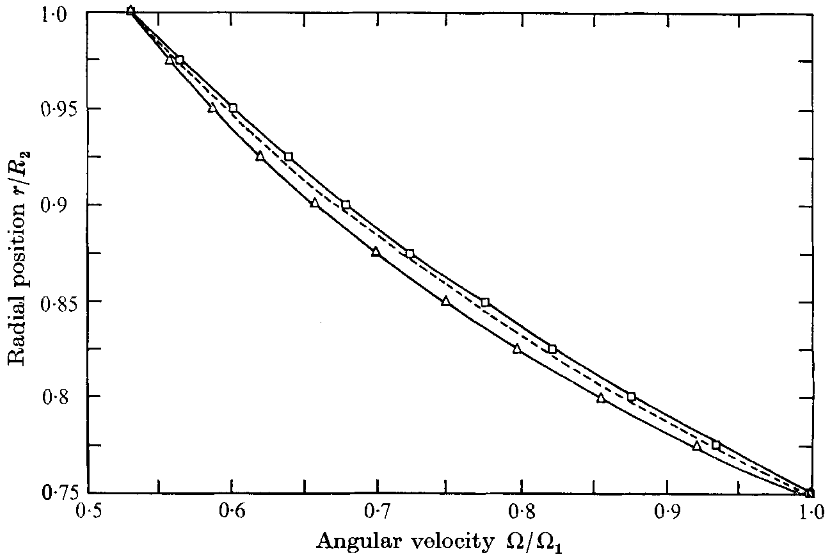

FIGURE 3. Angular velocity as a function of radius for different viscosity ratios. △, $\Phi T = 2$, $\mu_a/\mu_b = 0{\cdot}02$, $\hat{C} = 0{\cdot}15$; □, $\Phi T = 2$, $\mu_a/\mu_b = 50$, $\hat{C} = 0{\cdot}85$; - - - -, $\mu_a/\mu_b = 1$

## 4. Perturbation equations

To analyze the stability of the system of equations (10) to (14) with respect to infinitesimal, axisymmetric disturbances, we express each dependent variable as a sum of a stationary value (basic flow) and a perturbation quantity. We let

$$\left.\begin{aligned} &V_r = u', \quad V_\theta = r\Omega_0 + v', \quad V_z = w', \\ &P = P_0 + p', \quad J_r = j_r', \quad J_z = j_z', \\ &C = C_0 + c', \quad \rho = \rho_0 + \rho', \quad \mu = \mu_0 + \mu'. \end{aligned}\right\} \tag{18}$$

Equations (18) are substituted into the axisymmetric form of (10) to (14), the squares of perturbation quantities are neglected, and the basic flow is subtracted to give

$$\frac{\hat{\rho}}{\rho_b}\beta\rho_0\left(\frac{\partial c'}{\partial t} + u'\frac{dC_0}{dr}\right) - \left(\frac{1}{r}\frac{\partial ru'}{\partial r} + \frac{\partial w'}{\partial z}\right) = 0, \tag{19a}$$

$$\frac{1}{r}\frac{\partial ru'}{\partial r} + \frac{\partial w'}{\partial z} + \epsilon\left(\frac{1}{r}\frac{\partial rj_r'}{\partial r} + \frac{\partial j_z'}{\partial z}\right) = 0, \tag{19b}$$

$$\begin{aligned} \rho_0\left(\frac{\partial u'}{\partial t} - 2\Omega_0 v'\right) - \rho' r\Omega_0^2 = -\frac{\partial p'}{\partial r} + \frac{1}{T^{\frac{1}{2}}}\left\{\frac{\partial}{\partial r}\left[2\mu_0\frac{\partial u'}{\partial r} + \lambda_0\left(\frac{1}{r}\frac{\partial ru'}{\partial r} + \frac{\partial w'}{\partial z}\right)\right]\right. \\ \left. + \mu_0\frac{\partial}{\partial z}\left(\frac{\partial u'}{\partial z} + \frac{\partial w'}{\partial r}\right) + \frac{2\mu_0}{r}\left(\frac{\partial u'}{\partial r} - \frac{u'}{r}\right)\right\}, \end{aligned} \tag{19c}$$

