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## FREE SURFACES INDUCED BY THE MOTION OF VISCOELASTIC FLUIDS

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Free surfaces are sensitive to the state of stress in fluids. The striking variations in the shape of free surfaces induced by the motion of viscoelastic fluids chart the competing effects of elasticity, normal stresses and inertia in the fluid. We would like to know how to decipher these charts because the deciphering challenges our understanding of mechanics and introduces new methods of rheometry with good potential for characterizing important properties of viscoelastic fluids.

The analysis of the shape of free surfaces on moving viscoelastic fluids is plagued by two difficulties: the domain of flow is unknown and the constitutive equation is unknown except when evaluated on certain asymptotic limits of allowed dynamic processes, like viscometric flow and slow flow. And even in these asymptotic limits the constitutive equations are known only up to undetermined constants and functions whose values are the proper goal of rheometrical measurements.

The problem of the shape of free surfaces induced by motions of viscoelastic fluids which perturb the state of rest is the most congenial to analysis because general, but tractable, representations of the stress in terms of integrals (with unknown kernels) may be assumed and because the nonlinear problem of free surfaces perturbing the rest state can be studied by the method of domain perturbations. We may describe this type of analysis as the simultaneous perturbation of the domain and constitutive equation. To explain the principle behind this type of analysis we turn now to a conceptually simple problem, free of rheological complications, in which the principles involved may be clearly seen.

#### 1. A MODEL PROBLEM FOR DOMAIN PERTURBATIONS OF THE REST STATE

We suppose that there is an analytic function  $\mathcal{F}(\phi)$  which is not known except that in a neighborhood of  $\phi = 0$  it has a Taylor series

$$\mathcal{F}(\phi) = \frac{1}{2} a \phi^2 + \frac{1}{3!} b \phi^3 + \dots \quad (1.1)$$

where  $a = \mathcal{F}_{\phi\phi}(0)$  and  $b = \mathcal{F}_{\phi\phi\phi}(0)$ . We may think that  $\mathcal{F}(\phi)$  is representative of the nonlinear part of the stress in some fictitious material. Our idea is to find the Taylor coefficients for (1.1) by measuring the free surface induced by a dynamical process

$$F(\underline{x}) = \nabla^2 \phi + \mathcal{F}(\phi) = 0 \quad \text{in } \mathcal{V}_\delta \quad (1.2)$$

subject to the condition that

$$G(\underline{x}, \epsilon) = \phi(\underline{x}) - g(\underline{x}, \epsilon) \quad \text{on } \partial\mathcal{V}_\delta \quad (1.3)$$

where  $\mathcal{V}_\delta$  is a bounded region of space depending on a parameter  $\delta$  and  $g(\underline{x}, \epsilon)$  are given data depending on a parameter  $\epsilon$ .

It is instructive to carry out our analysis in easy stages. First we perturb  $\epsilon$ , leaving  $\delta$  fixed and assume that

$$\phi(\underline{x}, \epsilon) = \sum_0^k \frac{\epsilon^l}{l!} \phi_l(\underline{x}) \quad (1.4)$$

where, in  $\mathcal{V}_\delta$ ,

$$\nabla^2 \phi_0 + \mathcal{F}(\phi_0) = 0$$

$$\nabla^2 \phi_1 + \mathcal{F}_{\phi}(\phi_0) \phi_1 = 0 \quad (1)$$

$$\nabla^2 \phi_2 + \mathcal{F}_{\phi}(\phi_0) \phi_2 + \mathcal{F}_{\phi\phi}(\phi_0) \phi_1^2 = 0$$

and, on  $\partial\mathcal{V}_\delta$ ;

$$G_n = \phi_n(\underline{x}) - g_n(\underline{x}) = 0, \quad (1)$$

where  $g_n(\underline{x})$  is nth derivative of  $g(\underline{x}, \epsilon)$  at  $\epsilon = 0$ . If we could solve (1.5) and (1.6) for  $\phi_0$  we could find  $\phi_1, \phi_2$ , etc. as the solution of linear boundary value problems. It would be hard to solve (1.5)<sub>1</sub>, even if we knew  $\mathcal{F}(\phi_0)$ , because it is nonlinear. But we cannot solve (1.5)<sub>1</sub> at all because we don't know  $\mathcal{F}(\phi_0)$  except as a power series in a small neighborhood of zero.

The "rest state" is now defined as the state for which  $g_0(\underline{x})|_{\partial\mathcal{V}_\delta} = 0$ . We couple this definition with the assumption that (1.5)<sub>1</sub> has no solutions  $\phi_0 \neq 0$  when  $\phi_0(\underline{x})|_{\partial\mathcal{V}_\delta} = 0$ . Then  $\phi_0(\underline{x}) \equiv 0$  in  $\mathcal{V}_\delta$  and, replacing (1.5)<sub>2,3</sub> we get

$$\nabla^2 \phi_1|_{\mathcal{V}_\delta} = 0, \quad \phi_1|_{\partial\mathcal{V}_\delta} = g_1;$$

$$\nabla^2 \phi_2 + a \phi_1^2|_{\mathcal{V}_\delta} = 0, \quad \phi_2|_{\partial\mathcal{V}_\delta} = g_2$$

etc. These problems are linear and easy to solve even when a unknown (in fact  $\phi_2 = \phi_{21} + a\phi_{22}$ , where  $\phi_{21}$  and  $\phi_{22}$  are independent of  $a$ ). So we can use the solution which perturbs the rest state to find  $a$  and, with more work, we can get expressions which involve the other Taylor coefficients of (1.1) as well.

Now suppose that  $\epsilon$  is fixed and  $\delta$  varies. And suppose further that  $\mathcal{V}_0$  is some convenient domain whose point of convenience is, say, that  $\mathcal{V}_0$  has a high degree of symmetry. We are going to try to solve the dynamic problem in  $\mathcal{V}_\delta$  as a series whose coefficients can be determined from boundary value problems posed on the symmetric domain  $\mathcal{V}_0$ .

First we map  $\mathcal{V}_\delta$  into  $\mathcal{V}_0$  with an invertible one to one mapping, which is analytic in  $\delta$  and which takes points on  $\partial\mathcal{V}_\delta$  into  $\partial\mathcal{V}_0$ :

$$\underline{x} = \underline{x}(\underline{x}_0, \delta) = \sum \frac{\delta^n}{n!} \underline{x}^{[n]}(\underline{x}_0) \quad (\text{analytic in } \delta),$$

$$\underline{x}_0 = \underline{x}(\underline{x}_0, 0) \quad (\text{identity}),$$

$$\underline{x}_0 = \underline{x}^{-1}(\underline{x}, \delta) \quad (\text{inverse}),$$

$$\partial\mathcal{V}_\delta \leftrightarrow \partial\mathcal{V}_0$$

Let  $H(\underline{x}, \delta)$  be any function defined in the family of domains  $\mathcal{V}_\delta$  and introduce the notation:

$$\tilde{H}(\delta) = H(\underline{x}(\underline{x}_0, \delta), \delta),$$

$$H^{[n]}(\underline{x}_0) = \frac{d^n}{d\delta^n} \tilde{H}(0)$$

and

$$H^{<n>}(\underline{x}_0) = \left. \frac{\partial^n H(\underline{x}, \delta)}{\partial \delta^n} \right|_{\substack{\delta=0 \\ \underline{x}=\underline{x}_0}}$$

Connection formulas (the chain rule) connect  $H^{[n]}$  and  $H^{<n>}$ :

$$H^{[0]}(\underline{x}_0) = H^{<0>}(\underline{x}_0) = H(\underline{x}_0, 0),$$

$$H^{[1]}(\underline{x}_0) = H^{<1>}(\underline{x}_0) + \underline{x}^{[1]} \cdot \nabla H^{<0>},$$

$$H^{[2]}(\underline{x}_0) = H^{<2>}(\underline{x}_0) + 2\underline{x}^{[1]} \cdot \nabla H^{<1>} + \underline{x}^{[2]} \cdot \nabla H^{<0>} + (\underline{x}^{[1]} \cdot \nabla)^2 H^{<0>},$$

(1.9)

etc. It follows from the equations that

$$\tilde{F}(\delta) = F(\underline{x}(\underline{x}_0, \delta), \delta) = 0$$

when  $\underline{x} \in \mathcal{V}_\delta$ ,  $\underline{x}_0 \in \mathcal{V}_0$ , and that

$$\tilde{F}^{[n]}(0) = F^{[n]}(\underline{x}_0) = 0 \text{ in } \mathcal{V}_0. \quad (1.10)$$

We may easily establish by mathematical induction, using the connection formulas, that

$$F^{<n>}(\underline{x}_0) = 0 \text{ in } \mathcal{V}_0. \quad (1.11)$$

For example,  $F^{<0>}(\underline{x}_0) = 0$  in  $\mathcal{V}_0$  and  $F^{[1]}(\underline{x}_0) = F^{<1>}(\underline{x}_0) + \underline{x}^{[1]} \cdot \nabla F^{<0>}(\underline{x}_0) = F^{<1>}(\underline{x}_0) = 0$ . It is a bit more complicated at the boundary. Since  $G(\delta) = 0$  on  $\partial \mathcal{V}_\delta$ ,  $G^{[m]}(\underline{x}_0) = 0$  on  $\partial \mathcal{V}_0$  and tangential derivatives of  $G^{[m]}$  on  $\mathcal{V}_0$  must vanish but normal derivatives need not vanish. It follows that on  $\partial \mathcal{V}_0$

$$G^{[m]}(\underline{x}_0) = \left\{ \left( \frac{\partial}{\partial \epsilon} + \nu_n \frac{\partial}{\partial n} \right)^m G(\underline{x}, \epsilon) \right\}_{\substack{\epsilon=0 \\ \underline{x}=\underline{x}_0}} = 0 \quad (1.12)$$

where  $\underline{n}$  is the outward normal to  $\mathcal{V}_\delta$ ,  $\nu_n = \underline{x}^{[1]} \cdot \underline{n}$  and  $\underline{n} \cdot \nabla = \partial / \partial n$

We may seek the solution of (1.2) and (1.3) in  $\mathcal{V}_\delta$  as a series defined in  $\mathcal{V}_0$

$$\phi(\underline{x}(\underline{x}_0, \delta), \delta) = \sum_0^{\infty} \frac{\delta^n}{n!} \phi^{[n]}(\underline{x}_0) \quad (1.13)$$

where, in  $\mathcal{V}_0$ ,

$$\nabla^2 \phi^{<0>} + \mathcal{F}(\phi^{<0>}) = 0$$

$$\nabla^2 \phi^{<1>} + \mathcal{F}_\phi(\phi^{<0>}) \phi^{<1>} = 0 \quad (1.14)$$

$$\nabla^2 \phi^{<2>} + \mathcal{F}_\phi(\phi^{<0>}) \phi^{<2>} + \mathcal{F}_{\phi\phi}(\phi^{<0>}) \phi^{<1>^2} = 0$$

etc., and on  $\partial \mathcal{V}_0$

$$G^{[n]} = \phi^{[n]}(\underline{x}_0) - g^{[n]}(\underline{x}_0). \quad (1.15)$$

The problems (1.14), (1.15) are like (1.5) and (1.6). We can't solve them because  $\mathcal{F}(\phi)$  is given only as a Taylor series (1.1) with unknown coefficients and an unknown circle of convergence.

