

## Taylor vortices and the evaluation of material constants: a critical assessment

By W. M. JONES, D. M. DAVIES AND M. C. THOMAS

Department of Physics, University College of Wales, Aberystwyth

(Received 27 November 1972)

The second-order fluid model having the equation of state

$$p_{ik} = -pg_{ik} + \alpha_0 A_{ik}^{(1)} + \alpha_1 A_{im}^{(1)} A_k^{(1)m} + \alpha_2 A_{ik}^{(2)}$$

in the usual notation defined in §1.2 is normally considered adequate for describing the rheological behaviour of dilute polymer solutions. However, normal stress differences associated with the  $\alpha_i$  are too small to be measured and Graebel (1961) suggested that experiments on Taylor stability would yield information from which the  $\alpha_i$  could be evaluated. Since then a considerable literature has grown up and the work described here was done to test current ideas.

$\alpha_1$  and  $\alpha_2$  may be evaluated by one of three methods. (i) The value of the critical Taylor number  $Ta_c$  for the polymer solution is different from that for a Newtonian liquid; put  $\Delta = \delta Ta_c / Ta_c$ , where  $\delta Ta_c$  is the difference, then  $\Delta$  is related to  $\alpha_1$ ,  $\alpha_2$  and  $\eta$  ( $= R_1/R_2$ , where  $R_1$  and  $R_2$  are the radii of the cylinders in the Couette apparatus). By determining  $\Delta$  for two values of  $\eta$  the necessary two equations for determining  $\alpha_1$  and  $\alpha_2$  are obtained. (ii) The wavenumber  $\epsilon_c$  is also a function of  $\alpha_1$  and  $\alpha_2$  and values of  $\Delta$  and of  $\epsilon_c$  for a given  $\eta$  give the necessary two equations. (iii) The tangential stress  $G$  on the stationary outer cylinder is a linear function of  $1 - Ta_c/Ta$  when  $Ta > Ta_c$ ; the slope of the line is a function of  $\alpha_1$  and  $\alpha_2$  and the slope and the value of  $\Delta$  for given  $\eta$  give the necessary two equations in this case. Method (iii) is the most accurate experimental method.

The apparatus enabled experiments to be done for  $\eta = 0.50, 0.70, 0.80, 0.90, 0.925$  and  $0.950$ . Most experiments were done with the three narrowest gaps. Method (i) did not give physically meaningful results in that  $\alpha_2$  was positive for all solutions tested. Method (ii) can only give unique results when  $\eta = 0.95$ . The theory of method (iii) only applies to a narrow gap whereas for the other two methods the theory used was general. Results obtained by methods (ii) and (iii) were in agreement for  $\eta = 0.95$ . Average values found for a 250 p.p.m. aqueous solution of polyacrylamide were

$$(\alpha_1 + 2\alpha_2) = -(0.75 \pm 0.04) \text{ mg m}^{-1} \quad \text{and} \quad \alpha_2 = -(20.8 \pm 0.2) \text{ mg m}^{-1}.$$

The accuracy in the determination of values at other concentrations and for other materials was less (about 10%) because the number of determinations of each value was smaller. The ratio  $N_2/N_1$  of the second to the first normal stress coefficient is always negative and for the polyacrylamide solutions is constant ( $= 0.02$ ) and independent of concentration.

To achieve meaningful results  $Ta_c$  must be measured to within better than 1% and  $\epsilon_c$  and the slopes of the lines to within about 1%. Visual observation of  $Ta_c$  is not accurate enough. Suggestions for optimizing the experimental design are given.

## 1. Introduction

### 1.1. Newtonian fluids

Consider two coaxial cylinders with fluid between them; the inner cylinder is rotated and the outer held at rest. At a critical speed of rotation centrifugal forces overcome viscous forces and a steady secondary motion in the form of torroidal vortices spaced regularly along the axis of the cylinders ensues (figure 1). Taylor (1923) considered the onset of the secondary flow theoretically and arrived at a dimensionless number  $Ta$  as a criterion for the flow. When  $d \ll R_1 + R_2$ , 'narrow-gap' geometry,

$$Ta = \frac{4R_1 d^3}{R_1 + R_2} \left( \frac{\Omega}{\nu} \right)^2, \quad (1)$$

where  $\nu$  is the kinematic viscosity of the fluid. Denote the critical value of the Taylor number by  $Ta_c$  then when  $Ta$  is such that  $Ta_c < Ta < 1.5Ta_c$  the pattern of vortices is unaltered but their intensity increases as  $Ta$  increases. Taylor found that  $Ta_c = 3414$ . When the assumption of narrow-gap geometry is no longer valid  $Ta_c > 3414$ , the extent of the inequality increasing with  $d/R_1$  according to

$$Ta_c = 3414 / [(1 - 0.652d/R_1) + 0.0098(1 - 0.652d/R_1)^{-1}]. \quad (2)$$

Taylor reported (2) to be accurate to within 1% in a range of values of  $\eta (= R_1/R_2)$  from 0.70 to 1.00.

$l$ ,  $d$  and the wavenumber  $\epsilon$  are related through

$$\epsilon = \pi d / l \quad (3)$$

and Taylor showed that  $d = l$ , so that  $\epsilon = \pi$ . Taylor verified his predictions visually by injecting dye into the fluid from small holes drilled in the surface of the inner cylinder.

Lewis (1928) obtained values of  $Ta_c$  and of  $\epsilon_c$  in 'wide-gap' apparatus in which  $\eta = 0.585, 0.70, 0.773$  or  $0.855$  for three different liquids. For the widest gap there was a 14% maximum variation in the measured values of  $\epsilon_c$  and for the narrowest a  $4\frac{1}{2}$ % variation; in all cases  $\epsilon_c > \pi$  (average value = 3.27).

Chandrasekhar (1953) used a somewhat more precise numerical method than Taylor's and found that  $Ta_c = 3390$  for the narrow gap with  $\epsilon_c = 3.12$ . Chandrasekhar (1958) also considered a special case ( $\eta = 0.5$ ) of the wide-gap geometry, and he showed that  $Ta_c = 6295$  with  $\epsilon_c = 3.2$  (i.e.  $l = 0.983d$ ). Donnelly (1958) and Donnelly & Fultz (1960) experimentally verified those theoretical predictions.

When the vortices occur extra power must be supplied to the rotating cylinder to maintain them. Taylor (1936) measured the resulting extra torque and related a friction factor to a Reynolds number for the system. Stuart (1958) calculated the extra torque using an energy-balance equation and Donnelly & Simon (1960)

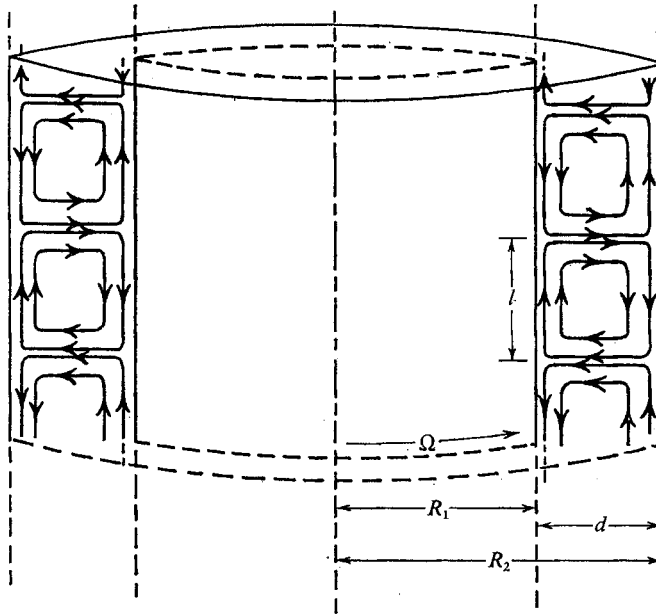


FIGURE 1. Axial distribution of Taylor vortices.  $l$  is the 'cell size'.

found a good fit between the experimental torque and calculated torque for  $Ta$  'not much larger' than  $Ta_c$ . Davey (1962) modified Stuart's work by taking into account the harmonics, as well as the fundamental mode, of the perturbations of the mean flow and obtained an expression for the tangential stress  $G$  on either cylinder for wide-gap as well as narrow-gap geometry. His equation can be written (for  $Ta > Ta_c$ ) as

$$\frac{Gd}{2\pi\mu R_1^3 \Omega h} = \frac{2R_2^2}{R_1(R_1 + R_2)} + \frac{\delta(R_1 + R_2)}{2R_1} \left(1 - \frac{Ta_c}{Ta}\right), \quad (4)$$

in which  $h$  is the height of the outer cylinder, over which the resultant torque is measured,  $\mu$  is the shear viscosity of the fluid and  $\delta$  is a function which is numerically equal to 1.528 for narrow-gap geometry whereas for the particular wide-gap case  $\eta = 0.5$ ,  $\delta = 0.838$ . When  $Ta < Ta_c$  the equation is made valid by writing  $\delta = 0$ . Donnelly (1965) developed an electrical method involving the measurement of the circulation of ions within the vortices, to determine the intensity (that is, amplitude) of the perturbation velocities. Donnelly & Schwarz (1965) report several consequential measurements of  $Ta_c$ , the drag and the amplitude for different values of  $\eta$ ; in an appendix to the paper Roberts describes a method of theoretically determining  $Ta_c$  for the wide-gap geometry and tabulates values of  $Ta_c$  as a function of  $\eta$  for values of  $\eta$  between 0.70 and 1.0.

Coles (1965) made an elaborate visual study of Taylor vortices and found an azimuthal wavy structure setting in when  $Ta \simeq 1.5Ta_c$ . The work is relevant to us in that it set an upper limit to the value of  $Ta$  for the theories we have used to be valid. (Debler, Fünér & Schaaf (1968) measured  $Ta_c$  and the drag for values of  $\eta$  in the range 0.5 to 0.95 and found a change in the torque as the wave pattern set in.)