$$\begin{aligned} \rho_0\left(\frac{\partial v'}{\partial t} + u'\frac{dr\Omega_0}{dr} + u'\Omega_0\right) = \frac{1}{T^{\frac{1}{2}}}\left\{\frac{\partial}{\partial r}\left[\mu_0\left(\frac{\partial v'}{\partial r} - \frac{v'}{r}\right) + \mu'\left(r\frac{d\Omega_0}{dr}\right)\right]\right. \\ \left. + \mu_0\frac{\partial^2 v'}{\partial z^2} + 2\mu'\frac{d\Omega_0}{dr} + 2\frac{\mu_0}{r}\left(\frac{\partial v'}{\partial r} - \frac{v'}{r}\right)\right\}, \end{aligned} \tag{19d}$$

$$\rho_0\frac{\partial w'}{\partial t} = -\frac{\partial p'}{\partial z} + \frac{1}{T^{\frac{1}{2}}}\left\{\frac{1}{r}\frac{\partial}{\partial r}\left[\mu_0 r\left(\frac{\partial u'}{\partial z} + \frac{\partial w'}{\partial r}\right)\right] + 2\mu_0\frac{\partial^2 w'}{\partial z^2} + \lambda_0\frac{\partial}{\partial z}\left(\frac{1}{r}\frac{\partial ru'}{\partial r} + \frac{\partial w'}{\partial z}\right)\right\}, \tag{19e}$$

$$j_r' = -\rho_0\left(\frac{\partial c'}{\partial r} + f_0\frac{\partial p'}{\partial r} + f'\frac{dP_0}{dr}\right), \tag{19f}$$

$$j_z' = -\rho_0\left(\frac{\partial c'}{\partial z} + f_0\frac{\partial p'}{\partial z}\right). \tag{19g}$$

A solution of (19) will be sought in terms of a superposition of normal modes of the form

$$u'(r,z,t) = u(r)\exp(i\alpha z + \sigma t), \tag{20}$$

where $\sigma$ is generally complex. The other perturbation quantities are represented in a similar functional form. The $r$-dependent function of a perturbation quantity uses the same symbol without the prime. Furthermore, since $\rho$, $\mu$ and $f$ are known functions of $C$, they are expanded about $C_0$ and their perturbations are replaced by perturbations in $C$. Hence,

$$\rho' = -\beta\frac{\hat{\rho}}{\rho_b}\rho_0^2 c', \quad \mu' = \frac{d\mu_0}{dC_0}c', \quad f' = \frac{df_0}{dC_0}c'. \tag{21}$$

The substitution of (20) into (19) leaves only derivatives with respect to $r$ which are represented by the notation:

$$D = \frac{d}{dr}, \quad D_* = \frac{d}{dr} + \frac{1}{r}. \tag{22}$$

Substituting (20), (21), and (22) into (19) and eliminating $j_r'$ and $j_z'$ from (19$b$) by the use of (19$f$) and (19$g$), we obtain the governing differential equations for the perturbations:

$$\frac{\beta\hat{\rho}}{\rho_b}\rho_0(\sigma c + uDC_0) = D_* u + i\alpha w, \tag{23$a$}$$

$$D_* u + i\alpha w = \epsilon\left\{D_*\left[\rho_0\left(Dc + f_0 Dp - \frac{Df_0}{f_0}DC_0 c\right)\right] - \alpha^2\rho_0(c + f_0 p)\right\}, \tag{23$b$}$$

$$\begin{aligned}\rho_0\left(\sigma u - 2\Omega_0 v + \frac{\beta\hat{\rho}}{\rho_b}\rho_0 r\Omega_0^2 c\right) = -Dp + \frac{1}{T^{\frac{1}{2}}}\Big\{&D(2\mu_0 Du) + D[\lambda_0(D_* u + i\alpha w)] \\ &+ \mu_0 i\alpha(Dw + i\alpha u) + 2\frac{\mu_0}{r}(Du - u/r)\Big\},\end{aligned} \tag{23$c$}$$

$$\begin{aligned}\rho_0(\sigma v + urD_*\Omega_0 + u\Omega_0) = \frac{1}{T^{\frac{1}{2}}}\Big\{&D\left[\mu_0(Dv - v/r) + \frac{df_0}{dC_0}rD\Omega_0 c\right] - \mu_0\alpha^2 v \\ &+ 2\frac{df}{dC_0}D\Omega_0 c + 2\frac{\mu_0}{r}(Dv - v/r)\Big\},\end{aligned} \tag{23$d$}$$

$$\rho_0\sigma w = -i\alpha p + \frac{1}{T^{\frac{1}{2}}}\{D_*[\mu_0(Dw + i\alpha u)] - 2\mu_0\alpha^2 w + \lambda_0 i\alpha(D_* u + i\alpha w)\}. \tag{23$e$}$$