The "rest state" for the domain perturbation, like the "rest state" for the perturbation of the boundary data may be defined by the condition  $g^{[0]}(\underline{x}_0) |_{\partial \mathcal{V}_0} = 0$ . This condition implies that  $\phi^{[0]} = \phi^{<0>} = 0$  on  $\partial \mathcal{V}_0$ , hence,  $\phi^{<0>} = 0$  in  $\mathcal{V}_0$  and

$$\nabla^2 \phi^{<1>} |_{\mathcal{V}_0} = 0, \quad \phi^{<1>} |_{\partial \mathcal{V}_0} = g^{[1]}(\underline{x}_0) \quad (1.16)$$

$$\nabla^2 \phi^{<2>} + a \phi^{<1>^2} |_{\mathcal{V}_0} = 0, \quad \phi^{[2]} |_{\partial \mathcal{V}_0} = \phi^{<2>} + 2\nu_n \frac{\partial}{\partial n} \phi^{<1>} = g^{[2]}(\underline{x}_0) \quad (1.17)$$

etc. The linear problems (1.16), (1.17) and higher order problems are solvable and not too hard to actually solve, even when  $a$  is unknown.

In our rheological problems the boundary data ( $\epsilon$  in our first example) perturb the boundary ( $\delta$  in our second example). So we may put  $\epsilon = \delta$  and construct a simple example of a domain perturbation of the rest state with a free surface. By a free surface we understand that there is a one ( $\epsilon$ ) parameter family of domains  $\mathcal{V}_\epsilon$  which are unknown. Supposing now that our dynamical process (1.1) and (1.2) hold in  $\mathcal{V}_\epsilon$  we might expect solutions in each and every  $\mathcal{V}_\epsilon$  corresponding to some possibly small  $\epsilon$  interval of the origin. But no, this will not be possible because in addition to (1.2) we pose an additional boundary condition, which is analogous to, but much simpler than, the condition that the jump in the normal component of stress is balanced by surface tension times mean curvature. Because we have this extra condition we can't solve (1.1) and (1.2) in every  $\mathcal{V}_\epsilon$ ; the extra condition can be satisfied only when  $\mathcal{V}_\epsilon$  is properly chosen.

As an example of the foregoing consider the two dimensional problem specified in polar coordinates  $(r, \theta)$  in Fig. 1.1 where the boundary data  $g(\theta, \epsilon)$  are given and correspond to a rest state  $g(\theta, 0) = g^{<0>}(\theta) = 0$ . The dynamical process  $\phi(r, \theta, \epsilon)$  and the function  $f(\theta, \epsilon)$  which gives the boundary  $r = 1 + f(\theta, \epsilon)$  of  $\mathcal{V}_\epsilon$  are unknown.

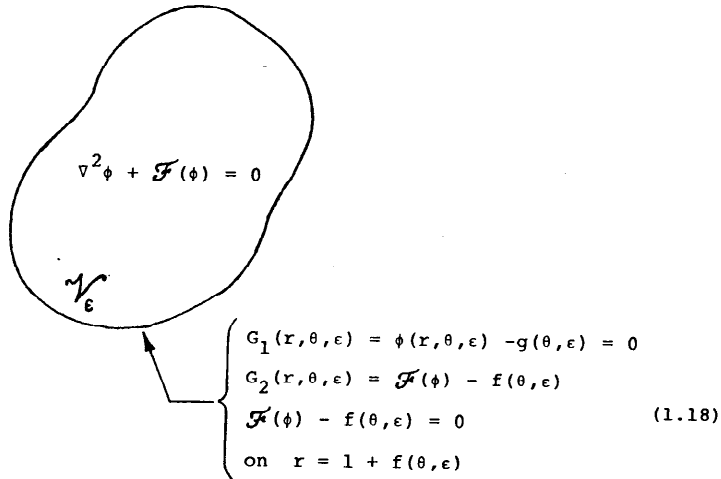


Fig. 1.1 Model of a free surface problem

We remind the reader that our aim is to show how to find  $\mathcal{F}(\phi)$ ; that is, the Taylor coefficients in (1.1) by (fictitious) experimental measurements of the (made up) boundary  $r = 1 + f(\theta, \epsilon)$ .

First we solve (1.18) when  $\epsilon = 0$  and we find that  $\nabla^2 \phi + \mathcal{F}(\phi) = 0$  in  $\mathcal{V}_0$  with  $\phi(r, \theta, 0) = 0$  on  $r = 1 + f(\theta, 0)$  has  $\phi \equiv 0$  in  $\mathcal{V}_0$ . Then, since  $\mathcal{F}(0) = 0, f(\theta, 0) = 0$  so that the reference configuration  $\mathcal{V}_0$  is the unit circle  $r_0 = 1$ . We seek the solution of (1.18) in powers of  $\epsilon$

$$\begin{pmatrix} \phi(r, \theta, \epsilon) \\ f(\theta, \epsilon) \end{pmatrix} = \sum_0^{\infty} \frac{\epsilon^n}{n!} \begin{pmatrix} \phi^{[n]}(r_0, \theta_0) \\ f^{[n]}(\theta_0) \end{pmatrix}$$

where  $f^{[0]}(\theta_0) = \phi^{[0]}(r_0, \theta_0) = 0$  and  $\mathcal{V}_\epsilon$  and  $\mathcal{V}_0$  are related by a shifting map

$$\theta = \theta_0$$

$$r = r_0(1 + f(\theta_0, \epsilon))$$

having all the properties required of (1.8). For the shifting map the deformation of  $\mathcal{V}_0$  is along rays and

$$\underline{x}^{[n]} = \underline{e}_r r^{[n]} = \underline{e}_r r_0 f^{[n]}(\theta_0).$$

Note that for any function  $f(\theta)$  of  $\theta = \theta_0$  alone, we have  $f^{<n>}(\theta) = f^{[n]}(\theta)$ . Using the connection formulas (1.9) we find that

$$\phi^{[1]}(r_0, \theta_0) = \phi^{<1>}(r_0, \theta_0)$$

and

$$\phi^{[2]}(r_0, \theta_0) = \phi^{<2>}(r_0, \theta_0) + 2r_0 f^{[1]}(\theta_0) \frac{\partial \phi^{<1>}}{\partial r_0}(r_0, \theta_0).$$

On the boundary of  $\mathcal{V}_0$ ,  $r_0 = 1$  we have, from (1.18)<sub>3</sub> that  $\mathcal{F}_\phi(0) \phi^{<1>}(1, \theta_0) - f^{[1]}(\theta_0) = 0$ . Since  $\mathcal{F}_\phi(0) = 0$ , we find that  $f^{[1]}(\theta_0) = 0$ . The boundary value problems satisfied by  $\phi^{<1>}(r_0, \theta_0)$  and  $\phi^{<2>}(r_0, \theta_0)$  are given in Figs 1.2 and 1.3.

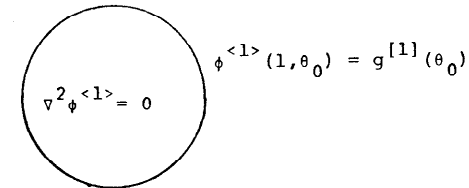


Fig. 1.2 The problem satisfied by  $\phi^{<1>}(r_0, \theta_0)$

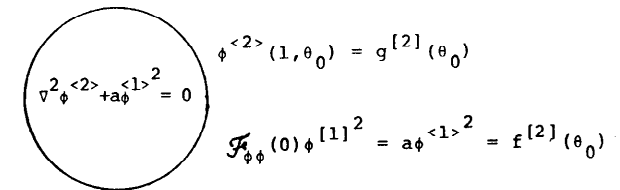


Fig. 1.3 The problem satisfied by  $\phi^{<2>}(r_0, \theta_0)$

These problems are easy to solve. We find from Fig. 1.3 that

$$f^{[2]}(\theta_0) = a \phi^{<1>^2}(1, \theta_0)$$

where  $\phi^{<1>}(r_0, \theta_0)$  is the solution of the problem shown in Fig. 1.2. It follows that the first approximation to the shape of  $\mathcal{V}_\epsilon$  is given by

$$r = 1 + f(\theta, \epsilon) = 1 + a\phi^{<1>2}(1, \theta)\epsilon^2 + o(\epsilon^3).$$

The next approximation depends on b as well as a. We may therefore deduce the values of derivatives of  $\mathcal{F}(\phi)$  at  $\phi = 0$  by monitoring the changes in the shape of  $\mathcal{V}_\epsilon$  when  $\epsilon$  is near to zero.