1.2. *Non-Newtonian fluids*

No consistent notation has been used in writing equations of state in papers on Taylor stability so it is important to define notation and to re-write equations of previous workers, in the brief necessary review that follows. A recent interpretation of our work with slightly elastic liquids has arisen as a consequence of the theoretical work of Chan Man Fong (1970*a, b*) and the notation we use is his; the relevant equation of state for the second-order fluid is then

$$p_{ik} = -pg_{ik} + \alpha_0 A_{ik}^{(1)} + \alpha_1 A_{im}^{(1)} A_k^{(1)m} + \alpha_2 A_{ik}^{(2)}, \quad (5)$$

in which  $p$  is the hydrostatic pressure and  $g_{ik} = 1$  when  $i = k$  and  $g_{ik} = 0$  when  $i \neq k$ ;  $A_{ik}^{(1)} = v_{i,k} + v_{k,i}$ , where  $v_i$  is the velocity in the liquid conjugate to the direction  $x_i$  and in the usual way the subscript ‘ $k$ ’ signifies  $\partial/\partial x_k$ ;

$$A_{ik}^{(n+1)} = \partial(A_{ik}^{(n)})/\partial t + v^j A_{ik,j}^{(n)} + A_{ij}^{(n)} v_{,k}^j + A_{kj}^{(n)} v_{,i}^j.$$

In a simple shear flow convention has the velocity in the direction  $x_1$  and the velocity gradient in the direction  $x_2$  such that  $v_1 = \gamma x_2$  and  $v_2 = v_3 = 0$ . Equation (5) then gives

$$p_{12} = \alpha_0 \gamma, \quad p_{11} - p_{22} = N_1 \gamma^2, \quad p_{22} - p_{33} = N_2 \gamma^2, \quad (6a)$$

where

$$N_1 = -2\alpha_2, \quad N_2 = (\alpha_1 + 2\alpha_2). \quad (6b)$$

The  $N_i$  are the normal stress coefficients;  $p_{11} - p_{22}$  is the first normal stress difference and  $p_{22} - p_{33}$  is the second normal stress difference. For the linear displacement of an element of fluid to be in the direction of the applied stress  $\alpha_2$  must be negative so  $N_1$  must be positive; there is no restriction on the sign of  $N_2$ . Previous workers (to be referred to) suggest that  $|N_2|/|N_1| \ll 1$ .

It is usual to non-dimensionalize the parameters occurring in (6*b*). In the case of Couette flow this is done by dividing the equations by  $2\rho d^2$  and writing  $P_i$  for  $N_i/2\rho d^2$  and  $k_i$  for  $\alpha_i/2\rho d^2$  to give

$$P_1 = -2k_2, \quad P_2 = k_1 + 2k_2; \quad (6c)$$

$\rho$  is the density of the fluid.

Couette flow between rotating cylinders is a practical example of a simple shear flow. Oldroyd (1950) considered viscoelastic materials in Couette flow when the inner cylinder only is rotated; the consequential distribution of normal stress differences led to an interpretation of the Weissenberg effect. However, it was ten years before Graebel (1961) provided equations to enable  $\alpha_1$  (the ‘cross-viscosity’) to be determined from measurements of normal stresses in Couette flow and found that the predicted percentage contribution of the non-Newtonian behaviour to the stresses was small. On the other hand Graebel found that even ‘small amounts of elasticity’ would affect the stability. When  $\alpha_1$  was positive the flow was destabilized. He did not discuss fully the case of  $\alpha_1$  negative but there was evidence to suggest that the flow would be stabilized in that case. Thomas & Walters (1964) used an equation of state designated ‘liquid  $B^1$ ’ which was very similar to the earlier one of Oldroyd (1950). The distribution of normal stresses was as given by (5) with  $\alpha_1 = |2\alpha_2|$ , that is,  $N_2 = 0$ . They found that the flow was

destabilized, the relevant elastic parameter being  $k_1$ ; furthermore, the wavenumber increased relative to that of a Newtonian liquid. (When  $k_1 = 0.005$ ,  $Ta_c$  was smaller by 9% and  $\epsilon_c$  greater by 3%.) Chan Man Fong (1965) considered the same problem using a liquid model  $A^1$  in which the distribution of normal stresses was consistent with setting  $\alpha_1 = 0$  in (5), and the relevant elastic parameter in this case was  $P_2$ . The elasticity stabilized the flow and diminished the wavenumber, having an effect opposite to that for liquid model  $B^1$ . Rubín & Elata (1966), in a review of previous work, refer to Datta's theoretical work on stability using (5) as the model. The relevant elastic parameter in that work was  $P_2 R_1/d$ ; when  $P_2 > 0$  the flow was destabilized and the wavenumber increased whereas when  $P_2 < 0$  the flow was stabilized and the wavenumber diminished. Rubín & Elata also refer to the 'Ericksen anisotropic fluid' and point out that a choice of positive values for some of the parameters will lead to the prediction of flow stabilization with no change in wavenumber. They describe their own experiments with aqueous solutions of polyacrylamide, of polyox and of guar gum and report that in all cases and for all concentrations investigated the flow was stabilized with no change in wavenumber in accord with Ericksen's anisotropic fluid model. Rubín, Elata & Poreh (1968) report on theoretical work with yet another liquid model designated liquid  $C^1$ ; model  $C^1$  differs from  $B^1$  in that, for  $B^1$ ,  $p_{22} = p_{33} = 0$  whereas for  $C^1$  only  $p_{33} = 0$ ; in both  $N_2 = 0$ . Model  $C^1$  predicted destabilization accompanied by a decrease in wavenumber. The predictions of all the models so far discussed are summarized in figure 2.

It is clear from the above discussion that values of  $P_1$  and of  $P_2 R_1/d$  are relevant in discussing Taylor stability. The relative roles of the two were clarified by Ginn & Denn (1969) using narrow-gap theory and (5). They found positive values of both  $P_1$  and  $P_2$  to be destabilizing and negative values of  $P_2$  to be stabilizing; whether destabilization or stabilization occurs depends on the relative magnitudes of  $P_1$  and  $P_2 R_1/d$ .  $\epsilon_c$  increased with increasing  $P_1$  and with increasing  $P_2 R_1/d$ . Graphs showing  $Ta_c$  and  $\epsilon_c$  as functions of  $P_1$  and  $P_2 R_1/d$  were produced by numerical techniques. Denn & Roisman (1969) tested the theory in apparatus affording three choices of  $R_1/d$ , in the range 20–50; measurement of torque was used to detect the onset of instability: the transition was sharp but 'the uncertainty in establishing the critical Taylor number is of the order of 10%'. Dilute solutions of six different polymers of various concentrations were used in the experiments. To analyse the results it was necessary to assume that  $\alpha_1$  and  $\alpha_2$  were functions of the shear rate. They put  $-\alpha_2 = a\gamma^b$  and  $\alpha_1 + 2\alpha_2 = 2\lambda a\gamma^b$ , where  $\lambda = N_1/N_2$ . Using the results for the three gaps 'the three parameters  $a$ ,  $b$  and  $\lambda$  were then chosen such that the Taylor numbers predicted by the stability theory corresponded to the experimental Taylor number'. For only two solutions were consistent values of  $a$ ,  $b$  and  $\lambda$  obtained. For them, values of  $\alpha_2$  were in agreement with values of  $\alpha_2$  obtained by extrapolating the results of rheogoniometer experiments done with relative high concentrations, to the low concentrations used in the Couette-flow experiments.  $\lambda$  was found to be negative and  $|\lambda| \leq 0.05$ ;  $b$  was found to be 2.0.

Hayes & Hutton (1970) worked with aqueous solutions of two types of polyacrylamide, and with polyox solutions, and found stabilization which increased

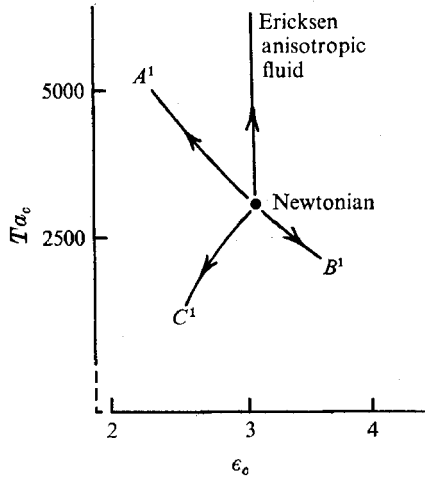


FIGURE 2. Summary of the theoretical predictions of various liquid models. Elasticity increasing in direction of arrows.

with concentration with no change in wavenumber, in all cases ( $\eta = 0.90$  and  $0.94$ ). Using Ginn & Denn's theoretical diagrams they found all their results to lie in a region where  $N_2$  and  $N_1$  were negative; therefore they concluded that the model (5) cannot be valid in their situation. (Their results are on the other hand in accord with the predictions from the Ericksen anisotropic fluid.) Goddard & Miller (1967), Karlsson, Sokolov & Tanner (1971), Lockett & Rivlin (1968) and Smith & Rivlin (1972) consider the second-order fluid having three material constants to be inadequate to describe results of experiments on Taylor stability and each suggests that several material constants (up to nine) would be necessary.

All the above theoretical work involved the assumption of narrow-gap geometry. It was possible that the disagreement between second-order-fluid theory and the experimental results could be attributed to that assumption rather than inadequacies in the fluid model. Chan Man Fong (1970*b*) used model (5) to discuss the general case of the wide gap in such a way that the narrow gap was a special case. The numerical results he published were insufficient to produce a set of comprehensive curves relating  $T a_c$  to  $\alpha_1$  and  $\alpha_2$  but a three-dimensional surface could be produced. A drawing of such a surface is shown in figure 3. Chan Man Fong also considered the effect of elasticity on  $\epsilon_c$ . The recent purpose of the work to be described in this paper was to see if  $\alpha_i$  could be evaluated by comparing results on Taylor stability with Chan Man Fong's theoretical predictions.