The dominant dimensionless groups in (23) are the Taylor number and the Schmidt number. For most liquid mixtures, $S \sim 10^5 - 10^6$ while $T^{\frac{1}{2}} \sim 10^3$. Therefore, since $\beta \sim 10^0$ and $\epsilon \sim 10^{-8} - 10^{-9}$ the right-hand side of equation (23$b$) is very small. We are, therefore, interested in the limit as $\epsilon \to 0$, i.e. we restrict the density changes to pure convective changes. The density changes are carried out by the perturbation velocities alone, since the momentum diffusivity $\nu$ is much larger than the mass diffusivity $D_{ab}$. The limit $\epsilon \to 0$ implies, however, a singular perturbation and a first-order solution obtained by putting $\epsilon = 0$ is valid only for small time. This is precisely the region of interest in linear stability since only the initial time behaviour is sought. It should be noted that the classical linearization of (1) to (3) is, in itself, a singular perturbation valid for small times only.

Indeed, several earlier investigators of stability of stratified flow (cf. Yih 1965) neglected mass diffusivity. Kulinski (1966), in his analysis of the transient Couette flow problem of a heterogeneous fluid, compares the short time scale required for establishing the velocity profile to the long mass diffusion scale required for the final concentration profile.

Letting $\epsilon \to 0$ and eliminating $c$, $p$, and $w$ from (23), we can write the perturbation equations as

$$\begin{aligned}(2\rho_0\Omega_0)v = \rho_0\sigma&\left(1 + \frac{\beta\hat{\rho}}{\rho_b}f_0\omega^2/\sigma^2\right) - \frac{\sigma}{\alpha^2}D(\rho_0 D_* u) \\ &+ \frac{1}{\alpha^2 T^{\frac{1}{2}}}(DD_* - \alpha^2)[\mu_0(DD_* - \alpha^2)]u - 2\alpha^2[D(\eta f\omega)u],\end{aligned} \tag{24$a$}$$

$$\rho_0\sigma v = -\rho_0(D_* r\Omega_0)u + \frac{1}{T^{\frac{1}{2}}}\{\mu_0(DD_* - \alpha^2)\, v - \eta f_0\,\omega(Dv - v/r)\}$$
$$+\frac{1}{T^{\frac{1}{2}}}\left\{\frac{1}{r^2}D\left[(r^3\eta f_0\,\omega D\Omega_0)\frac{u}{\sigma}\right]\right\}, \qquad (24b)$$

where $$\omega(r) = \rho_0 r\Omega_0^2 \qquad (25)$$

and $$\eta(r) = d\mu_0/dC_0. \qquad (26)$$

The boundary conditions governing the differential equations are, to order $\epsilon$, given by

$$u = Du = v = 0 \quad \text{at} \quad r = R_1/R_2, 1. \qquad (27)$$

The condition on the concentration perturbation

$$\int_{R_1/R_2}^{1} cr\,dr = 0 \qquad (28)$$

cannot be satisfied because of the nature of the singular perturbation implied by $\epsilon \to 0$. However, the conditions that $c = 0$ at $r = R_1/R_2$ and 1 are satisfied by (23*a*), and Kulinski (1966) shows that (28) is satisfied to order $[C_0(1) - C_0(R_1/R_2)]$ in this case. The calculations shown in figure 1 illustrate that this is adequate.

Equations (24) may be analyzed more conveniently by rewriting them in terms of a sixth-order equation in the complex variable $u$ alone. In operator form, we can write this single equation in $u$ as

$$L_1\sigma^2 u + L_2\sigma u + L_3 u + L_4(1/\sigma)\, u = 0, \qquad (29)$$

where $L_1$, $L_2$, $L_3$ and $L_4$ are second, fourth, sixth, and second order, respectively and are given as