## 2. CONSTITUTIVE EXPRESSIONS WHICH PERTURB THE REST STATE OF A SIMPLE FLUID

In our rheological studies the role of the non-linear function  $\mathcal{F}(\phi)$  is assumed by the nonlinear functional

$$\mathcal{F}[\underline{G}(s)] = \underline{T} + p\underline{1} \quad (2.1)$$

giving the constitutively determined part of the stress  $\underline{T}$  of an incompressible fluid. The argument functions in the domain of  $\mathcal{F}$  are symmetric tensor valued functions of  $s=t-\tau$  for fixed t and  $\underline{x}$ , called histories,

$$\underline{G}(s) = \underline{F}_t^T(\tau) \cdot \underline{F}_t(\tau) - \underline{1}, \quad \underline{G}(0) = 0$$

which are defined on the relative deformation tensor  $\underline{F}_t(\tau) = \nabla_{\underline{x}} \underline{X}_t(\underline{x}, \tau)$  where  $\underline{X}_t(\underline{x}, \tau) = \underline{g}(\underline{X}, \tau)$  is the position vector of the particle  $\underline{X}$  which is presently at  $\underline{x}$ . Equation (2.1), which assumes that the stress depends only on the first spatial gradient of the deformation, is still too general to be used to solve the problems which lead to understanding how viscoelastic fluids respond to applied forces.

It is well known that the extra stress vanishes on the zero history  $\mathcal{F}(0) = 0$ . It is then natural to seek expansions of  $\mathcal{F}(\underline{G}(s))$  in terms of functional expansions on the rest history

$$\begin{aligned} \mathcal{F}[\underline{G}(s)] &= \mathcal{F}[0; \underline{G}(s)] + \mathcal{F}_1[0; \underline{G}(s), \underline{G}(s)] \\ &+ \mathcal{F}_2[0; \underline{G}(s), \underline{G}(s), \underline{G}(s)] + \dots \end{aligned} \quad (2.2)$$

where

$$\mathcal{F}_n[0; \underline{G}_1(s), \underline{G}_2(s), \dots, \underline{G}_n(s)]$$

is an n linear form. We shall assume (Green and Rivlin (1), Coleman and Noll (2), Pipkin (3)) that these n-linear forms may be represented by iterated integrals of the form

$$\int_0^\infty \dots \int_0^\infty K_{ijkl\dots mnv}(s_1, s_2, \dots, s_n) G_{kl}(s_1) \dots G_{mv}(s_n) ds_1 ds_2 \dots ds_n.$$

It is very easy to find the isotropic forms of these integrals from invariance theory for a single tensor  $\underline{G}(s)$  (see Exercise 94.7 in Joseph, (4)). If we introduce the notation  $\mathcal{F}^{(n)}(\underline{G}(s))$  for the partial sum of (2.2) after n terms it is easy to show

that for  $n = 1, 2, 3, 4$ , we get

$$\mathcal{F}^{(1)} = \int_0^\infty \zeta(s) \underline{G}(s) ds, \quad (1)$$

$$\begin{aligned} \mathcal{F}^{(2)} &= \mathcal{F}^{(1)} + \int_0^\infty \int_0^\infty \{ \beta(s_1, s_2) \underline{G}(s_1) \cdot \underline{G}(s_2) \\ &+ \alpha(s_1, s_2) [\text{tr } \underline{G}(s_1)] \underline{G}(s_2) \} ds_1 ds_2, \end{aligned} \quad (1)$$

$$\begin{aligned} \mathcal{F}^{(3)} &= \mathcal{F}^{(2)} + \int_0^\infty \int_0^\infty \int_0^\infty \{ \psi_1(s_1, s_2, s_3) \underline{G}(s_1) \cdot \underline{G}(s_2) \cdot \underline{G}(s_3) \\ &+ \psi_2(s_1, s_2, s_3) [\text{tr } \underline{G}(s_1)] \underline{G}(s_2) \cdot \underline{G}(s_3) \\ &+ \psi_3(s_1, s_2, s_3) [\text{tr } \underline{G}(s_1)] [\text{tr } \underline{G}(s_2)] \underline{G}(s_3) \\ &+ \psi_4(s_1, s_2, s_3) \text{tr}[\underline{G}(s_1) \cdot \underline{G}(s_2)] \underline{G}(s_3) \} ds_1 ds_2 ds_3, \end{aligned}$$

$$\begin{aligned} \mathcal{F}^{(4)} &= \mathcal{F}^{(3)} + \int_0^\infty \int_0^\infty \int_0^\infty \int_0^\infty \{ \phi_1(s_1, s_2, s_3, s_4) \underline{G}(s_1) \cdot \underline{G}(s_2) \cdot \underline{G}(s_3) \cdot \underline{G}(s_4) \\ &+ \phi_2(s_1, s_2, s_3, s_4) [\text{tr } \underline{G}(s_1)] \underline{G}(s_2) \cdot \underline{G}(s_3) \cdot \underline{G}(s_4) \\ &+ \phi_3(s_1, s_2, s_3, s_4) [\text{tr } \underline{G}(s_1)] [\text{tr } \underline{G}(s_2)] \underline{G}(s_3) \cdot \underline{G}(s_4) \\ &+ \phi_4(s_1, s_2, s_3, s_4) [\text{tr } \underline{G}(s_1)] [\text{tr } \underline{G}(s_2)] [\text{tr } \underline{G}(s_3)] \underline{G}(s_4) \\ &+ \phi_5(s_1, s_2, s_3, s_4) [\text{tr}(\underline{G}(s_1) \cdot \underline{G}(s_2))] \underline{G}(s_3) \cdot \underline{G}(s_4) \\ &+ \phi_6(s_1, s_2, s_3, s_4) [\text{tr}(\underline{G}(s_1) \cdot \underline{G}(s_2))] [\text{tr } \underline{G}(s_3)] \underline{G}(s_4) \\ &+ \phi_7(s_1, s_2, s_3, s_4) [\text{tr}(\underline{G}(s_1) \cdot \underline{G}(s_2) \cdot \underline{G}(s_3))] \underline{G}(s_4) \} \\ &ds_1 ds_2 ds_3 ds_4 \end{aligned}$$

where  $\beta(s_1, s_2)$ ,  $\psi(s_1, s_2, s_3)$  and  $\phi(s_1, s_2, s_3, s_4)$  are unchanged transposition of arguments; e.g.,  $\beta(s_1, s_2) = \beta(s_2, s_1)$ . Higher order expressions giving  $\mathcal{F}^{(n)}$  ( $n > 4$ ) may be written down by spection.

A distinction is usually made between small strain expansions which lead to integral expressions for  $\mathcal{F}^{(n)}$  and slow motion expansions which lead to

$$\mathcal{F}^{(1)} = \nu \underline{A}_1, \quad (2.7)$$

$$\mathcal{F}^{(2)} = \alpha_1 \underline{A}_2 + \alpha_2 \underline{A}_1^2, \quad (2.8)$$

$$\mathcal{F}^{(3)} = \beta_1 \underline{A}_3 + \beta_2 (\underline{A}_2 \underline{A}_1 + \underline{A}_1 \underline{A}_2) + \beta_3 (\text{tr } \underline{A}_2) \underline{A}_1, \quad (2.9)$$

$$\mathcal{F}^{(4)} = \gamma_1 \underline{A}_4 + \gamma_2 (\underline{A}_3 \underline{A}_1 + \underline{A}_1 \underline{A}_3) + \gamma_3 \underline{A}_2^2 \quad (2.10)$$

$$+ \gamma_4 (\underline{A}_2 \underline{A}_1^2 + \underline{A}_1^2 \underline{A}_2) + \gamma_5 (\text{tr } \underline{A}_2) \underline{A}_2$$

$$+ \gamma_6 (\text{tr } \underline{A}_2) \underline{A}_1^2 + [\gamma_7 \text{tr } \underline{A}_3 + \gamma_8 (\text{tr } \underline{A}_1 \underline{A}_2)] \underline{A}_1$$

where  $\nu, \alpha_1, \dots, \gamma_8$  are constants and  $\underline{A}_n = \underline{A}_n[\underline{U}(\underline{x}, t)]$  are Rivlin-Ericksen tensors,

$$(\underline{A}_1)_{ij} = \frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}$$

and

$$(\underline{A}_{n+1})_{ij} = \left[ \frac{\partial}{\partial t} + U_k \frac{\partial}{\partial x_k} \right] (\underline{A}_n)_{ij} + (\underline{A}_n)_{ik} \frac{\partial U_k}{\partial x_j} + (\underline{A}_n)_{jl} \frac{\partial U_l}{\partial x_i}. \quad (2.11)$$

We think that the expansions for small strains and for slow motions are not distinct because (2.7,8,9,10) arise as a special case of (2.3,4,5,6) when the motion is slow. By a slow motion we mean a velocity field of the form

$$\underline{U}(\underline{x}, t, \epsilon) = \epsilon \underline{u}(\underline{x}, t'), \quad t' = \epsilon t \quad (2.12)$$

where  $t'$  derivatives of  $\underline{u}(\underline{x}, t')$  of all orders are bounded fields. The  $n$ th acceleration

$$\frac{d^n \underline{U}(\underline{x}, t, \epsilon)}{dt^n} = \left( \frac{\partial}{\partial t} + \underline{U} \cdot \nabla \right)^n \underline{U} = \epsilon^{n+1} \left( \frac{\partial}{\partial t'} + \underline{u} \cdot \nabla \right)^n \underline{u}$$

tends to zero as  $\epsilon^{n+1}$  as  $\epsilon \rightarrow 0$ . Slow unsteady motions and slow steady motions share the property that their velocity is given by  $\epsilon$  times a bounded field and their  $n$ th acceleration by  $\epsilon^{n+1}$  times a bounded field.