However, before going on to describe our work, a further relevant connexion between elasticity and Taylor vortices must be mentioned. During secondary flow in elastic liquids  $G$  as given by (4) may be less than anticipated (cf. Jones & Marshall 1969). Chan Man Fong (1970*a*) discussed such 'drag reduction' for narrow-gap geometry in terms of a Reynolds number. Denn, Sun & Rushton (1971), on the other hand, also from narrow-gap theory, arrived at a modification of (4):

$$\frac{Gd}{2\pi\mu R_1^3 \Omega \bar{h}} = \frac{2R_2^2}{R_1(R_1 + R_2)} + \left( \frac{\delta(R_1 + R_2)}{2R_1} - 6P_1 + 6P_2 \frac{R_1}{d} \right) \left( 1 - \frac{T a_c}{T a} \right). \quad (7)$$

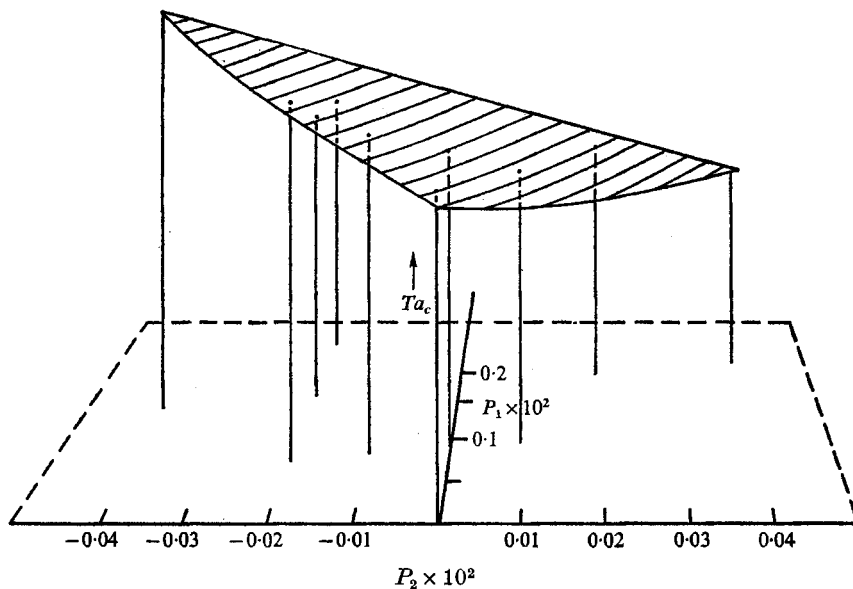


FIGURE 3.  $Ta_c$  as a function of  $P_1$  and  $P_2$  for  $\eta = 0.95$ . Theoretical results taken from Chan Man Fong's (1970*b*) paper with his numbers converted to our definition of  $k_i$ .

Equation (7) is a relationship between  $\alpha_1$  and  $\alpha_2$ . A second relationship – albeit a complicated one requiring a numerical solution – is the one mentioned above, obtained from observation of differences in  $Ta_c$  between solution and solvent. Denn, Sunn & Rushton state that those two relationships, obtained for the same value of  $\eta$ , can be used to determine  $\alpha_1$  and  $\alpha_2$  but they quote no values determined in such a manner.

Our experiments were initially concerned with testing apparatus and verifying that there were changes in the value of  $Ta_c$  and in the amount of drag in dilute polymer solutions compared with Newtonian liquids (Marshall 1967; Jones & Marshall 1969),  $\eta = 0.95$ . The experiments were extended to find the effect of temperature on  $Ta_c$  and on drag reduction (Davies 1968). Recently, Davies (1972) has done experiments with various values of  $\eta$  ranging from 0.5 to 0.95 and has measured the cell size. During the work it became apparent that three-dimensional surfaces such as those in figure 3 were too crude for finding the  $\alpha_i$  for the small changes in  $Ta_c$  which were being observed and H. Holstein of the Computer Science Department (UCW) produced a set of curves (figures 5 and 6) which could be quickly used to find the  $\alpha_i$  from measurements of  $Ta_c$  and  $\epsilon_c$ . Finally it was found that measurement of  $Ta_c$  to within  $\pm 1\frac{1}{2}\%$  (as done by Davies) was not accurate enough and M. C. Thomas has re-done some of his work to determine  $Ta_c$  to within  $\pm 0.8\%$ , which is the best that can be done with our apparatus. All our work is reported in this paper together with our assessment of the study of Taylor vortices as a means of determining the material constants of dilute polymer solutions.

The work is considered fundamentally important in that despite the long standing of equations such as (5) experimental evaluation of the  $\alpha_i$  has not been

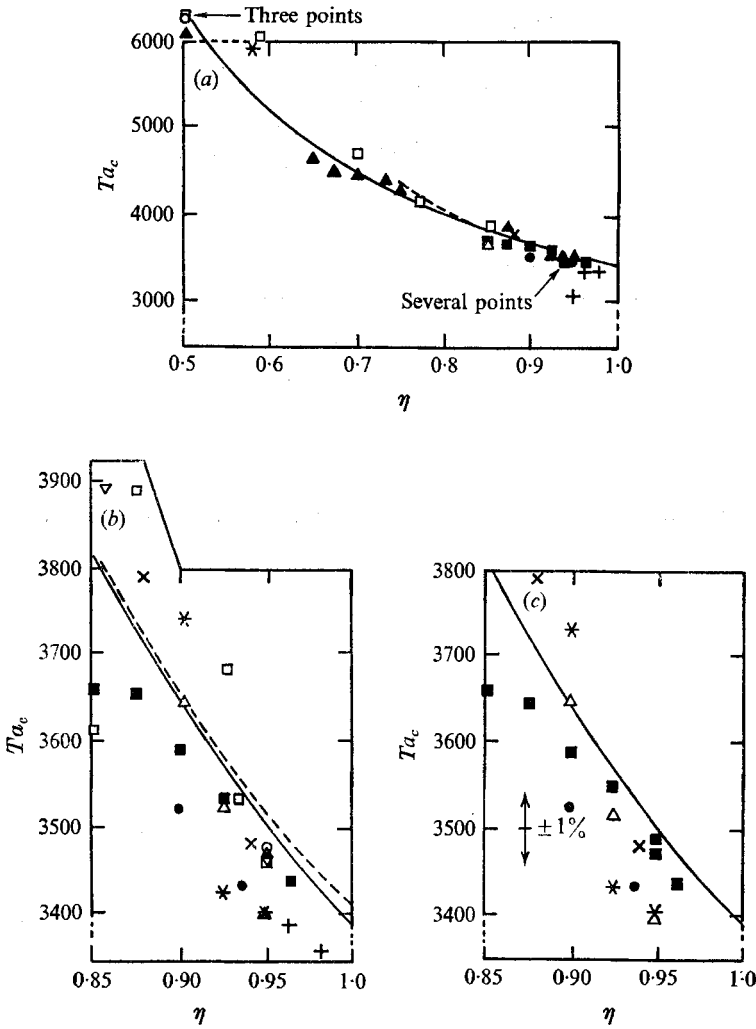


FIGURE 4.  $T\alpha_c$  as a function of  $\eta$  for Newtonian fluids. Points are experimental. ----, theories of Taylor and of Roberts; —, Holstein's theory. (a) All results. (b) Section of (a) enlarged. (c) Results of those claiming 1% accuracy or better. \*, DMD;  $\Delta$ , MCT;  $\blacksquare$ , Donnelly *et al.*;  $\bullet$ , Hayes & Hutton;  $\times$ , Taylor.

achieved for 'weak' elastic fluids such as dilute polymer solutions; it is considered industrially important because when the  $\alpha_i$  are known (5) could be used to predict results in other geometries.

While the theoretical and experimental work referred to above was being done other more qualitative experiments were carried out. Some of the results of that work were used in preparing figure 4; see Merrill, Mickley & Ram (1962), Song & Tsai (1966), Giesekeus (1966) and Bailey (1969).



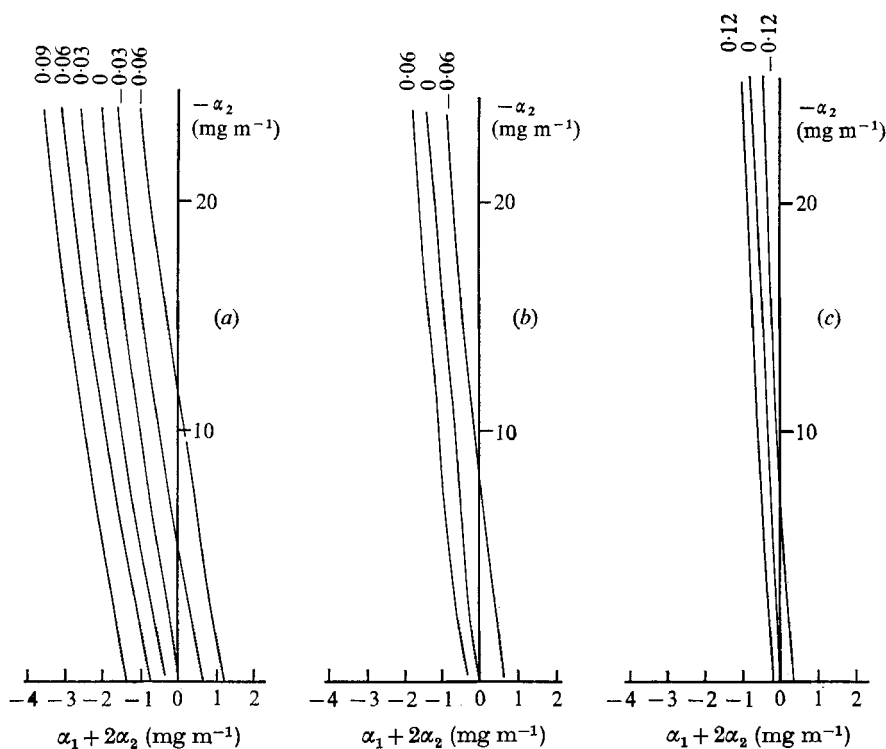


FIGURE 5. Theoretical curves of constant  $\Delta$  as a function of the  $\alpha_i$ .  
 (a)  $\eta = 0.90$ . (b)  $\eta = 0.925$ . (c)  $\eta = 0.95$ .