$$L_1 = \rho_0 T\left(DD_* - \alpha^2 + \frac{D\rho_0}{\rho_0}D_*\right),$$
$$L_2 = -T^{\frac{1}{2}}\Bigg\{(DD_* - \alpha^2)\,[\mu_0(DD_* - \alpha^2)] + 2\alpha^2 D^2\mu_0$$
$$-2\Omega_0\,[\mu_0(DD_* - \alpha^2) + (D\mu_0)\,(D - 1/r)]\left[\frac{1}{2\Omega_0}\left(DD_* - \alpha^2 + \frac{D\rho_0}{\rho_0}D_*\right)\right]\Bigg\},$$
$$L_3 = 2\Omega_0[\mu_0(DD_* - \alpha^2) + (D\mu_0)\,(D - 1/r)]$$
$$\times\left\{\frac{1}{2\rho_0\Omega_0}(DD_* - \alpha^2)[\mu_0(DD_* - \alpha^2)] + \frac{1}{2\rho_0\Omega_0}(2\alpha^2 D^2\mu_0)\right\}$$
$$-\alpha^2 T\left[\rho_0\beta\frac{\hat{\rho} f_0\omega^2}{\rho_b} + 2\rho_0\Omega_0(D_* r\Omega_0)\right];$$
$$L_4 = 2\Omega_0\alpha^2\,T^{\frac{1}{2}}\Bigg\{[\mu_0(DD_* - \alpha^2) + D\mu_0(D - 1/r)]\left(\frac{\beta\hat{\rho} f_0\omega^2}{2\rho_b\Omega_0}\right)$$
$$-\frac{1}{r^2}D(r^3 D\Omega_0 D\mu_0) - rD\Omega_0 D\mu_0 D\Bigg\}.$$

The coefficients of the operators are functions of the basic flow and the axial wavelength. Hence, they are real but not constant. If one takes the case of no density change ($\beta = 0$), $L_4$ is identically equal to zero. Since $L_3$ has the form of the classical Taylor problem in this case, (24) reduces to the problem discussed by Sparrow *et al.* (1964) with $\sigma \equiv 0$.

When density changes are included, a non-oscillatory neutral solution with $\sigma \equiv 0$ is not possible because of the singularity. We must then consider both real and imaginary parts of $\sigma$ and real and imaginary parts of $u$. This is a direct result of the neglect of diffusion because the density perturbations can only relax themselves through the formation of an axial wave. When

$$\sigma = \sigma_r + i\sigma_i, \tag{30}$$

the exponential factor in the perturbation variables can be rewritten

$$[\exp(\sigma_r t)][\exp i(\alpha z + \sigma_i t)], \quad \text{and} \quad (\alpha z + \sigma_i t)$$

can be considered as a wave form or oscillation. Because heterogeneous shear flow problems are known to have oscillatory solutions, the complete formulation requires consideration of real and imaginary parts of the velocities

$$u = u_r + iu_i, \quad v = v_r + iv_i. \tag{31}$$

After substitution of (30) and (31) into (24) and (27), a twelfth-order differential system of real functions results.

$$\begin{aligned} &\{(DD_* - \alpha^2)[\mu_0(DD_* - \alpha^2)] + 2\alpha^2 D^2\mu_0\}\begin{pmatrix} u_r \\ u_i \end{pmatrix} - T^{\frac{1}{2}}[D(\rho_0 D_*) - \rho_0\alpha^2] \\ &\qquad \times \begin{pmatrix} \sigma_r u_r - \sigma_i u_i \\ \sigma_i u_r + \sigma_r u_i \end{pmatrix} + \frac{\alpha^2 T^{\frac{1}{2}}\rho_0\beta\hat{\rho}f_0\omega^2}{\rho_b(\sigma_r^2 + \sigma_i^2)}\begin{pmatrix} \sigma_r u_r + \sigma_i u_i \\ -\sigma_i u_r + \sigma_r u_i \end{pmatrix} = \alpha^2 T^{\frac{1}{2}} 2\rho_0\Omega_0\begin{pmatrix} v_r \\ v_i \end{pmatrix}, \end{aligned} \tag{32}$$

$$\begin{aligned} &[\mu_0(DD_* - \alpha^2) + D\mu_0(D - 1/r)]\begin{pmatrix} v_r \\ v_i \end{pmatrix} - \rho_0 T^{\frac{1}{2}}\begin{pmatrix} \sigma_r v_r - \sigma_i v_i \\ \sigma_i v_r + \sigma_r v_i \end{pmatrix} \\ &\qquad = \rho T^{\frac{1}{2}}(D_* r\Omega_0)\begin{pmatrix} u_r \\ u_i \end{pmatrix} + \frac{1}{r^2}D\left[r^3 D\mu_0 D\Omega_0\begin{pmatrix} \sigma_r u_r + \sigma_i u_i \\ -\sigma_i u_r + \sigma_r u_i \end{pmatrix}\right]. \end{aligned} \tag{33}$$