To obtain (2.7,8,9,10) from (2.6) when the motion is slow we note that since  $\underline{A}_n[\underline{U}(\underline{x}, t, \epsilon)]$  satisfies (2.11)

$$\underline{A}_n[\underline{U}(\underline{x}, t, \epsilon)] = \epsilon^n \underline{A}_n[\underline{u}(\underline{x}, t')] \quad (2.13)$$

where  $\underline{A}_n[\underline{u}(\underline{x}, t')]$  satisfies

$$\underline{A}_{n+1}[\underline{u}] = \epsilon \left[ \frac{\partial \underline{A}_n}{\partial t'} + \underline{u} \cdot \nabla \underline{A}_n + \underline{A}_n \cdot \nabla \underline{u} + (\underline{A}_n \cdot \nabla \underline{u})^T \right],$$

$$\begin{aligned} \underline{G}(s, \epsilon) &= \sum_{n=1}^{\infty} (-1)^n \frac{s^n}{n!} \underline{A}_n[\underline{U}(\underline{x}, t, \epsilon)] \\ &= \sum_{n=1}^{\infty} (-1)^n \frac{n(\epsilon s)^{n-1}}{n!} \underline{A}_n[\underline{u}(\underline{x}, t')] \end{aligned} \quad (2.14)$$

Substituting (2.14) into (2.6) we find that (2.7) arises as the coefficient of  $\epsilon$ , (2.8) as the coefficient of  $\epsilon^2$ , (2.9) as the coefficient of  $\epsilon^3$ , and (2.10) as the coefficient of  $\epsilon^4$ , where the constants arise as moments of the material functions<sup>1</sup>. For example (see Coleman & Markovitz (5)),

$$\nu = - \int_0^{\infty} s \zeta(s) ds, \quad \alpha_1 = \frac{1}{2} \int_0^{\infty} s^2 \zeta(s) ds$$

and

$$\alpha_2 = \int_0^{\infty} \int_0^{\infty} \beta(s_1, s_2) s_1 s_2 ds_1 ds_2.$$

### 3. CANONICAL FORMS OF THE STRESS FOR THE PERTURBATION OF THE REST STATE<sup>2</sup>

We turn now to the problem of free surfaces on viscoelastic fluids in arbitrary time-dependent motions which perturb the rest state. We consider a one-dimensional family of problems depending on an externally given parameter  $\epsilon$ . When  $\epsilon = 0$ , the fluid is at rest. For example, in the problem of the free surface on top of viscoelastic fluid induced by rotation of a rod the angular velocity of the rod is given by  $\epsilon f(t)$  where  $f(t)$  is a prescribed

<sup>1</sup> This reduction was carried out for the fluid of second grade by Coleman and Markovitz (5) and for the fluid of third grade in steady flow by Schowalter (6). To make the reduction at third, fourth and higher orders it is convenient to use trace identities for the Rivlin-Ericksen tensors evaluated on isochoric flows (see Spencer & Rivlin (7)).

<sup>2</sup> The material in this section gives a simplification, due to Joseph, of the theory of domain perturbations of the rest state with unsteady motion (Joseph (4), Chap. XIII). In the old theory we used the material mapping,  $\underline{X} \mapsto \underline{x}_\epsilon$ , defined by  $\underline{\xi}(\underline{X}, t, \epsilon) = \underline{x}_\epsilon$ ,  $\underline{\xi}(\underline{X}, t, 0) = \underline{X}$ , as a domain mapping. In the present, improved theory we do not introduce  $\underline{\xi}(\underline{X}, t, \epsilon)$  but work exclusively with the relative position vector  $\underline{x}_t(\underline{X}, t, \epsilon)$ . We are then able to define simpler mappings of the domain, like (3.16), which are not material mappings and are just like the domain mappings we use to study steady flow.

function of the time  $t$ . The coordinates of the rest state may be identified with the particle labels  $\underline{x}$ . The domain  $\mathcal{V}_\epsilon(t)$  occupied by the fluid depends on  $t, \epsilon$  but  $\mathcal{V}_0$  is a fixed domain independent of  $t$ .

We begin by introducing kinematics for perturbations of the rest state. The relative position vector is given by

$$\underline{x}_t(\underline{x}, \tau, \epsilon) = \underline{x} + \sum_1 \underline{x}_t^{<n>}(\underline{x}, \tau) \frac{\epsilon^n}{n!} \quad (3.1)$$

When  $\epsilon = 0$

$$\underline{x}_t(\underline{x}, \tau, 0) = \underline{x} \quad (3.2)$$

is independent of  $\tau$  because the fluid is at rest. Moreover, since

$$\underline{x}_t(\underline{x}, t, \epsilon) = \underline{x} \quad (3.3)$$

it follows that

$$\underline{x}_t^{<n>}(\underline{x}, t) = 0, \quad n \geq 1. \quad (3.4)$$

The velocity of the particle at time  $\tau$  which at time  $t$  is at place  $\underline{x}$  (the particle at  $\underline{x}$  when  $\epsilon = 0$ ) is given by

$$\begin{aligned} \underline{U}(\underline{x}_t(\underline{x}, \tau, \epsilon), \tau, \epsilon) &\equiv \tilde{\underline{U}}(\underline{x}, \tau, \epsilon) = \frac{\partial \underline{x}_t(\underline{x}, \tau, \epsilon)}{\partial \tau} \\ &= \sum_1 \tilde{\underline{U}}^{<n>}(\underline{x}, \tau) \frac{\epsilon^n}{n!} = \sum_1 \frac{\epsilon^n}{n!} \frac{\partial \underline{x}_t^{<n>}(\underline{x}, \tau)}{\partial \tau}. \end{aligned} \quad (3.5)$$

Hence

$$\underline{x}_t^{<n>}(\underline{x}, \tau) = \int_{\tau}^t \tilde{\underline{U}}^{<n>}(\underline{x}, \tau') dt', \quad (3.6)$$

where, by differentiating (3.5)<sub>1</sub> with respect to  $\epsilon$  at  $\epsilon=0$ , we find that

$$\tilde{\underline{U}}^{<1>}(\underline{x}, \tau) = \underline{U}^{<1>}(\underline{x}, \tau)$$

$$\tilde{\underline{U}}^{<2>}(\underline{x}, \tau) = \underline{U}^{<2>}(\underline{x}, \tau) + 2 \underline{x}_t^{<1>} \cdot \nabla \underline{U}^{<1>}(\underline{x}, \tau)$$

etc. We note that since

$$\nabla_{\underline{x}} \cdot \underline{U}(\underline{x}, \tau, \epsilon) = 0, \quad \underline{x} = \underline{x}_t(\underline{x}, \tau, \epsilon)$$

is an identity in  $\mathcal{V}$

$$\nabla_{\underline{x}} \cdot \underline{U}^{<n>}(\underline{x}, \tau) = 0$$

in  $\mathcal{V}$  and, taking  $\epsilon=0$ ,  $\underline{x} = \underline{x}$  we have

$$\nabla \cdot \underline{U}^{<n>}(\underline{x}, \tau) = 0$$

for  $n \geq 0$ .

With these kinematic preliminaries aside we turn next to the stresses. We confine our attention to the second order theory and seek to purge  $\mathcal{F}^{(2)}$  of redundant terms. We first rewrite (2.4)

$$\begin{aligned} \mathcal{F}^{(2)} &= \int_0^\infty G(s) \underline{\mathbb{J}}(t-s, \epsilon) ds \\ &+ \int_0^\infty \int_0^\infty \gamma(s_1, s_2) \underline{\mathbb{J}}(t-s_1, \epsilon) \cdot \underline{\mathbb{J}}(t-s_2, \epsilon) ds_1 ds_2 + O(\epsilon^3) \end{aligned} \quad (3)$$

where

$$\underline{\mathbb{J}}(t-s, \epsilon) = \underline{F}_t^T(t-s, \epsilon) \cdot \underline{A}_1[\underline{U}(\underline{x}, t-s, \epsilon)] \cdot \underline{F}_t(t-s, \epsilon), \quad (3)$$

$$(\underline{\mathbb{J}})_{ij} = \frac{\partial \xi_k}{\partial x_i} \left[ \frac{\partial U_m}{\partial \xi_k} + \frac{\partial U_k}{\partial \xi_m} \right] \frac{\partial \xi_m}{\partial x_j} = \frac{\partial U_m}{\partial x_i} \frac{\partial \xi_m}{\partial x_j} + \frac{\partial \xi_k}{\partial x_i} \frac{\partial U_k}{\partial x_j}.$$

The material constant  $\alpha(s_1, s_2)$  does not appear in the second order theory because  $G(s, \epsilon) = O(\epsilon^2)$ ,  $\text{tr } G(s, 0) = 0$  (for incompressible materials) and  $\text{tr } G(s, \epsilon) = O(\epsilon^2)$ . The material functions  $G(s)$  and  $\gamma(s_1, s_2)$  are, respectively, the linear and quadratic shear relaxation moduli,

$$\frac{dG}{ds} = \tau(s), \quad \gamma(s_1, s_2) = \frac{\partial^2 \beta(s_1, s_2)}{\partial s_1 \partial s_2}.$$

$G(s)$  and  $\gamma(s_1, s_2)$  vanish with sufficient rapidity to justify the integration by parts leading to (3.7). These moduli are related to the constants of the fluid of grade two by

$$\begin{aligned} \mu &= \int_0^\infty G(s) ds, \\ \alpha_1 &= - \int_0^\infty s G(s) ds, \\ \alpha_2 &= \int_0^\infty \int_0^\infty \gamma(s_1, s_2) ds_1 ds_2. \end{aligned} \quad (3)$$

The canonical forms of the stress for perturbations of the rest state arise from the perturbation of the equations of motion

$$F(\underline{x}, \epsilon) = \rho \left( \frac{\partial \underline{U}}{\partial t} + \underline{U} \cdot \nabla \underline{U} \right) + \nabla p - \nabla \cdot \underline{\mathcal{G}}[\underline{G}(s)] = 0. \quad (3.10)$$