## 2. Theory

From figure 4 it is seen that there is a good agreement between the theoretical curves, but the 'best' experimental results (figure 4c) are with one exception 1–2% less than the theoretical values. Possible reasons for the difference are examined in § 5 but as a consequence of it  $\{Ta'_c - Ta_c\}/Ta_c = \Delta$ , rather than  $Ta'_c$ , has been determined theoretically as a function of the  $\alpha_i$ , where  $Ta'_c$  refers to the elastic liquid and  $Ta_c$  to Newtonian liquids (figure 5). (Note the greater sensitivity of  $\Delta$  to the  $\alpha_i$  as  $\eta$  increases.) Experimental determination of  $\Delta$  for any two gaps should produce two curves giving values of  $\alpha_1$  and  $\alpha_2$  at the point of intersection.

An alternative to working with different gaps is to measure  $Ta_c$  and  $\epsilon_c$  in the same gap (figure 6). It is seen that curves of constant  $\Delta$  and constant  $\bar{\epsilon}$  cross for the case when  $\eta = 0.95$  only, so results in the other two gaps would not give unique values for the  $\alpha_i$ .

Another alternative is to measure  $\Delta$  and the drag reduction in a given gap. Then it is convenient to alter the form of (7) by substituting for  $P_1$  and  $P_2$  and by collecting terms in  $R_1$ ,  $R_2$  and  $d$  on the right-hand side, thus giving

$$\frac{G}{2\mu\pi\Omega h} = \frac{\kappa\phi P}{\mu} = A + \{\delta B + \psi(\alpha_1, \alpha_2)\} \left(1 - \frac{Ta_c}{Ta}\right), \quad (8)$$

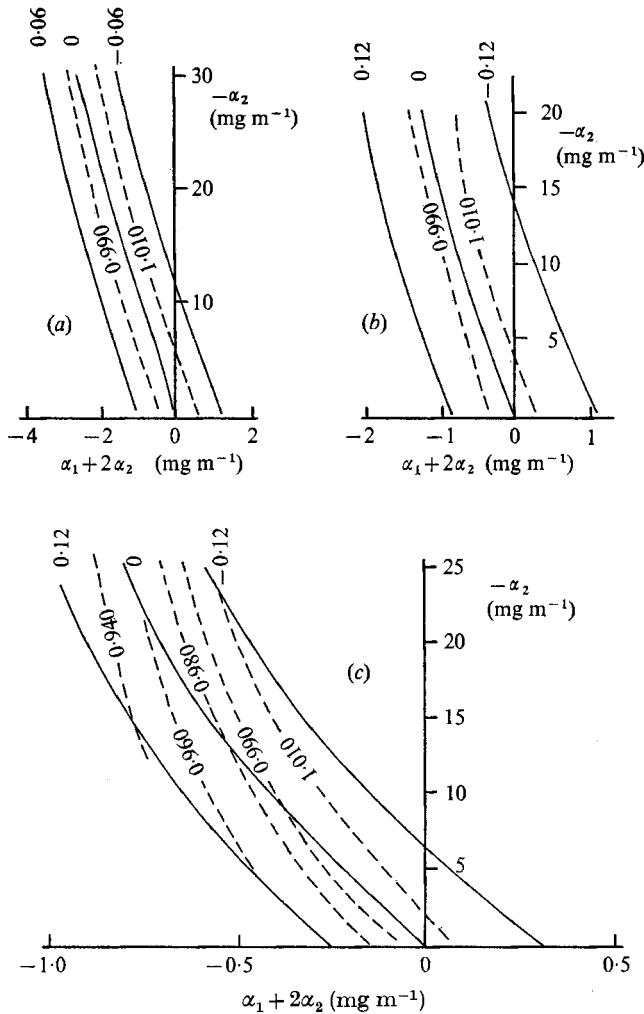


FIGURE 6. Theoretical curves of constant  $\Delta$  (solid lines) and of constant  $\bar{\epsilon}$  (broken lines) as a function of the  $\alpha_i$ . ( $\bar{\epsilon} = (\epsilon_c \text{ for non-Newtonian fluid})/(\epsilon_c \text{ for water})$ .) (a)  $\eta = 0.90$ . (b)  $\eta = 0.925$ . (c)  $\eta = 0.95$ .

where  $\kappa$  is a constant of the apparatus,  $\phi$  is the angular twist of the torsion fibre supporting the outer cylinder,  $P$  is the period of rotation of the inner cylinder,

$$A = 2R_1^2 R_2^2 / d(R_1 + R_2), \quad B = R_1^2 (R_1 + R_2) / 2d,$$

$$\psi(\alpha_1, \alpha_2) = \frac{3}{\rho} \left( \frac{R_1}{d} \right)^4 \left[ \alpha_1 + 2\alpha_2 \left( 1 + \frac{d}{R_1} \right) \right]. \quad (9)$$

When primary flow only occurs  $\phi P/\mu$  is constant ( $= A/\kappa$ ) and an average experimental value  $(\phi P/\mu)_{av}$  may be obtained from several measurements in the primary-flow region. Hence  $\kappa = A/(\phi P/\mu)_{av}$  is obtained. If  $|\phi|$  is written for  $(\phi P/\mu)/(\phi P/\mu)_{av}$ , equation (8) becomes, for Newtonian fluids,

$$|\phi| = 1.0 + \delta\eta(1 - Ta_c/Ta) \quad (10)$$

( $B/A = \eta$  to within  $\frac{1}{3}\%$  for  $\eta$  from 0.90 to 1.00).

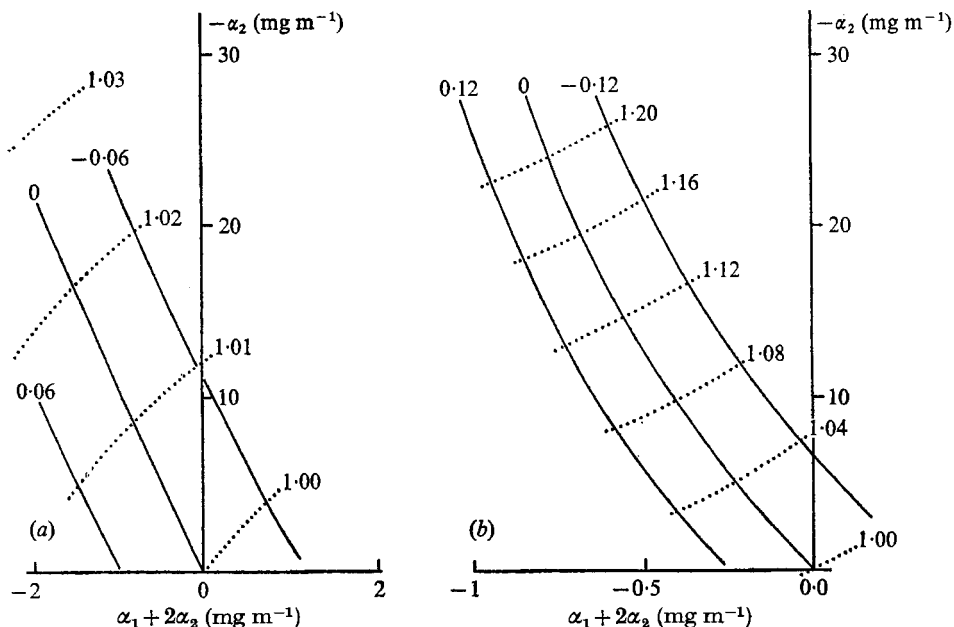


FIGURE 7. Theoretical curves of constant  $\Delta$  (solid lines) and of constant  $s_1/s_2$  (dotted lines) as a function of the  $\alpha_i$ . (a)  $\eta = 0.90$ . (b)  $\eta = 0.95$ .

Straight lines should be obtained when  $|\phi|$  is represented as a function of  $1 - T\alpha_o/Ta$ , and  $\delta$  may be found from the slope  $s_1$  of the line obtained with Newtonian liquids. The ratio  $s_1/s_2$ , where  $s_2$  is the slope of the line obtained with the elastic liquid, gives

$$s_1/s_2 = \delta B / (\delta B + \psi(\alpha_1, \alpha_2)). \quad (11)$$

Values of the  $\alpha_i$  may then be obtained from curves such as those in figure 7. (It should be noted that (10) and (11) are independent of  $\kappa$  and therefore experiments done with different torsion fibres could be used for comparing slopes.)

### 3. Experiments

The basic apparatus (in which  $\eta = 0.95$ ) and the experimental procedure were described by Jones & Marshall (1969); since then other inner cylinders have been provided enabling experiments to be done for  $\eta = 0.925, 0.90, 0.80, 0.70$  and  $0.50$ . Furthermore, a transparent outer cylinder (of Perspex) has been fabricated which enables visual experiments to be made for each gap. A streak of ink is placed down the wall of the inner cylinder and the apparatus is assembled and immersed. In primary laminar flow the ink spreads around the inner cylinder but at the point of instability the ink circulates in the vortices and is photographed; the print is measured to give the cell size.

The largest value of  $\Delta$  we have measured was  $0.12$  but most values lay between  $\pm 0.03$ . It will be appreciated therefore that accurate determination of the material constants can only arise if  $Ta_c$  is determined with precision. A discussion of the accuracy of the apparatus is therefore relevant.

Of the quantities in the expression for  $Ta$ , equation (1),  $R_1$  and  $R_2$  can be measured to within  $\pm 0.0001$  cm thus giving  $d$  to within  $\pm 0.0002$  cm. The largest possible error in  $Ta_c$  due to that cause is therefore 6% when  $\eta = 0.95$  and since the error is constant, then, for example, if the observed  $\Delta$  is 0.03 then the 6% error in  $Ta_c$  means  $\Delta$  could lie anywhere between 0.028 and 0.032.

The accuracy of the value substituted for  $\nu$  depends entirely on the accuracy to which the temperature of the fluid is known. The bath temperature can be kept constant to within  $\pm 0.2^\circ\text{C}$  and the temperature during any one determination of  $\phi$  and  $P$  measured to within  $\pm 0.1^\circ\text{C}$ . That means that  $\mu$  and hence  $\nu$  is known to within  $\pm 0.2\%$  and any value of  $Ta$  to within  $\pm 0.4\%$ . But these are now random errors and if the value of  $\Delta$  is 0.030 the uncertainty in  $Ta_c$  will mean that the range of possible values of  $\Delta$  is 0.022 to 0.038.