The boundary conditions are

$$u_r = Du_r = v_r = u_i = Du_i = v_i = 0 \quad \text{at} \quad r = R_1/R_2, 1. \tag{34}$$

The problem now is to find the values of $T$ and $\sigma_i$ which will lead to the solution of (32) and (33), once $\alpha$, $\sigma_r$, $\Phi$, $R_1/R_2$, and $\Omega_2/\Omega_1$ are prescribed. A minimum value of $T$ can be found by varying the parameters $\alpha$ and $\sigma_r$. Of course, any two of these parameters can be considered eigenvalues of the differential equation. However, $\Phi$, $R_1/R_2$, and $\Omega_2/\Omega_1$ are physical constants; the values of $\alpha$ for minimum $T$ will be close to those found by Sparrow *et al.* (1964), and $\sigma_r$ will generally be zero. Therefore, $T$ and $\sigma_i$ are the logical choice for independent eigenvalues.

The solution of the system of equations (32) and (33) with boundary conditions (34) follows Lanczos (1961) and is essentially the same technique employed by Sparrow *et al.* (1964) and Harris & Reid (1964). It involves choosing six linearly independent trial values each of $D^2u_r$, $D^2u_i$, $D^3u_r$, $D^3u_i$, $Dv_r$, $Dv_i$ at $r = R_1/R_2$, and generating six linearly independent solutions, $u^j$ ($j = 1, \ldots, 6$), by forward integration. To satisfy the boundary conditions (34) at $r = 1$, a system of six homogeneous algebraic equations of the form $\xi_j \Lambda_k^j = 0$ results. The coefficients in the equations, $\Lambda^j_k$, consist of the six trial solutions ($j$) described and the six boundary conditions ($k$) of the form

$$\Lambda^j_1 = u^j_r(1), \quad \Lambda^j_2 = u^j_i(1), \quad \Lambda^j_3 = Du^j_r(1), \quad \ldots,$$

$$\Lambda^j_6 = v^j_i(1).$$

The determinant of the coefficients, det $(\Lambda^j_k)$, will vanish if, and only if, the eigenvalues have been properly chosen. Hence, to obtain the eigenvalues we must find the zero of the function

$$\det = \det(T, \sigma_i) \tag{35}$$

after the other parameters, i.e. $\alpha$, $\sigma_r$, $\Omega_2/\Omega_1$, $R_1/R_2$, $\beta$, $M$, $\Phi$, $\hat{C}$ and $\mu_a/\mu_b$, have been specified. Equation (35) is calculated as a surface which touches the det $= 0$ plane only at unique values of $T$ and $\sigma_i$. The calculation procedure uses a Gauss elimination technique that continuously renormalizes and diagonalizes the determinant. A numerical check on the accuracy of the results was performed by repeating a typical calculation on two computers having different accuracies. The first was an IBM 360-40 with six significant figures and the second a CDC-6600 with 31 significant figures. The results agree to three places.

## 5. Results and discussion

Equations (32) and (33) were integrated for a number of different conditions. The results of one such calculation are shown qualitatively in figure 4 for

$$\Phi = 0{\cdot}7 \times 10^{-8}.$$

Solutions exist only for the particular combinations of $T$, $\sigma_i$, and $\sigma_r$ given by this curve. The homogeneous solution is given by the $\Phi = 0$ curve and lies wholly in the $\sigma_r - T$ plane. The departure from aperiodic motion occurs in the neighbourhood of $\sigma_r = 0{\cdot}015$. The oscillation frequency appears to decrease asymptotically towards zero with $\sigma_i = 0{\cdot}0001$ being the cutoff value with which to determine any substantial change with $\sigma_i$. As the neutral point ($\sigma_r = 0$) is approached, the Taylor number decreases and the frequency increases to a maximum value of 0·0112.

The oscillatory motion is due mainly to the restoring force of the pressure gradient in the presence of a stable density stratification. When a lighter fluid particle is displaced toward the heavier fluid, the pressure gradient drives it back toward the centre and vice versa. In the absence of diffusion, either in this case or in other cases of density stratification, (23*a*) shows that density perturbations must be carried along disturbance streamlines in the radial direction and cannot be transmitted axially. Hence, when the radial velocity disturbance is positive, buoyancy effects tend to drive the particles toward the centre and, when the radial velocity is negative, the particles are driven toward the outside. These restoring forces coupled with the axial periodicity of the radial velocity give rise to the oscillations.