Since (3.10) is an identity in  $\mathcal{V}$ ,  $F^{<n>}(\underline{x}) = 0$  in  $\mathcal{V}$  for all  $n \geq 0$ . To expand  $\underline{\mathcal{G}}$ , at constant  $\underline{x}$  we write

$$(\underline{\mathcal{U}})_{ij} = \frac{\partial \underline{U}}{\partial x_i} \cdot \frac{\partial \chi_t}{\partial x_j}(\underline{x}, \tau, \epsilon) + \frac{\partial \underline{U}}{\partial x_j} \cdot \frac{\partial \chi_t}{\partial x_i}(\underline{x}, \tau, \epsilon) \quad (3.11)$$

and

$$\begin{aligned} \frac{\partial \underline{U}}{\partial x_i} \cdot \frac{\partial \chi_t}{\partial x_j}(\underline{x}, \tau, \epsilon) &= \sum_{\ell=1}^{\infty} \frac{\epsilon^\ell}{\ell!} \frac{\partial \underline{U}^{<\ell>}}{\partial x_i} \\ &+ \sum_{\ell=1}^{\infty} \sum_{m=1}^{\infty} \frac{\epsilon^{m+\ell}}{m! \ell!} \frac{\partial \underline{U}^{<\ell>}}{\partial x_i} \cdot \frac{\partial \chi_t^{<m>}}{\partial x_j} \end{aligned}$$

Collecting results we find that in  $\mathcal{V}$ ,

$$\rho \frac{\partial \underline{U}^{<1>}}{\partial t}(\underline{x}, t) + \nabla p^{<1>} - \int_0^\infty G(s) \nabla^2 \underline{U}^{<1>}(\underline{x}, t-s) ds = 0 \quad (3.12)$$

and

$$\begin{aligned} \rho \left[ \frac{\partial \underline{U}^{<2>}}{\partial t} + 2 \underline{U}^{<1>} \cdot \nabla \underline{U}^{<1>} \right] + \nabla p^{<2>} - \int_0^\infty G(s) \nabla^2 \underline{U}^{<2>}(\underline{x}, t-s) ds \\ - 2 \nabla \cdot \int_0^\infty G(s) \left\{ \chi_t^{<1>} \cdot \nabla \underline{A}_1(s) + \underline{A}_1(s) \cdot \nabla \chi_t^{<1>} + \nabla \chi_t^{<1>} \cdot \underline{A}_1(s) \right\} ds \\ - 2 \nabla \cdot \int_0^\infty \int_0^\infty \gamma(s_1, s_2) \underline{A}(s_1) \cdot \underline{A}(s_2) ds_1 ds_2 = 0 \end{aligned} \quad (3.13)$$

where  $\chi_t^{<1>} = \chi_t^{<1>}(\underline{x}, t-s)$ ,  $\underline{A}_1(s) = \underline{A}_1[\underline{U}^{<1>}(\underline{x}, t-s)]$  and  $\nabla \cdot \underline{U}^{<1>} = \nabla \cdot \underline{U}^{<2>} = 0$ . Equations (3.12) and (3.13) are to be solved subject to boundary conditions arising from the perturbation of given  $\chi_t^{<1>}$  data. When this data is steady we find that  $\chi_t^{<1>}(\underline{x}, t-s) = -s \underline{U}^{<1>}(\underline{x})$  and

$$\nabla p^{<1>} - \mu \nabla^2 \underline{U}^{<1>}(\underline{x}) = 0 \quad (3.14)$$

and

$$\begin{aligned} 2\rho \underline{U}^{<1>} \cdot \nabla \underline{U}^{<1>} + \nabla p^{<2>} - \mu \nabla^2 \underline{U}^{<2>} \\ - 2 \nabla \cdot (\alpha_1 \underline{A}_2 + \alpha_2 \underline{A}_1^2) = 0. \end{aligned} \quad (3.15)$$

The perturbation problems may be solved sequentially. First we find  $\underline{U}^{<1>}$  from (3.12). Then we compute  $\chi_t^{<1>}(\underline{x}, \tau)$  from (3.6). Then we compute  $\underline{U}^{<2>}$  from (3.13). We may then compute  $\chi_t^{<2>}(\underline{x}, \tau)$  from (3.6) and proceed to higher orders. Recent work of Slemrod (8) guarantees that, for the type of boundary conditions occurring in the applications, these perturbation problems are uniquely solvable.

In treating free surface problems which perturb the rest state with unsteady motions we confront two problems. We need an iterative procedure for computing the free surface like that described in Section 1 which will allow us to simultaneously generate the history of the deformation tensor  $\underline{F}(\tau, \epsilon)$  which is necessary to compute the stress. An iterative procedure of this kind has been given by Joseph (4) and applied to the problem of the free surface on the top of a fluid between cylinders undergoing torsional oscillations (Joseph and Beavers, (9)). In discussing this procedure it is best to separate the problem of computing histories from the domain perturbation. We therefore start our discussion by considering problems in which the region occupied by the fluid is constant. As in the model problem of Section 1 this assumption restricts the preliminary analysis to a perturbation of boundary data at a fixed boundary, independent of  $\epsilon$  and  $t$ .

We now suppose that the domain occupied by the fluid depends on  $\epsilon$  as in the rod climbing problem shown in Fig. 3.1. The analysis of this difficult problem closely follows along the lines of

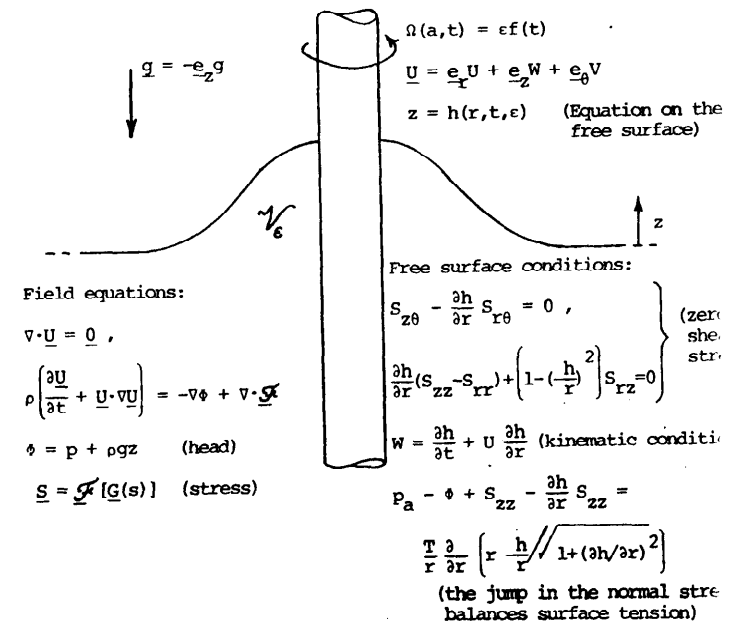


Fig. 3.1 Governing equations for unsteady rod climbing. The constants  $[\rho, g, p_a, T]$  = [density, gravity, atmospheric pressure, surface tension].



the discussion of the model problem analyzed in Section 1. We define a good mapping  $\mathcal{V}_\epsilon(t) \leftrightarrow \mathcal{V}_0$

$$\mathcal{V}_\epsilon(t) = [r, \theta, z: r \geq a, 0 \leq \theta \leq 2\pi, z \leq h(r, t, \epsilon)] ,$$

$$\mathcal{V}_0 = [r_0, \theta_0, z_0: r_0 \geq a, 0 \leq \theta_0 \leq 2\pi, z_0 \leq 0]$$

with a shifting transformation

$$\begin{aligned} \theta &= \theta_0, \\ r &= r_0, \\ z &= z_0 + h(r_0, t, \epsilon), \end{aligned} \quad (3.16)$$

The differentiation rules discussed in Section 1 apply here with obvious modifications introduced by the fact that in the present problem the shift is along vertical rather than radial lines ( $\underline{x}^{[n]} = \underline{e}_z h^{[n]}(r_0, t)$ ). The solution in  $\mathcal{V}_\epsilon(t)$  is obtained as a series in  $\mathcal{V}_0$  whose coefficients are substantial derivatives following the mapping

$$\begin{pmatrix} \underline{u}(r, z, t, \epsilon) \\ \chi_t(r, z, t, \epsilon) \\ \phi(r, z, t, \epsilon) \\ h(r, t, \epsilon) \end{pmatrix} = \sum_{n=1}^{\infty} \frac{\epsilon^n}{n!} \begin{pmatrix} \underline{u}^{[n]}(r_0, z_0, t) \\ \chi_t^{[n]}(r_0, z_0, t) \\ \phi^{[n]}(r_0, z_0, t) \\ h^{[n]}(r_0, z_0, t) \end{pmatrix} .$$

The substantial derivatives may be written in terms of partial derivatives holding  $\underline{x}$  fixed through connection formulas like (1.9). For example, since  $(\cdot)^{[0]} = (\cdot)^{<0>} = 0$

$$\begin{aligned} (\cdot)^{[2]} &= (\cdot)^{<2>} + 2\underline{x}^{[1]} \cdot \nabla (\cdot)^{<1>} \\ &= (\cdot)^{<2>} + 2h^{[1]} \frac{\partial}{\partial z_0} (\cdot)^{<1>} . \end{aligned}$$

The equations which govern the partial derivatives in  $\mathcal{V}_0$  are (3.12) and (3.13) with  $\underline{x} = \underline{x}_0$  and  $p = \phi$  (the head). At the rod surface,  $r_0 = a$

$$\underline{u}^{<1>} = \underline{e}_\theta a f(t), \quad \underline{u}^{<n>} = 0 \quad (n > 1) .$$

The equations which hold on the free surface are expressed on  $\partial\mathcal{V}_0$  in terms of partial derivatives through the connection formulas as in Section 1.

This completes our derivation of the theory of free surfaces induced by time-dependent motions of a viscoelastic fluid which perturb the rest state. We turn next to applications and results.

#### 4. STEADY ROD CLIMBING<sup>3</sup>

Here we consider the well-known fact that certain viscoelastic fluids will climb rotating rods of small diameter. The mathematical problem is that shown in Fig. 3.1 with  $f(t) = 1$  and the analysis follows along lines laid out in Section 3.

##### 4.1 Symmetry

From symmetry considerations following a change in the sign of  $\epsilon$  it is easy to establish that the azimuthal velocity and shear stresses are odd functions of  $\epsilon$ , while the secondary motions, head and free surface are even functions of  $\epsilon$ .