The accuracy of  $\Omega_c$  depends on the accuracy to which the onset of the instability can be observed from the torque measurement. Donnelly (1958) has established that there is a discontinuity in the torque at the transition and approaching this discontinuity using our apparatus is limited by the precision to which the speed of rotation can be adjusted. The adjustment is such that, at the critical condition when  $P \simeq 2.00$  s, the period cannot be adjusted to within better than 0.01 s, and in an 'average' experiment the uncertainty is nearer 0.015 s, hence  $P_c \simeq (2.000 \pm 0.008)$  s, leading to a possible 0.8% error in  $Ta_c$  and a possible range of values of  $\Delta$  from 0.14 to 0.46 when  $\Delta \simeq 0.30$ . (In a very few experiments the adjustment can by chance, be better; our best value of  $P_c$  was  $(1.698 \pm 0.002)$  s, leading to a 0.2% error in  $Ta_c$ .)

The above limits on accuracy can only be achieved by careful experimentation. Most of our early experiments were done with the idea that the Taylor vortices were very sensitive to elasticity; at that time repeated experiments showed our values of  $Ta_c$  to be reproducible to within around  $\pm 1\frac{1}{2}\%$ . The above analysis would suggest that the best we could hope to achieve would be a  $\pm 0.7\%$  probability error or a 1.2% uncertainty, as a consequence of the random errors. The best we have achieved by repeated measurements of  $Ta_c$  is  $\pm 0.8\%$ .

The visual technique of finding  $Ta_c$  is not accurate enough. In the first place embryo cells, which do not fill the gap, appear when  $Ta < Ta_c$ , so one has to note carefully when the vortices first fill the gap and that is difficult to do. (Hayes & Hutton (1970) also report on those difficulties.) We were only able satisfactorily to see the transition because of prior knowledge from torque measurements.

Another measurement to be made in the experiments is of  $\bar{\epsilon}$ . The length of annulus visible through the outer cylinder is 4.40 cm and hence, when  $\eta = 0.95$ , there are 44 cells, whose average size  $l$  is to be determined. However, the axial distribution of the sizes of the cells (figure 8) is such that the root-mean-square deviation from the mean of the sizes shown is 3.5%. Repeated measurement of the average cell size also gives large variations. The eight values of the average cell size determined from eight photographs each taken after the apparatus had been dismantled and re-assembled, showed a r.m.s. deviation of 4% from the mean of the eight values. To determine  $\bar{\epsilon}$  so that consequential inaccuracies in the  $\alpha_i$  are no worse than those resulting from uncertainty in  $\Delta$  would require 1% accuracy in the cell size and therefore photographs of 130 repeated experiments

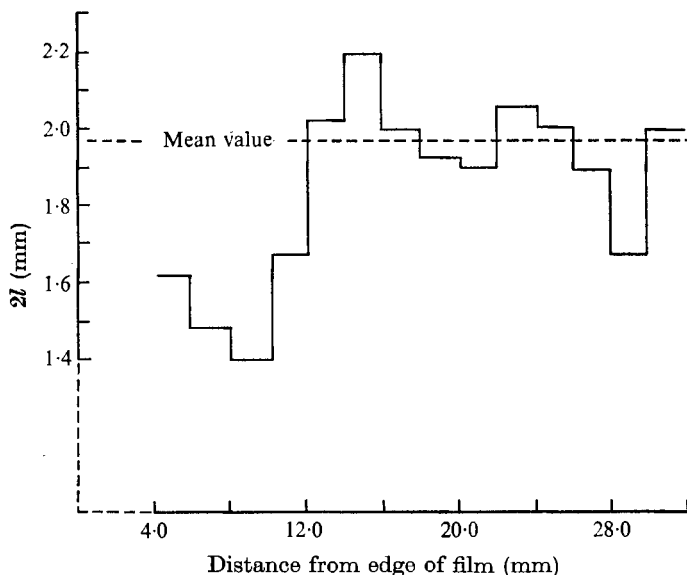


FIGURE 8. Axial variation of cell size.  $\eta = 0.95$ .

of the cell pattern at the transition. To sum up: cell size is relatively insensitive to elasticity and to aggravate the situation the cell pattern itself makes accurate measurement impractical. Nevertheless, even with such limited accuracy as can be achieved, the results will show that trends in the values of the  $\alpha_i$  can be established using figure 6 (c).

The method using figure 7 is applicable to narrow gaps only and involves the measurement of  $\phi$  as a function of  $P$ . Three factors affecting the accuracy have to be taken into account.

(i) A zero error  $\delta\phi$  can occur in setting the torsion head, which alters the reading of  $|\phi|$  from unity to  $1.0 \pm \delta\phi/\phi$ . In some of our results there is evidence of a contribution from  $\delta\phi$  but in such cases  $|\phi|$  reaches the value unity (i.e.  $\delta\phi \ll \phi$ ) well before secondary flow sets in, when the measurement of  $s_1$  and  $s_2$  becomes relevant.

(ii) Error due to small oscillations of the outer cylinder during secondary flow can be reduced to within required limits by taking a sufficient number of readings. (When  $T\alpha_c$  is measured to within 0.8%,  $s_1/s_2$  must be measured to within 0.7% to have no more than a comparable effect on the  $\alpha_i$ .)

(iii) There is a force of static friction in the teltales which increases as the torque applied through the suspension to the outer cylinder increases, so that, when the viscosities of aqueous glycerol solutions were obtained from the torque in the solution relative to that in water (using water as standard) the values were too large: in fact, the larger the viscosity the larger the discrepancy. When the ratio of the values of  $\mu$  for glycerol and water was 1.20, the  $\mu$  for glycerol was 2% too large. Since in all these measurements  $|\phi| = 1.0$  throughout the laminar flow region, the frictional force called into play must be proportional to  $\gamma$  thus effectively increasing  $\mu$  by a constant  $\delta\mu$  in any one experiment. (Donnelly (1958) gives

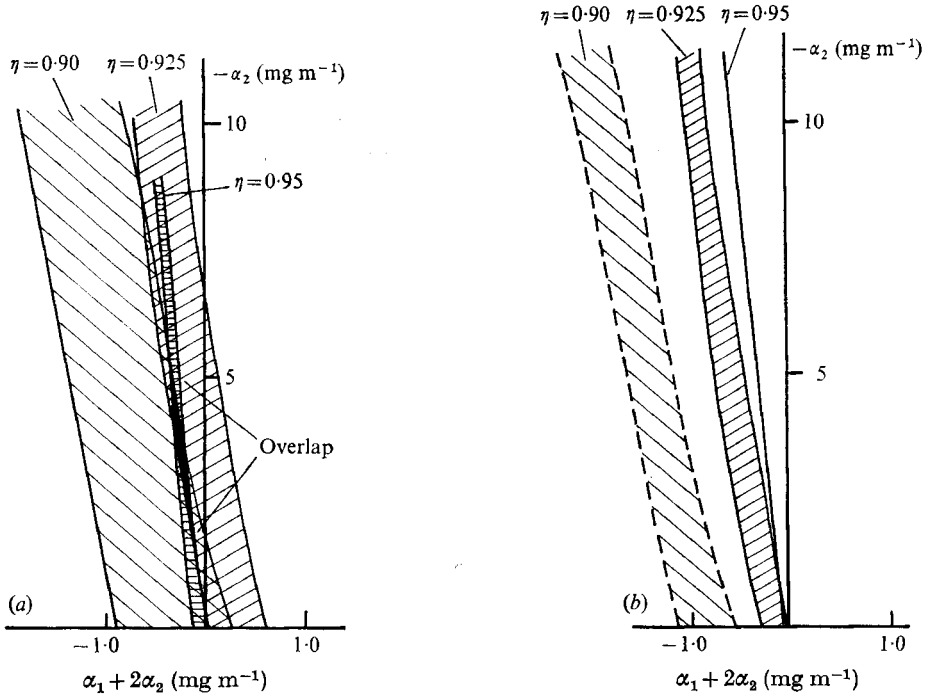


FIGURE 9. Experimental values of  $\Delta$  superposed on the theoretical curves of figure 5 together with experimental tolerances.

values of quantities which can be used to show that he was finding similar effects in his apparatus.) The ultimate effect of the frictional force is the same as would be obtained if  $\kappa$  [equation (8)] were increased for that particular experiment and therefore the value of  $\psi(\alpha_1, \alpha_2)$  found from (11) will not be influenced by the friction. We have also found that  $Ta_c$  is not affected by the friction provided that the value of  $\mu$  as found in the Ostwald viscometer is used in the calculation of  $Ta_c$  and not the apparent viscosity obtainable from the Couette apparatus; that means that the friction does not affect the sensitivity of the instrument to the onset of the Taylor instability. Also the viscosities of the test materials cannot be found sufficiently accurately *in situ*. But the important conclusion is that experimental data of torque measurements must be normalized as in (10) for accurate conclusions (within better than 2%) to be drawn concerning relative drag in separate experiments on secondary flow.