Figure 5 is a projection of the $T = T(\sigma_r, \sigma_i)$ curve on the $T - \sigma_r$ plane. At a Taylor number of $27{\cdot}0 \times 10^5$, $\Phi = 0{\cdot}7 \times 10^{-8}$ corresponds to a density change of 0·05 % and $\Phi = 2{\cdot}5 \times 10^{-8}$ corresponds to a density change of 0·25 %. These modest changes in density decrease the growth rate by, respectively, 15 % and 45 %.

The intersections of the dotted lines with the solid lines indicate where $\sigma_i \neq 0$. If $\sigma_i$ were forced to equal zero, the particular curves would follow the dotted lines and become singular at $\sigma_r = 0$.

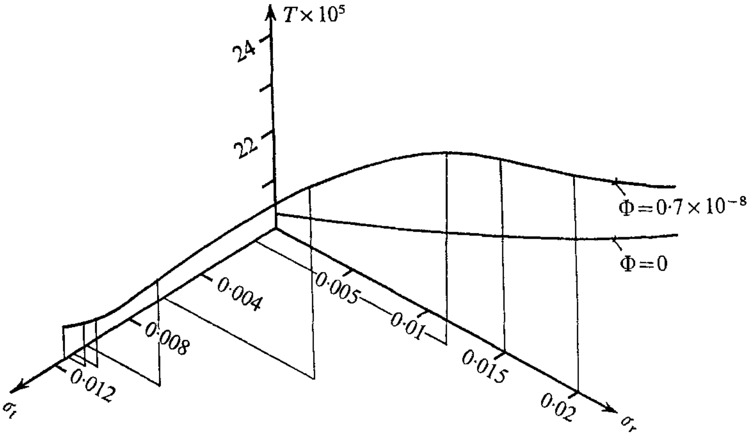

Figure 4. Taylor number variation with $\sigma_r$ and $\sigma_i$.

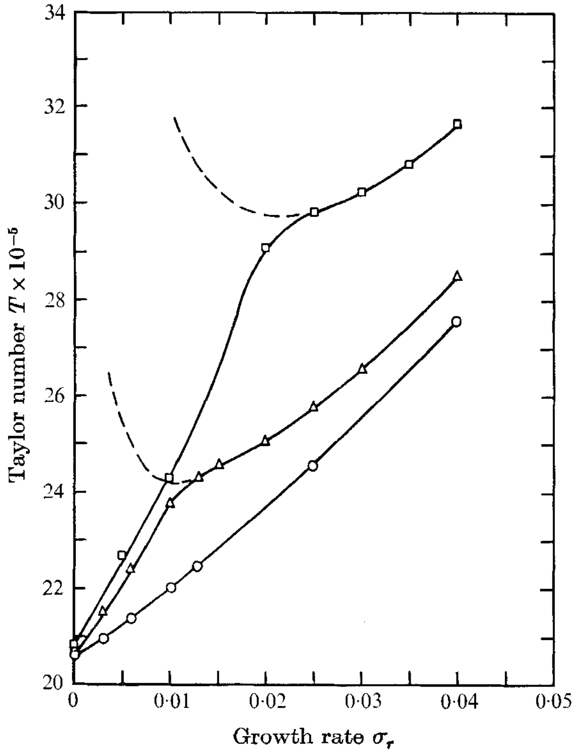

Figure 5. Taylor number as a function of growth rate with constants of figure 1.
○, $\Phi = 0$; △, $\Phi = 0{\cdot}7 \times 10^{-8}$; □, $\Phi = 2{\cdot}5 \times 10^{-8}$.

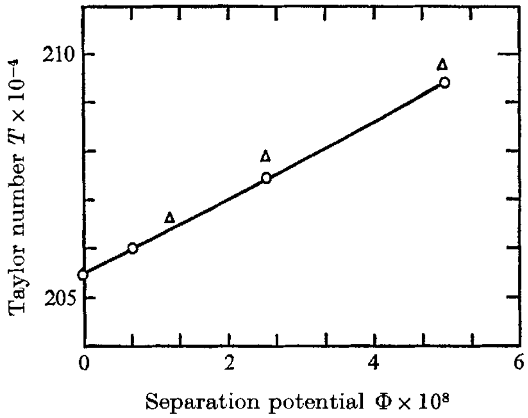

FIGURE 6. Critical Taylor number as a function of separation potential with $\sigma_r = 0$ and the constants of figure 1. △, $\mu_a/\mu_b = 75$; ○, $\mu_a/\mu_b = 50$.