##### 4.2 Physical Description

There is a neat sorting of the different characteristic physical effects into an association with the different powers of  $\epsilon$  in the series solution. When there is no rotation, the free surface is flat and the pressure is hydrostatic. At first order in  $\epsilon$ , there is a z-independent flow in circles with no change in the pressure or flat free surface. At order two, the pressure must equilibrate the central forces arising from centripetal accelerations and normal stress. The free surface acts as the barometer of the interior pressure distribution, rising where the interior pressure is greatest. The free surface can remain flat only if there is no motion. The departure from flatness of the free surface at order two requires that the azimuthal velocity, at order three, should come to depend on z. This is a consequence of the fact that the azimuthal component of the shear stress  $\underline{S} = \mathcal{F}[\underline{G}(s)]$

$$S_{n\theta} = S_{z\theta} - h'S_{r\theta} ,$$

which vanishes automatically for z-independent fields when the free surface is flat, can vanish when the free surface is not flat only when  $S_{z\theta} = h'S_{r\theta}$  does not vanish. The z dependence of the azimuthal field at third order is generated without changing the pressure or the shape of the free surface. The z-dependent azimuthal field, generated at order three, is associated at order four with forces that also depend on z; such forces inevitably exert torques in an azimuthal plane, and they lead to secondary motions.

##### 4.3 Steady Rod Climbing at Second Order

If we assume that the free surface at the rod surface makes a flat angle of contact it is not hard to show that

$$\underline{u}^{<1>} = \underline{e}_\theta a^2/r, \quad \underline{u}^{<2>} = 0 \quad \text{in } \mathcal{V}_0 ,$$

and

$$\frac{1}{2}\phi^{<2>} = \frac{2a^4}{r^4} \hat{\beta} - \frac{\rho a^4}{2r^2} \quad (4.1)$$

where  $\hat{\beta} = 3\alpha_1 + 2\alpha_2$  is the climbing constant. The coefficient  $h^{[2]}(x)$ , which gives the first deviation of the free surface from flatness,

<sup>3</sup> Joseph and Fosdick (10); Joseph, Beavers and Fosdick (11); Beavers and Joseph (12), (15).

$$h(r, \epsilon) = h^{[2]}(r) \epsilon^2 + O(\epsilon^4), \quad (4.2)$$

is determined from the problem

$$\frac{T}{r} (rh^{[2]})' - \rho g h^{[2]} = -\frac{2a^4}{r^4} \hat{\beta} + \frac{\rho a^4}{2r^2}, \quad (4.3)$$

$$h^{[2]}(a) = 0, \quad h^{[2]}(r) \rightarrow 0 \quad \text{as } r \rightarrow \infty.$$

#### 4.4 The Critical Radius and the Effects of Surface Tension

The fluid will climb the rod whenever  $\epsilon$  is small and wherever  $h^{[2]}(r) > 0$ . To see this, it is convenient to set  $T = 0$ . Then the free surface will rise if and only if  $\hat{\beta} > 0$  and

$$r^2 < 4\hat{\beta}/\rho. \quad (4.4)$$

Otherwise, inertia dominates, and the free surface sinks. Representative experimental values for  $\hat{\beta}$  near room temperatures are  $\hat{\beta} \approx 1$  for STP, and  $\hat{\beta} \approx 0.8$  and  $1.4$  for polyacrylamide, (Beavers and Joseph (12)). It is easy to verify that the ratio

$$|T(rh^{[2]})'| / \rho g r h^{[2]} \rightarrow 16T/\rho g r^2$$

as  $r \rightarrow 0$  when  $h^{[2]}$  is given (4.3) with  $T = 0$ . This ratio shows that when  $r$  is small surface tension should not be neglected (see Fig. 4.1). To determine the way in which the parameters  $a, \beta, \rho, g, T$  affect  $h^{[2]}$ , Joseph, Beavers and Fosdick (11) derived an accurate approximation to the problem (4.3). In the approximation, we write (4.3) as

$$T(rh^{[2]})' - \rho g a^2 h^{[2]} = -a^2 \phi^{<2>} + (r - a^2/r) T(rh^{[2]})', \quad (4.5)$$

The last term of (4.5) is zero when  $r = a$ . We set this term to

<sup>4</sup>Equation (4.4) gives a critical radius

$$r_c = 2\sqrt{\hat{\beta}/\rho} \quad (4.7)$$

corresponding to  $h^{[2]}(r_c) = 0$  in the absence of surface tension. For  $r < r_c$  normal stresses dominate and for  $r > r_c$  inertia dominates. The first theoretical analyses of rod climbing are due to Serrin (13) and Giesekus (14). Serrin studied the problem for a Reiner-Rivlin fluid with constant coefficients (for this mathematical fluid,  $\hat{\beta} = 2\alpha_2$ ), on the assumption that the free surface was nearly flat and that the  $z$  dependence of the solution and the secondary motions could be neglected. Surface tension was neglected. He interpreted a negative slope at  $r = a$  as a tendency to climb and a negative slope at  $r = b$  as a tendency to fall. He deduced a critical radius corresponding to the condition that  $dh^{[2]}(r_c)/dr = 0$  when  $T = 0$ . The physically dominant feature of rod climbing is that the normal stress effects die very rapidly like  $1/r^4$  when  $r$  is large. When a viscoelastic fluid is floated on the top of a Newtonian fluid, the viscoelastic fluid will climb down into the Newtonian fluid (see Section 4.7) and though the free surface has no critical radius it has a shape which is very close to one that does.

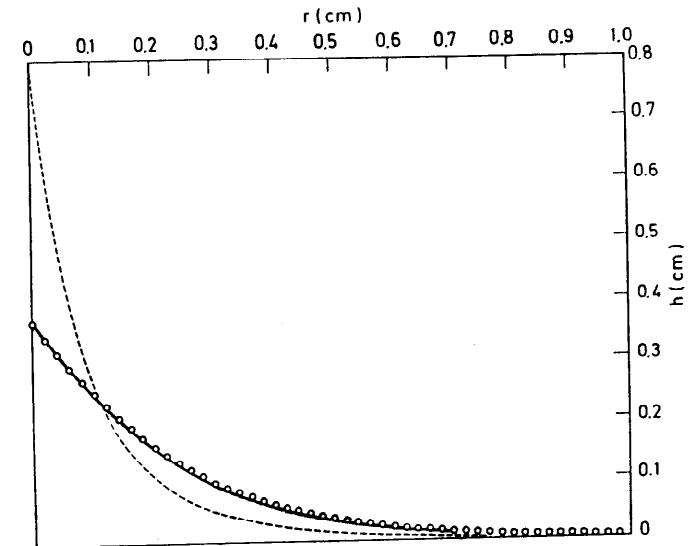


Fig. 4.1 The free surface on STP motor oil additive near a rod of radius  $a = 0.320$  cm rotating at 3.8 rev/sec, ( $\hat{\beta} = 0.63$ ): — experimental profile; - - - second order theory neglecting surface tension; o o o second order theory including surface tension (Joseph, Beavers and Fosdick (11)).

zero, and solve the remaining problem; then we restore the neglected term through successive approximations. The first approximation is very accurate, especially near the rod  $r = a$  (see Joseph, Beavers and Fosdick (11)):

$$\frac{1}{2} h^{[2]} \sim \frac{a}{2T\sqrt{s}} \left[ \frac{4\hat{\beta}}{4+\lambda} - \frac{\rho a^2}{2+\lambda} \right], \quad (4)$$

$\lambda^2 = a^2 s$  and  $s = \rho g/T$ . This expression shows that, when  $a$  is small,  $h^{[2]}$  is proportional to  $a$  and  $\hat{\beta}$ , and is inversely proportional to  $\sqrt{T}$ .

#### 4.5 Rheometrical Measurements of the Climbing Constant

We use the second-order theory to determine the values of constant  $\hat{\beta}$  from

$$h(r; \epsilon^2, \hat{\beta}, \rho, T) = \frac{1}{2} h^{[2]}(r; \hat{\beta}, \rho, T) \epsilon^2 + O(\epsilon^4). \quad (4)$$

When  $h, T$  and  $\rho$  are given, we may compute  $\hat{\beta}$  from (4.8). In Fig. 4.2 we have plotted typical examples of the height rise at  $r =$  against the square of the angular velocity  $\omega^2 = \epsilon^2/4\pi^2$ . The rise is nearly linear in  $\omega^2$  for values of  $\omega^2$  less than about 10. This suggests that there might be good agreement between the second-order theory given by (4.8) and the experimental observations.

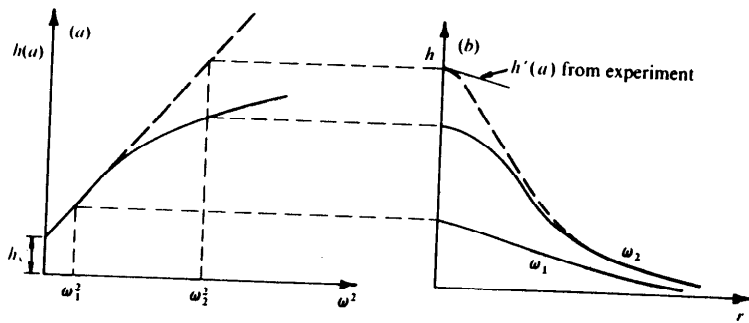


Fig. 4.2 (a) Method of slopes. (b) Method of profile fitting. ---, second order theory; —, experiment. Observed rise at  $r = a$  never seems to lie above second-order theory. Static rise in height  $h_s$  computed using observed contact angles. When the fluid does not wet the rod,  $h_s = 0$  and  $h'(a) = 0$ . In second-order theory  $\hat{\beta}$  is computed from (4.6) by method of slopes. Theoretical profiles computed numerically from first two terms of (4.9) using observed values of  $h'(a)$ , and values of  $\hat{\beta}$  taken from method of slopes. (Beavers and Joseph (12)).