## 4. Results

### 4.1. Using the values of $\Delta$ found for three different $\eta$ to determine $\alpha_i$

In figure 9 (a) the consequences of determining  $\Delta$  to within  $\pm 0.03$  are adequately illustrated. When  $\eta = 0.90$ ,  $\Delta$  was  $0.02 \pm 0.03$ ; when  $\eta = 0.925$ ,  $\Delta$  was  $-0.03 \pm 0.03$ ; when  $\eta = 0.95$ ,  $\Delta$  was  $0.025 \pm 0.03$ . The three bands of possible values of the  $\alpha_i$  all overlap in the region where  $\alpha_2$  lies between  $-3.2$  and  $-9.6$   $\text{mg m}^{-1}$  and  $\alpha_1 + 2\alpha_2$  lies between  $-0.1$  and  $-0.4$   $\text{mg m}^{-1}$ . For all the other materials and concentrations

Material	Concentration (p.p.m.)	100 $\Delta$	$\bar{\epsilon}$	$-(\alpha_1 + 2\alpha_2)$ (mg m <sup>-1</sup> )	$-\alpha_2$ (mg m <sup>-1</sup> )	$-\lambda$
Polyacrylamide	50	-2.7	1.012	0.03	2.0	0.01
	100	0.0	1.008	0.35	8.0	0.02
	250	0.0	0.982	0.70	20.0	0.02
'Kelzan'	10	4.5	0.962	0.75	18.5	0.02
	50	4.5	0.928	0.95	27.0	0.02
Polyox	50	2.5	0.995	0.42	8.6	0.02
	100	10.0	1.040	—	+ve	—
	250	7.5	1.032	—	+ve	—
	500	5.3	1.030	—	+ve	—

TABLE 1. Values of the material constants determined from figure 6 using experimental measurements of  $\Delta$  and of  $\bar{\epsilon}$  (temperature  $t = 25^\circ\text{C}$ ). Experimental accuracy is discussed in the text.

tested in the three gaps the three bands do not overlap in the region where  $\alpha_2$  is negative. To obtain an overlap the bands had to be extrapolated to positive  $\alpha_2$ . Consequently  $\alpha_1$  was equated to  $a + b\gamma^2$  and  $\alpha_1 + 2\alpha_2$  to  $\lambda(a + b\gamma^2)$ , in which  $a$ ,  $b$  and  $\lambda$  were regarded as constants (cf. Denn & Roisman 1969; see § 1), and the results in the three gaps were used to evaluate  $a$ ,  $b$  and  $\lambda$ . No real mathematical solution was obtained in any one case.

A factor affecting the results was the presence of slots inadvertently cut in the guard rings for admission of fluid to the apparatus. These slots went unnoticed until our experiments with the wider gaps were begun and then the experimental value of  $Ta_c$  for Newtonian fluids was 12% greater than the theoretical value; when the slots were sealed the theoretical and experimental values agreed. The effect was similar to that predicted by DiPrima (1960) when an axial flow is present, which suppresses the Taylor vortices. Griffiths & Thomas (1966) theoretically confirmed DiPrima's work but also showed that in an elastic liquid the suppression of the vortices is less marked. Some of the results shown in figure 4(a) suggest that a small axial flow was present in some sets of apparatus and since Newtonian and non-Newtonian fluids are not similarly affected erroneous results would be obtained for  $\Delta$ . Those of our earlier results which were repeated with the slots sealed, giving  $Ta_c$  to within  $\pm 1\frac{1}{2}\%$ , were not altered in the narrower gaps ( $\eta = 0.90, 0.925$  and  $0.95$ ). Nevertheless, we felt more accurate experiments were required with the slots sealed and the consequences for  $\Delta$  of measuring  $Ta_c$  to within  $\pm 0.08\%$  are shown in figure 9(b). But again no value of the  $\alpha_i$  was obtained in the region where  $\alpha_2$  is negative.

#### 4.2. Using values of $\Delta$ and of $\bar{\epsilon}$ to determine $\alpha_i$

Realistic values of the  $\alpha_i$  were obtained for polyacrylamide solutions and for Kelzan solutions but for the three highest concentrations of the polyox solutions  $\alpha_2$  is positive.

#### 4.3. Using values of $\Delta$ and of $s_1/s_2$ to determine $\alpha_i$

$s_1$  was obtained by drawing the best straight line through the experimental points obtained with water as the test fluid (figure 10a).  $s_2$  was then found from

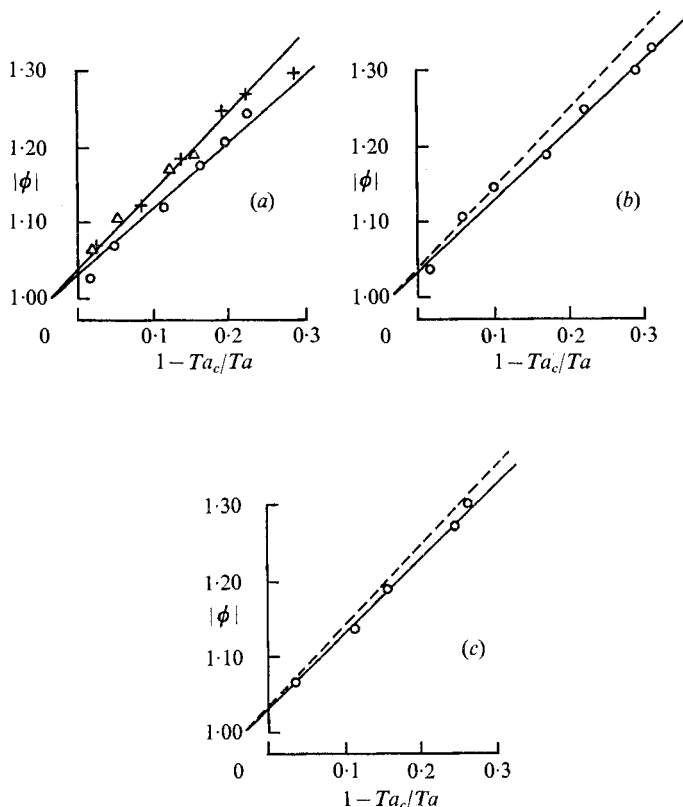


FIGURE 10.  $|\phi|$  as a function of  $1 - Ta_c/Ta$  for Newtonian (crosses and triangles) and non-Newtonian (circles) fluids.  $\eta = 0.95$ . Aqueous polyacrylamide solutions. (a) 250 p.p.m. (b) 100 p.p.m. (c) 50 p.p.m.

Concentration (p.p.m.)	$t$ ( $^{\circ}\text{C}$ )	$-(\alpha_1 + 2\alpha_2)$ ( $\text{mg m}^{-1}$ )	$-\alpha_2$ ( $\text{mg m}^{-1}$ )	$-\lambda$
250†	25	0.66	18.8	0.02
50	15	0.32	6.2	0.03
	25	0.40	11.2	0.02
	35	0.75	21.4	0.02
100	15	-0.30	-5.0	0.03
	25	0.22	4.4	0.03
	35	0.63	15.4	0.02
250	15	0.45	11.0	0.02
	25	0.78	24.0	0.02
	35	1.08	33.0	0.02
500	15	0.55	10.4	0.03
	25	0.78	19.2	0.02
	35	1.17	48.8	0.01

† Experiments of Marshall (1967); others from Davies (1972).

TABLE 2. Values of the material constants of aqueous polyacrylamide solutions as a function of temperature, determined from figure 7 using experimental measurements of  $\Delta$  and of  $s_1/s_2$  for  $\eta = 0.95$ . Experimental accuracy discussed in the text.



Concentration (p.p.m.)	$\eta$	100 $\Delta$	$-(\alpha_1 + 2\alpha_2)$ (mg m <sup>-1</sup> )	$-\alpha_2$ (mg m <sup>-1</sup> )	$\gamma$ (s <sup>-1</sup> )	$-\lambda$
50	0.95	2.25	0.35	6.2	183	0.03
	0.925	3.33	0.85	9.4	67	0.03
	0.90	5.0	1.9	10	33	0.10
100	0.95	1.5	0.6	14.5	187	0.02
	0.925	7.0	2.8	41.4	70	0.03
	0.90	9.0	5.2	44.1	35	0.06
250	0.95	6.0	0.89	20.4	210	0.02
	0.925	10.0	3.8	61.9	79	0.03
	0.90	12.0	18	184	40	0.05

TABLE 3. Values of the material constants of aqueous polyacrylamide solutions at 25 °C, determined from figure 7 using experimental measurements of  $\Delta$  and of  $s_1/s_2$ , for various values of  $\eta$ . Experimental accuracy discussed in the text.

experiments with the polymer solutions, by drawing the best straight lines with the additional restriction that they should pass through the same intercept on the abscissa as does the line for water (figures 10*a*, *b*, *c*). Values of the  $\alpha_n$  were then obtained from figure 7. That  $|\phi| \neq 1$  when  $1 - Ta_c/Ta = 0$  results from the experimental discontinuity that occurs in  $G$  when  $Ta = Ta_c$ , and the theory takes no account of such a discontinuity.

## 5. Discussion

### 5.1. $Ta_c$ as a function of $\eta$ : Newtonian fluids

The percentage differences between our experimental values of  $Ta_c$  and the theoretical values are within our error of measurement of  $d^3$ . (If we correct  $d^3$  accordingly only insignificant differences occur in our values of  $\Delta$ .) But the striking feature of figure 4 (*c*) is that all but two of the experimentally determined values of  $Ta_c$  are less than the theoretical ones and one of those two was obtained by D.M. Davies with slots in the guard ring of the apparatus. If differences between theory and experiment were due to errors in  $d$ , a random distribution of points about the theoretical curve might be expected. Suppose the experiments and theory disagree because energy is fed from the motor into the fluid, thus actuating the vortices at lower values of  $\Omega$  than that predicted by the ideal mathematical case. (Vortices can in fact be actuated as  $\Omega \rightarrow \Omega_c$  by tapping the apparatus lightly.) From considerations of energy balance it can be shown that

$$r_3/r_2 = r_1(1 - r_3) + r_3,$$

in which

$$r_1 = [Ta'_n/Ta_n]^{1/2}, \quad r_2 = [Ta'_p/Ta'_n]^{1/2}, \quad r_3 = [Ta_p/Ta_n]^{1/2},$$

where  $Ta'_n$  = the theoretical critical Taylor number (Newtonian),

$Ta_n$  = the observed critical Taylor number (Newtonian),

$Ta'_p$  = the theoretical critical Taylor number (non-Newtonian),

$Ta_p$  = the observed critical Taylor number (non-Newtonian).

By substitution of known values it is seen that  $r_3$  is negligibly different from  $r_2$ , that is, the observed  $\Delta$  will not differ significantly from the theoretical  $\Delta$  as a consequence of  $Ta_n$  being less than  $Ta'_n$ .