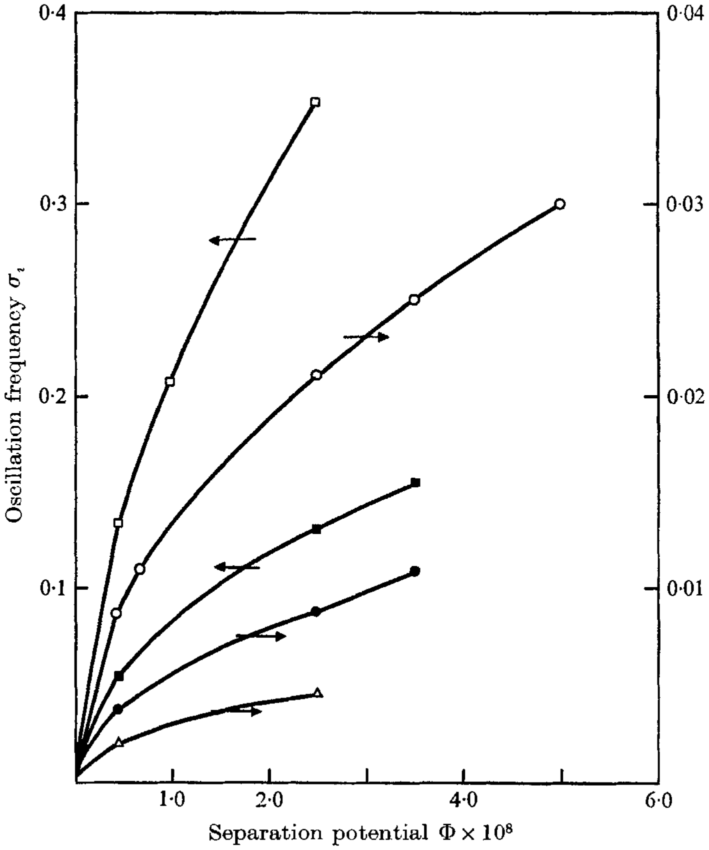

FIGURE 7. Critical oscillation frequency as a function of separation potential for different radius ratios with $\sigma_r = 0$ and the constants of figure 1. □, $R_1/R_2 = 0{\cdot}95$, $\Omega_2/\Omega_1 = 0{\cdot}85$; ■, $R_1/R_2 = 0{\cdot}95$, $\Omega_2/\Omega_1 = 0{\cdot}68$; ○, $R_1/R_2 = 0{\cdot}75$, $\Omega_2/\Omega_1 = 0{\cdot}53$; ●, $R_1/R_2 = 0{\cdot}75$, $\Omega_2/\Omega_1 = 0{\cdot}42$; △, $R_1/R_2 = 0{\cdot}5$, $\Omega_2/\Omega_1 = 0{\cdot}24$.

The critical Taylor number of the homogeneous case, $T_s$ and its corresponding critical wave-number $\alpha_s$ have been tabulated by Sparrow *et al.* (1964). The critical wave-number $\alpha$ for which $T$ is a minimum, experiences some variation from $\alpha_s$ as the curves move away from the neutral point and the homogeneous curve. The dominant $\alpha$ term in (32) and (33) is $\alpha^2 T$. After a $T_{\text{new}}$ has been determined on the basis of $\alpha_s$, the corresponding $\alpha_{\text{new}}$ is given approximately by $\alpha_{\text{new}} = \alpha_s(T_s/T_{\text{new}})^{\frac{1}{2}}$. This approximation eventually minimizes $T_{\text{new}}$, which is rather insensitive to small changes in wave-number. Therefore, the final value of $T_{\text{new}}$ is close to that found by using $\alpha_s$, even though $\alpha_{\text{new}}$ may change by 10 %.

Figure 6 shows that for $\mu_a/\mu_b > 1$, the critical Taylor number increases with separation potential and with viscosity ratio. The stabilizing effect of higher viscosity ratios is due to the fact that the velocity profile tends more toward solid body rotation, i.e. a more stable circulation distribution as shown in figure 3.