Such agreement was attained in Joseph, Beavers and Fosdick (11), even though in those experiments the condition  $h'(a) = 0$  was not satisfied; neither was  $h'(a)$  small. Joseph, Beavers and Fosdick (11) attributed the rise associated with the non-zero wetting angle  $h'|_{r=a} = -\eta$  to capillarity and replaced (4.8) with

$$h(r; \epsilon^2, \hat{\beta}, \rho, T, \eta) = h_s(r; \rho, T, \eta) + \frac{1}{2} h^{[2]}(r; \hat{\beta}, \rho, T) \epsilon^2 + O(\epsilon^2 \eta + \epsilon^4). \quad (4.9)$$

$h_s$  is the static rise, computed from

$$\frac{T}{r} \left( \frac{r h'_s}{(1+h_s)^2} \right)' - \rho g h_s = 0, \quad h'|_{r=a} = -\eta, \quad h_s \rightarrow 0 \text{ as } r \rightarrow \infty. \quad (4.10)$$

The static rise vanishes when  $\eta = 0$ . The plausible procedure (4.9) for computing  $h$  seemed to work, but had an ad hoc character, since  $\eta$  was not small and there was no a priori reason to neglect terms  $O(\epsilon^2 \eta)$ . In the experiments reported by Beavers and Joseph (12) we were able to control  $\eta$ , setting it to zero.

There are two methods we use to determine  $\hat{\beta}$  from (4.9) and the measured values of  $h = h_{\text{exp}}$ : the method of slopes and the method of profile fitting. The two methods are demonstrated in Fig. 4.2. To use the method of slopes, we need only measure the height rise  $h_{\text{exp}}$  at  $r = a$ ; from these measurements, we read off a slope and equate the theoretical and measured values

$$\frac{1}{2} h^{[2]}(a; \hat{\beta}, \rho, T) = dh_{\text{exp}}/d\epsilon^2.$$

This equation is then solved for  $\hat{\beta}$ . The method of slopes does not depend on the static climb  $h_s$ , but the only justification we have for assuming that  $dh_{\text{exp}}/d\epsilon^2$  is independent of  $\eta$  is experimental.

In the method of profile fitting, we choose the value of  $\hat{\beta}$  that gives the best fit over the entire profile. This method is judged successful if, at a given temperature, one and the same value of  $\hat{\beta}$  is determined for a single fluid from tests with rods of different radius rotating at different values of  $\epsilon$ . The method

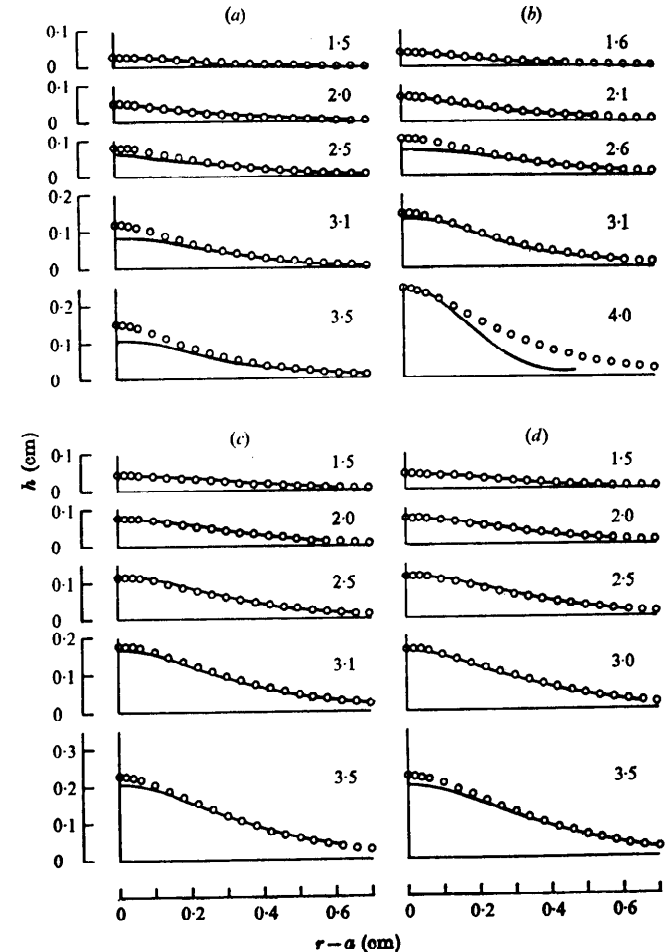


Fig. 4.3. Profile fitting for polyacrylamide solution. Comparison of observed with predicted profiles for low rotational speed. Temperature = 24°C.

	(a)	(b)	(c)	(d)
a (cm)	0.317	0.476	0.635	0.794

(Beavers and Joseph (12))

of profile fitting requires that one compute the static rise  $h_s$  from the experimentally measured values of  $\eta$ . The method of profile fitting is more accurate but more time-consuming than the method of slopes. In general, the determination of  $\hat{\beta}$  is achieved most efficiently and accurately by simultaneous use of both methods. Some representative comparisons between theory and experiment are exhibited in Fig. 4.3.

#### 4.6 The Effect of Temperature

Beavers and Joseph (12) measured  $\hat{\beta}$  at temperatures in the range 25 to 50°C. The variation of  $\hat{\beta}$  with temperature for STP is shown in Fig. 4.4. Obviously, temperature has a very big effect.

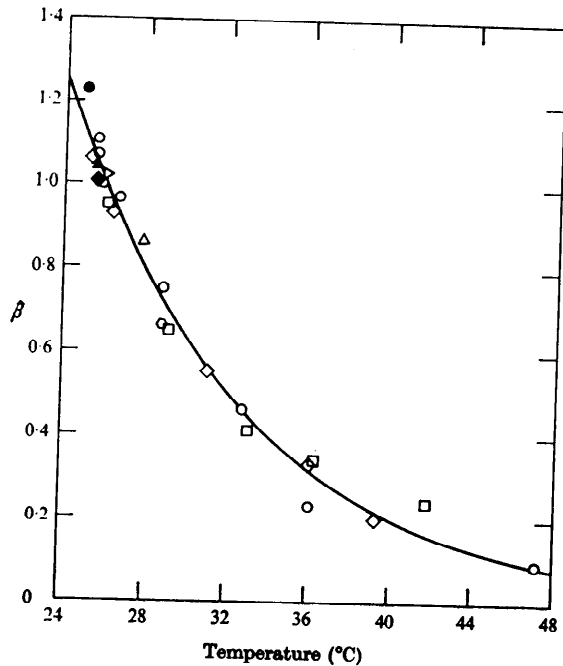


Fig. 4.4. Variation of  $\hat{\beta}$  ( $\text{g cm}^{-1}$ ) with temperature ( $^{\circ}\text{C}$ ), for STP. —, empirical relation  $\hat{\beta} = 20 \exp(-0.115T)$ ,  $25 < T < 50^{\circ}\text{C}$ .

Coated	Uncoated					
		◻	●	◆	▲	◻
a(cm)		0.159	0.317	0.476	0.635	0.794
			○	◊	△	□
						○

(Beavers and Joseph (12)).

#### 4.7 Normal Stress Amplifier

The perturbation analysis of rod climbing can be carried through second order without complication when a layer of liquid is floated on another immiscible liquid. Beavers and Joseph (15) have given an analysis and reported an experiment testing the analysis for the problem in which a non-Newtonian liquid (STP) is floated on water. The STP climbs up the rotating rod into the air and down the rod into the water. The down-climb is much larger than the up-climb, roughly in the ratio

$$\frac{h_B^{[2]}(a)}{h_T^{[2]}(a)} \sim \frac{\sqrt{T_T(\rho - \rho_a)}}{\sqrt{T_B(\rho_w - \rho)}} \frac{4 + a\sqrt{(\rho - \rho_a)g/T_T}}{4 + a\sqrt{(\rho_w - \rho)g/T_B}} \quad (4.11)$$

where the quantity  $h_B^{[2]}(a)$  is the down-climb at the bottom STP-water interface on the rod surface at  $r = a$ ,  $h_T^{[2]}(a)$  is the up-climb at the air-STP interface, and  $\rho$ ,  $\rho_w$  and  $\rho_a$  are densities of STP, water and air respectively, and  $T_T$  and  $T_B$  are surface tension coefficients (see Fig. 4.5). Equation (4.11) is derived from an

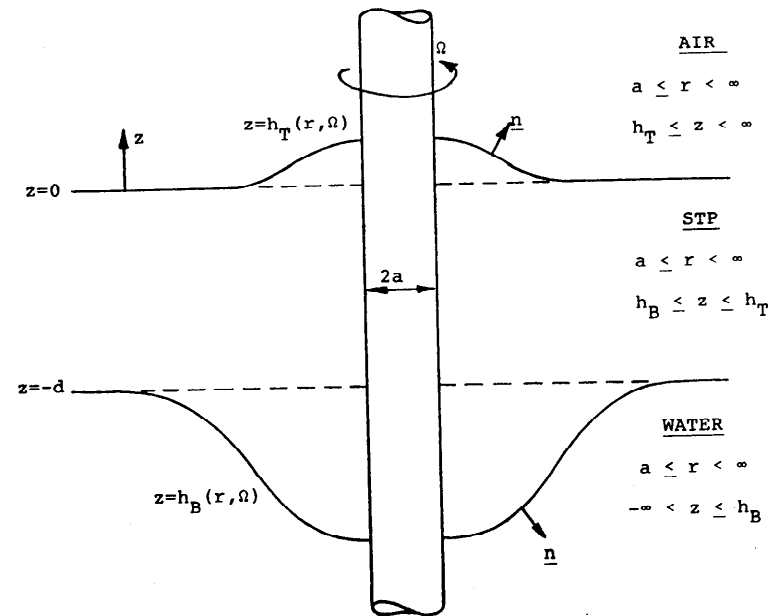


Fig. 4.5. Schematic sketch of the free surfaces on STP floating on water.

analysis of the type that leads to (4.6) except that the dynamical problem is solved in all three layers. Correct coupling conditions for the continuity of stresses at each of the two interfaces are prescribed, and it is not assumed that the interface is free of tangential tractions.

The analysis shows that the effect of inertia and gravity is to push the heavy fluid down near the rod. The normal stresses push the STP up into the air and down into the water. Hence, inertia and normal stresses are in conflict at the STP-air interface and in concert at the water-STP interface.