If on the other hand  $Ta_n < Ta'_n$  because of a faulty assumption in the theory such as, for example, that the perturbation velocities are much less than the rotational velocities in the fluid, then a theoretical re-assessment would be required to see if the observed  $\Delta$  could be significantly different from the theoretical  $\Delta$ . (The perturbation velocities cannot be much less than the rotational velocity near the outer cylinder, which is stationary.)

### 5.2. Use of $\Delta$ as a function of $\eta$ to determine $\alpha_i$

No physically meaningful result was obtained, that is,  $\alpha_2$  was always positive. If the conclusions of the previous paragraph are accepted the result could be attributed to the failure of the second-order fluid model. (See §5.6.) Even so figure 9 can be considered as an illustration of the potential accuracy of the method: because of the experimental uncertainty and the acute angles at which the curves meet, there will be a large range of possible values of the  $\alpha_i$ .

### 5.3. Use of $\Delta$ and $\bar{\epsilon}$ to determine $\alpha_i$

The large error in the measured value of  $\epsilon_c$  is not due to difficulty in the measurement of  $\epsilon_c$  but due to inherent variations in the forms the cells take up. Other workers (e.g. Coles 1965) have found variations in the number of cells that can form in what appear to be identical situations. Even so, the spread in the values of the  $\alpha_i$  obtained from relatively few (five to six) repeat measurements of  $\Delta$  and  $\bar{\epsilon}$  are more accurate than expected! That suggests that some other imponderable appears in the experiments, such as, let us suppose, favoured patterns of cell distributions (for solution and solvent alike) that recur in such a way that averages of  $\bar{\epsilon}$  are far more accurate than the collection of repeated values of  $\epsilon_c$  for solution and for solvent would indicate. So, in practice, the spread among the values obtained for the  $\alpha_i$  should be a guide to the accuracy rather than the spread among the values of  $\epsilon_c$ .

Positive values of  $\alpha_2$  were obtained for aqueous polyox solutions of the highest concentrations (table 1). Had the values of  $\mu$  as determined in the coaxial cylinder apparatus been used  $\Delta$  would have been negative and  $\alpha_2$  would then have been found to be negative. There is a sound argument for using the value of  $\mu$  determined *in situ* in this case because the viscosity of polyox solutions is notoriously shear dependent, but we have refrained from doing so for the sake of consistency. (The average shear rate in the Ostwald viscometer was about the same as that in the Couette apparatus but of course the shear rate at the capillary wall would be larger than the average.)

### 5.4. Use of $\Delta$ and $s_1/s_2$ to determine $\alpha_i$

Figure 7 shows that curves of constant  $s_1/s_2$  and constant  $\Delta$  cross approximately at right angles thus giving a better fix for the values of the  $\alpha_i$  than do the curves of figures 5 and 6. Thus for similar errors in  $\Delta$ ,  $\bar{\epsilon}$  and  $s_1/s_2$  this method of drag

reduction gives the most accurate determination of the  $\alpha_i$ . However, equation (9), relating  $\psi(\alpha_1, \alpha_2)$  to  $\alpha_1$  and  $\alpha_2$ , is only applicable to narrow-gap geometry and it is likely that the variation of the  $\alpha_i$  with gap size shown in table 3 is largely a consequence of the inapplicability of (9). But here again the exact form of (9) is not required for a discussion of the instrument as a means of determining the  $\alpha_i$ .

From Chan Man Fong's (1970*a*) work an exact expression may be obtained for  $\psi(\alpha_1, \alpha_2)$  for a narrow gap but the numerical solution of his form of the equation has not been attempted by us yet. Even so, for  $\eta = 0.95$  equation (9) must be as accurate as our experimental results because the  $\alpha_i$  determined from  $\Delta$  and  $\bar{e}$  agree with those determined from  $\Delta$  and  $s_1/s_2$ . If the theory were exact experimental difficulties could remain. In drawing the best straight lines for the Newtonian and non-Newtonian fluids through the same point on the abscissa (figure 9) we are assuming that energy imbalance which produces a discontinuity in the  $\phi P$ ,  $P^{-1}$  curves affects  $1 - Ta_c/Ta$  similarly, independently of the fluid, but perhaps that is justified by the discussion of  $r_1$ ,  $r_2$  and  $r_3$  in § 5.1, in that ratios of values of  $Ta$  are not affected by additional energy input (additional to the rotational energy). In this connexion it is interesting to see that Denn *et al.* (1971) draw best straight lines through the experimental points so that they pass through  $1 - Ta_c/Ta = 0$ . Inspection of their results will show that their experimental point would be a better fit on lines passing through a negative value of  $1 - Ta_c/Ta$  as in figure 9.

A further feature of our results to be commented on is the values we find for  $\delta$ . The theoretical value of  $\delta$  for narrow-gap geometry is 1.53 and when  $\eta = 0.5$  the value is 0.83 (Davey 1962). The values we find when  $\eta = 0.95$ , 0.925 and 0.90 are 1.09, 1.15 and 1.61 respectively. It is relevant to note that the best straight lines do pass through  $Ta_c/Ta = 1$  when  $\eta = 0.90$  whereas for the other two gaps they do not. If the straight lines were drawn to pass through  $Ta_c/Ta = 1$  when  $\eta = 0.95$  and when  $\eta = 0.925$ , the slopes would be greater and near 1.5. It seems, therefore, that the presence of a discontinuity in the experiments has resulted in the slope of the best lines being smaller than that predicted theoretically, and when the discontinuity is absent, as in the case of  $\eta = 0.90$ , the experimentally determined value agrees with the theoretical prediction. It is also pertinent to note that experimental and theoretical values of  $Ta_c$  agree (figure 4*c*) when  $\eta = 0.90$ , consistent with our view that discontinuities and the disagreement between  $Ta_c$  values are both related to energy fed in other than from rotation. But as shown in § 5.1 the values of  $\Delta$  are not affected.

It might now be asked whether, since experimentally  $\delta \neq 1.53$ , the theory being applied to determine  $\psi(\alpha_1, \alpha_2)$  is quantitatively applicable. To answer the fact, the ratio of the slopes of the curves for the Newtonian and non-Newtonian fluids has been used so that differences between theory and experiment in a multiplying constant such as  $\delta$  should cancel out.

### 5.5. Considerations concerning the improvement of the apparatus

The most accurate of the three methods of determining the  $\alpha_i$ , given an accurate numerical solution of the equations involved, is that involving  $\Delta$  and  $s_1/s_2$ . All the measurements here are measurements of torque alone. At  $Ta_c$  the magnitude

of the torque per unit length of cylinder depends on  $\eta$  only so there is no optimum radius for accuracy in determining  $\Delta$ . Also the ratio  $s_1/s_2$  is independent of  $R_1$  and  $R_2$  and of  $h$  and no increase in sensitivity is obtained by changing the dimensions. It is true that  $G$  increases with  $h$  but as  $h$  increases so does the necessity of a more robust suspension and so a loss of sensitivity in measuring  $\phi$ .

Figures 5, 6 and 7 show that the  $\alpha_i$  are more accurately determinable by all the methods as  $\eta$  increases. We have tried an inner cylinder in our apparatus such that  $\eta = 0.98$  but we have found it tedious to align and then the speed of rotation is such that as  $\Omega \rightarrow \Omega_c$  the mechanical vibrations lead to fluctuations which are prohibitively large; and the outer cylinder sticks. We have tried no experiment with cylinders giving values of  $\eta$  between 0.95 and 0.98 but it would obviously be advantageous to discover from practical experience which value of  $\eta$  would be the largest usable.

As  $\eta$  increases so does the rate of shear at the critical value of the angular speed and consequently so does the frictional heating per unit volume at  $\Omega_c$  increase with  $\eta$ . An order-of-magnitude calculation will show that when  $\eta = 0.98$  (assuming it could be set up in our apparatus) all the heat generated could be dissipated by a temperature differential within the liquid of  $\simeq 10^{-4}^\circ\text{C}$ , for the dilute solutions having viscosities within 40% of that water. For a larger apparatus the temperature differential would be less. So any misgivings concerning frictional heating in the narrower gaps can be set aside for dilute polymer solutions, in considering the design of the apparatus.

A further guide line for the design comes about from consideration of the likely range of values of the  $\alpha_i$  to be measured. Figures 6 and 7 show that if  $|\alpha_2| < +2.5 \text{ mg m}^{-1}$  then the apparatus should be designed such that  $\eta > 0.95$  and from considerations already given this would imply larger radii of cylinders to yield a wider gap (in absolute terms) to facilitate the setting up of the apparatus. Also as already stated torque measurements involving  $Ta_c$  and the drag during secondary flow are the most sensitive and consequently the design of apparatus should be a suspended outer cylinder on a torsion fibre. But a note of caution must be struck. Calculation of  $\Delta$  and  $\bar{e}$  for  $\eta = 0.98$  (figure 11) for low values of the  $\alpha_i$  illustrates how sensitive the Taylor instabilities are then to elasticity, but the figure also shows that the same value of  $\Delta$  can be obtained for different  $\alpha_i$  and only  $s_1/s_2$  (or  $\bar{e}$ ) fixes the values of the  $\alpha_i$ ; the measurement of  $\bar{e}$  is relatively inaccurate, so ambiguities could arise. But values of  $\Delta$  of order unity would be observed for the values of the  $\alpha_i$  which we have observed in our experiments. The design of the apparatus would then involve a large range of rotational speeds with associated difficulties of adjustments and accompanying vibration. A value of  $\Delta$  of 0.50 could be measured to within 2%, that is  $\pm 0.01$ , and that would be adequate;  $\Delta = 0.50$  when  $0.95 < \eta < 0.98$ . Further calculation would indicate the value of  $\eta$  for the range of  $\alpha_i$  to be measured.

Should it be necessary to measure  $\bar{e}$  rather than  $s_1/s_2$  to determine  $\alpha_i$  then  $\Delta$  could be measured in an apparatus such as ours and a long coaxial cylinder apparatus built with the same values of  $R_1$  and  $R_2$  to determine the cell size. The restriction on the length would be the number of cells to be observed to attain the required accuracy.