Figure 7 shows the change in the corresponding critical oscillation frequency with separation potential for different radius ratios, $R_1/R_2$, and velocity ratios, $\Omega_2/\Omega_1$. The values for $R_1/R_2 = 0.95$ are generally an order of magnitude higher than those for $R_1/R_2 = 0{\cdot}75$. The critical Taylor number at $R_1/R_2 = 0{\cdot}95$ varies from $1{\cdot}376 \times 10^8$ $(\Phi = 0)$ to $1{\cdot}805 \times 10^8$ $(\Phi = 2{\cdot}5 \times 10^{-8})$; a change of more than 31 %. These large changes are due to the greater magnitude of Taylor number at this radius ratio. Representative density changes for $R_1/R_2 = 0.95$ are of the order of 5 %.

## 6. Conclusion

We have shown that, in the absence of diffusion, a mixture of two fluids in a circular Couette flow geometry exhibits a new kind of Taylor instability to axisymmetric linear disturbances.This instability is characterized by the formation of axial waves as the fundamental unstable mode.

The presence of small density stratification (approximately 0.05 % change across the gap) due to centrifuging causes (i) oscillations at frequencies that increase with increased component separation; (ii) a stabilizing effect when density changes alone are considered; and (iii) a stabilizing effect when the lighter fluid is more viscous and a destabilizing effect when the converse is true.

The solution technique developed is able to predict quantitatively the stability of a two-component rotating Couette flow for any gap size, for any angular velocity ratio, and for any liquid mixture where the behaviour of the density and the viscosity is consistent with the governing assumptions.

We speculate that the secondary unstable modes, which include asymmetric waves, would be shifted substantially from the fundamental mode because the growth rate is much lower than in the corresponding homogeneous case. The next step should be the inclusion of asymmetric waves. This would test the axisymmetric assumption and would determine the limit of negative angular velocity ratio for axisymmetric disturbances.

This work was supported by the National Science Foundation under Grant GK-911 and by the U.S. Atomic Energy Commission. The authors are grateful to Dr E. S. Kulinski for discussions on the paper.

## REFERENCES

CHANDRASEKHAR, S. 1961 *Hydrodynamic and Hydromagnetic Stability*. Oxford University Press.

DEGROOT, S. R. & MAZUR, P. 1962 *Non-equilibrium Thermodynamics*. North Holland.

HARRIS, D. L. & REID, W. H. 1964 On the stability of viscous flow between rotating cylinders. Part 2. Numerical analysis. *J. Fluid Mech.* **20**, 95.

HIRSCHFELDER, J. O., CURTISS, C. F. & BIRD, R. B. 1954 *Molecular Theory of Gases and Liquids*. John Wiley.

KRUEGER, E. R., GROSS, A. & DIPRIMA, R. C. 1966 On the relative importance of Taylor-vortex and non-axisymmetric modes in flow between rotating cylinders. *J. Fluid Mech.* **24**, 521.

KULINSKI, E. S. 1966 Rotating Couette flow of a binary liquid. *Phys. Fluids*, **9**, 1207.

LANCZOS, G. 1961 *Linear Differential Operators*, chap. 9. Van Nostrand.

LIN, C. C. 1955 *The Theory of Hydrodynamic Stability*. Cambridge University Press.

SARIC, W. S. 1968 Hydrodynamic stability of heterogeneous Couette flows. Ph.D. thesis, Illinois Institute of Technology, Chicago, Illinois.

SNYDER, H. A. 1968 Stability of rotating Couette flow. I. Asymmetric wave forms. *Phys. Fluids*, **11**, 728.

SPARROW, E. C., MUNRO, W. D. & JONSSON, V. K. 1964 Instability of the flow between rotating cylinders – the wide gap problem. *J. Fluid Mech.* **20**, 35.

TAYLOR, G. I. 1923 Stability of a viscous liquid contained between two rotating cylinders *Trans. Roy. Soc.* A **223**, 289.

WALOWIT, J., TSAO, S. & DIPRIMA, R. C. 1964 Stability of flow between arbitrarily spaced concentric cylindrical surfaces including the effect of a radial temperature gradient. *J. Appl. Mech.* **31**, 585.

YIH, C. S. 1961 Dual rule of viscosity in the instability of revolving fluids in variable density. *Phys. Fluids*, **4**, 806.

YIH, C. S. 1965 *Dynamics of Nonhomogeneous Fluids*. Macmillan.