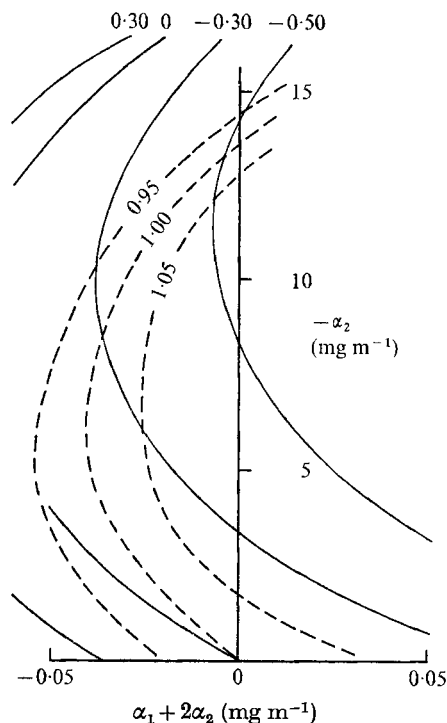


FIGURE 11. Theoretical curves of constant  $\Delta$  (solid lines) and of constant  $\bar{\Delta}$  (broken lines) as a function of low values of the  $\alpha_i$ .  $\eta = 0.98$ .

### 5.6. Comments on the results

It has been necessary to determine  $Ta_c$  to within better than 1% and it can be seen that relatively imprecise measurements could lead to very dissimilar results and that is the likely reason for the conflicting reports in the literature concerning the stability of dilute polymer solutions. Also, it is apparent from our results that a small amount of disturbance of the flow (due to slots) which does not alter the value of  $Ta_c$  by more than 1% does influence the value of  $\Delta$ , in that destabilization was observed in some instances whereas without the slots there was stabilization in all cases. Lastly, as mentioned above, we found visual methods of determining  $Ta_c$  to be too inaccurate to be considered.

In precise experiments (measurement of  $Ta_c$  to within better than 1%)  $\Delta$  is (i) positive, (ii) increases with concentration and (iii) increases as  $\eta$  decreases (table 3). The  $|\alpha_i|$  (i) increase with increasing concentration and (ii) increase with increasing temperature.  $\lambda$  is always negative and  $|\lambda|$  lies between 0.02 and 0.07. There is evidence that the  $\alpha_i$  are functions of  $\gamma$  in that determining  $\Delta$  as a function of  $\eta$  gives no negative value for  $\alpha_2$ . (The fact that the  $\alpha_i$  vary with  $\eta$  when determined from  $\Delta$  and  $s_1/s_2$  could be a misapplication of the theory.)

Many more experiments have been done with polyacrylamide solutions at 25°C than with any other solution at any other temperature. Average values of the  $\alpha_i$  with r.m.s. deviations, found from all methods giving negative values for  $\alpha_2$ ,

Concentration (p.p.m.)	$-(\alpha_1 + 2\alpha_2)$ (mg m <sup>-1</sup> )	$-\alpha_2$ (mg m <sup>-1</sup> )	$-\lambda$
50	$0.26 \pm 0.11$	$6.5 \pm 0.9$	0.02
100	$0.39 \pm 0.11$	$9.0 \pm 1.0$	0.02
250	$0.75 \pm 0.04$	$20.8 \pm 0.2$	0.02

TABLE 4. Summary of the values found for the material constants of aqueous polyacrylamide solutions at 25 °C.

are given in table 4. The relatively low deviation for the 250 p.p.m. solution results because it was used as a test solution in trying out experiments with polymer solutions so there are many more results. It is seen that  $\lambda$  is constant, small (0.02) and negative. Tanner (1972) reported on several measurements of the ratio of the normal stress coefficients (but of more concentrated solutions) and quoted values of around  $-0.15$  to  $-0.05$ .

Concerning the validity of the second-order equation, then, the equation is satisfactory in that meaningful results (just cited) are obtained from measurement of  $\Delta$ ,  $\bar{\epsilon}$  and  $s_1/s_2$  in the same gap but is unsatisfactory in that when the  $\alpha_i$  are determined from  $\Delta$  as a function of  $\eta$  meaningful results are not obtained. The shear times  $\gamma^{-1}$  in the three gaps (30, 15 and 5 ms respectively) were of the order of the relaxation times (10–50 ms) given by  $2\alpha_2/\alpha_0$ . So elastic effects which are a function of the gap size might be expected. If a third-order equation were necessary to describe fluid behaviour there should be some dependence of  $\alpha_0$  on  $\gamma$  and there is evidence for that in the more concentrated solutions. Use of the third-order equation will require the evaluation of five material constants and therefore measurement of five experimental variables; those would be  $\mu$ ,  $\Delta$ ,  $\eta$ ,  $\bar{\epsilon}$  and  $s_1/s_2$ . When the effect of elasticity on the stability of the azimuthal wavy structure that occurs at  $1.5Ta_c$  is calculated using principles discussed by DiPrima (1961) and Davey, DiPrima & Stuart (1968) then a sixth experimental variable could be introduced. The present state of our work is the application of third-order theory to our results to see if the ‘constants’ of the third-order equation are in fact constant, over a wider range of conditions than are the constants of the second-order equation.

## 6. Conclusion

Given an accurate theory involving just two material constants (other than shear viscosity), the study of Taylor instabilities is a sensitive means of evaluating them, capable of determining values of order 1 mg m<sup>-1</sup>. The most sensitive combination of measurements to use is that of the change in the Taylor number together with that of drag reduction in secondary flow.  $\eta$  should be chosen so that  $\Delta \simeq 0.50$  for the particular  $\alpha_i$ .

The authors are grateful to Dr K. Walters of the Department of Applied Mathematics of this College, for his advice and help in many valuable discussions.

## REFERENCES

- BAILEY, B. J. 1969 *Nature*, **222**, 373.
- CHANDRASEKHAR, S. 1953 *Proc. Roy. Soc. A* **216**, 293.
- CHANDRASEKHAR, S. 1958 *Proc. Roy. Soc. A* **246**, 301.
- CHAN MAN FONG, C. F. 1965 *Rheol. Acta*, **4**, 37.
- CHAN MAN FONG, C. F. 1970a *Appl. Sci. Res.* **23**, 16.
- CHAN MAN FONG, C. F. 1970b *Z. angew. Math. Phys.* **21**, 977.
- COLES, D. 1965 *J. Fluid Mech.* **21**, 385.
- DAVEY, A. 1962 *J. Fluid Mech.* **14**, 336.
- DAVEY, A., DIPRIMA, R. C. & STUART, J. T. 1968 *J. Fluid Mech.* **31**, 17-52.
- DAVIES, D. M. 1968 M.Sc. thesis, University of Wales.
- DAVIES, D. M. 1972 Ph.D. thesis, University of Wales.
- DEBLER, W., FÜNER, E. & SCHAAF, B. 1968 *Appl. Mech. Proc. 12th Int. Cong. Appl. Math.* p. 158.
- DENN, M. M. & ROISMAN, J. J. 1969 *A.I.Ch.E. J.* **15**, 454.
- DENN, M. M., SUN, Z.-S. & RUSHTON, B. D. 1971 *Trans. Soc. Rheol.* **15**, 415.
- DIPRIMA, R. C. 1960 *J. Fluid Mech.* **9**, 621.
- DIPRIMA, R. C. 1961 *Phys. Fluids*, **4**, 751.
- DONNELLY, R. J. 1958 *Proc. Roy. Soc. A* **246**, 312.
- DONNELLY, R. J. 1965 *Proc. Roy. Soc. A* **283**, 509.
- DONNELLY, R. J. & FULTZ, D. 1960 *Proc. Roy. Soc. A* **258**, 101.
- DONNELLY, R. J. & SCHWARZ, K. W. 1965 *Proc. Roy. Soc. A* **283**, 531.
- DONNELLY, R. J. & SIMON, N. J. 1960 *J. Fluid Mech.* **7**, 401.
- GIESEKUS, H. 1966 *Rheol. Acta*, **5**, 239.
- GINN, R. F. & DENN, M. M. 1969 *A.I.Ch.E. J.* **15**, 450.
- GODDARD, J. D. & MILLER, C. 1967 *University of Michigan Tech. Rep.* no. 06673-8-T.
- GRAEBEL, W. P. 1961 *Phys. Fluids*, **4**, 362.
- GRIFFITHS, J. D. & THOMAS, R. H. 1966 *J. Mécanique*, **5**, 101.
- HAYES, J. W. & HUTTON, J. F. 1970 *Progr. Heat Transfer*, **5**, 195.
- JONES, W. M. & MARSHALL, D. E. 1969 *J. Phys. D* **2**, 809.
- KARLSSON, S. K. F., SOKOLOV, M. & TANNER, R. I. 1971 *Chem. Engng Prog. Symp. Ser.* **67**, 11.
- LEWIS, J. W. 1928 *Proc. Roy. Soc. A* **117**, 388.
- LOCKETT, F. J. & RIVLIN, R. S. 1968 *J. Mécanique*, **7**, 475.
- MARSHALL, D. E. 1967 M.Sc. thesis, University of Wales.
- MERRILL, E. W., MICKLEY, H. S. & RAM, A. 1962 *J. Fluid Mech.* **13**, 86.
- OLDBROYD, J. G. 1950 *Proc. Roy. Soc. A* **200**, 523.
- RUBIN, H. & ELATA, C. 1966 *Phys. Fluids*, **9**, 1929.
- RUBIN, H., ELATA, C. & POREH, M. 1968 *Rheol. Acta*, **7**, 340.
- SMITH, M. M. & RIVLIN, R. S. 1972 *J. Mécanique*, **11**, 69.
- SONG, C. S. & TSAI, P. Y. 1966 *University of Minnesota, Project Rep.* no. 84.
- STUART, J. T. 1958 *J. Fluid Mech.* **4**, 1.
- TANNER, R. I. 1972 *VIth Int. Cong. Rheol.*, Lyons.
- TAYLOR, G. I. 1923 *Trans. Roy. Soc. A* **223**, 289.
- TAYLOR, G. I. 1936 *Proc. Roy. Soc. A* **157**, 546.
- THOMAS, R. H. & WALTERS, K. 1964 *J. Fluid Mech.* **18**, 33.