In the experiments reported by Beavers and Joseph (15) the values of the ratio (4.11) are, respectively, (3.7, 3.9, 4.3) for three different rods of radius  $a = (0.479, 0.627, 0.951 \text{ cm})$ . The down-climb/up-climb ratio (4.11) may be further approximated by  $h_B^{[2]}(a)/h_T^{[2]}(a) \sim [(\rho - \rho_a)/(\rho_w - \rho)]^{1/2} \sim 2.7$ . The value of  $\hat{\beta}$  for STP was computed from the observed up-climb and the observed down-climb. There is presumably one and only one value of  $\hat{\beta}$  for STP at a given temperature. The value of  $\hat{\beta}$  computed from the up-climb, from the down-climb and from the independent measurements of Beavers and Joseph (12) are in good agreement with one another.

The configuration shown in Fig. 4.5 can be regarded as a normal stress amplifier because the down-climb can be arbitrarily amplified by making the density difference at the lower interface very small. A similar analysis has been constructed when STP is floated on a polyacrylamide solution instead of water. Experiments of A. Siginer, completed but not yet reported, show good agreement between theory and experiment.

#### 4.8 Limits of Applicability of the Second Order Theory

Joseph, Beavers & Fosdick (11) developed an *ad hoc* criterion to identify the region of validity of the second order theory. They argued that the rise in height could be given as the leading term of a power series in the Froude number  $F = \epsilon^2 \ell / g$ ,

$$h(r, \epsilon) = \frac{1}{2} \frac{g}{\ell} h^{[2]}(r) F + O(F^2) \quad (4.12)$$

Here  $\ell$  is a characteristic length depending on the radius of the rod and the fluid. This length is not uniquely given and Joseph, Beavers and Fosdick guessed, using (4.5), that

$$\ell^2 = 0[gh^{[2]}] \quad \text{and} \quad gh^{[2]} \sim \hat{\beta} a \left(\frac{g}{\rho T}\right)^{1/2}$$

The criterion for the second order theory was that the Froude number  $F$  should be less than one

$$F = \frac{\epsilon^2 \ell}{g} = \frac{\epsilon^2}{g} \left[ \hat{\beta} a \left(\frac{g}{\rho T}\right)^{1/2} \right]^{1/2} < 1 \quad (4.13)$$

This criterion works remarkably well, though it is based on little more than a guess.

The problem of error estimates for perturbation solutions is very complicated. The problem is that even for steady flow, the higher order terms depend on many independent constants whose values and sign are unknown.

The higher order theory of rod-climbing through fourth order has recently been completed by J. Yoo (16) and will be reported in a Ph.D dissertation presently under preparation. Yoo's work is very extensive because he has solved a very difficult problem with many parameters. Yoo's analysis treats the problem when the fluid is between rotating cylinders. He derives a new biorthogonal eigenfunction expansion theory for Stokes flow between cylinders to solve his problem.

The equations which govern at third and fourth order, when surface tension is neglected, was worked out by Joseph and Fosdick (10). At second order the free surface changes shape without generating secondary motions. The azimuthal component of the shear traction on the free surface cannot remain zero on a pure radial azimuthal velocity field when  $dh/dr \neq 0$ . Hence the azimuthal velocity field at third order varies with depth. This vertical stratification of velocity induces torques in azimuthal planes at fourth order leading to secondary motions. The motion at fourth order is governed by the stream function  $\psi^{<4>}$  satisfy (Joseph and Fosdick, (10))

$$\mu \mathcal{L}^2 \psi^{<4>} + 8 \frac{\partial}{\partial z_0} \left\{ \frac{4}{r^2} B (\alpha_1 + \alpha_2) \left( \frac{\partial v^{<3>}}{\partial r} - \frac{v^{<3>}}{r} \right) + \rho v^{<1>} v^{<3>} \right\} = 0 \quad (4)$$

in  $\mathcal{V}_0$ , where  $\mathcal{L}^2 = \frac{\partial^2}{\partial r^2} + \frac{\partial^2}{\partial z_0^2} - \frac{1}{r} \frac{\partial}{\partial r}$

$$\text{and } \psi^{<4>} = \frac{\partial \psi^{<4>}}{\partial r} = 0 \quad \text{at } r = a, b \quad (4)$$

$$\text{and } \frac{\partial \psi^{<4>}}{\partial r} = \frac{\partial^2 \psi^{<4>}}{\partial r^2} - \frac{\partial^2 \psi^{<4>}}{\partial z_0^2} = 0 \quad \text{at } z = 0, -\infty \quad (4)$$

In (4.14),  $v^{<1>} = Ar + B/r$  is the azimuthal component of velocity at first order,

$$\text{where } A = \frac{b^2 \lambda - a^2}{b^2 - a^2}, \quad B = \frac{a^2 b^2 (1 - \lambda)}{b^2 - a^2}$$

and  $\lambda$  is the ratio of the angular velocity of the outer cylinder to that of the inner cylinder;  $v^{<3>}$  is the azimuthal component velocity at third order;  $\mu$  and  $\rho$  are the viscosity and density of the fluid;  $\alpha_1$  and  $\alpha_2$  are constants of the fluid of second grade. It is found that

$$v^{<3>} = v_{31}(r) + v_{32}(r, z_0, \hat{\beta}, \rho, T)$$

Hence, only  $v_{32}$  enters into (4.14).

<sup>5</sup> The results of Yoo's analysis, including the height rise at fourth order will also be published elsewhere.

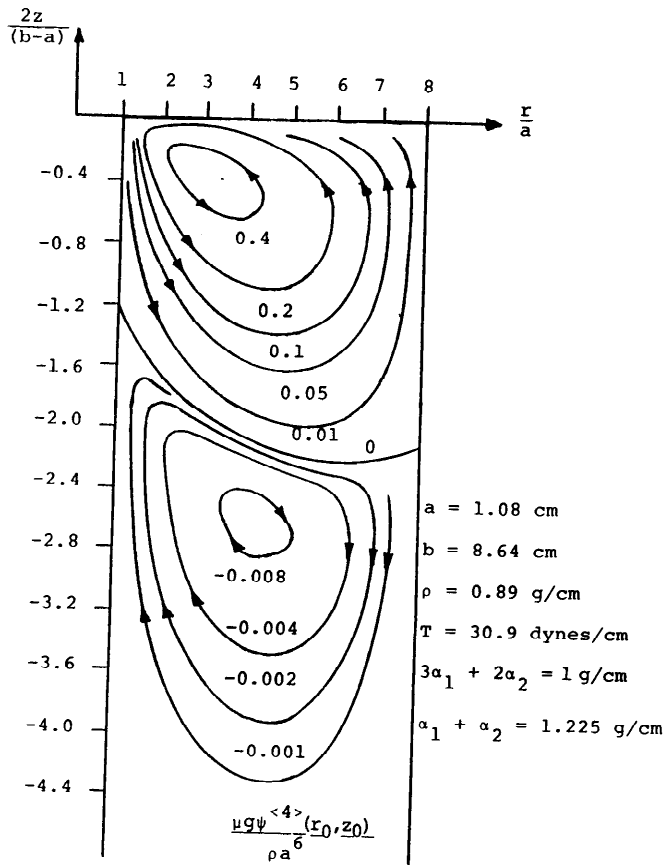


Fig. 4.6 Streamlines of the secondary motion. The level lines of  $\psi^{<4>}(r_0, z_0)$  are shown in the reference configuration. Streamlines of the low speed secondary motion can be obtained by inverting the shifting map,  $r = r_0$ ,  $z = z_0 + h(r_0, \epsilon)$ , using the low speed approximation  $h(r, \epsilon) \sim \frac{1}{2} h^{[2]}(r) \epsilon^2 + \frac{1}{4!} h^{[4]}(r) \epsilon^4$ .

The following remarkable result follows from the argument just given: The secondary motion induced by the slow rotation of a rod in any simple fluid appears first at fourth order but the viscoelastic contribution depends only on the constants  $\beta = 3\alpha_1 + 2\alpha_2$  and  $\alpha_1 + \alpha_2$  of the second order fluid. The streamlines of slow secondary motion have the same shape in all fluids having the same material properties  $(\rho, T, \beta, \alpha_1 + \alpha_2)$ .

In Fig. 4.6 we graphed the level lines of the stream function for one case which occurs in experiments. We note that the secondary motion is dominated by one edge eddy which, contrary to intuition, makes the fluid wind up on the free surface of the bubble and sink near the rod. This circulation is in agreement with the observations of Saville (17) and with our own preliminary observations at low speeds (see Fig. 4.7).

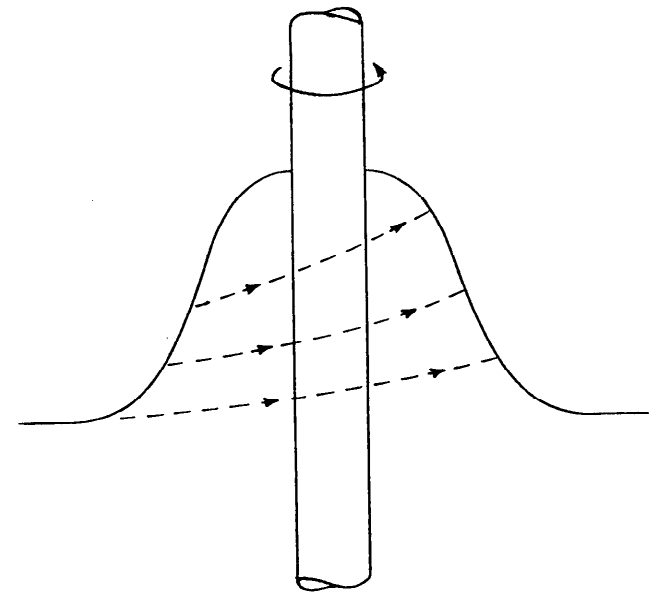


Fig. 4.7 Sketch illustrating the upward motion of a fluid particle on the free surface of the bubble.

## 5. UNSTEADY ROD CLIMBING

Everything which is reported in this section was unknown before 1974 and was discovered by us.

### 5.1 Torsional Oscillations of a Rod

Interesting things happen when a rod in a viscoelastic fluid is set into torsional oscillations. These things were discovered first by analysis (Joseph (9, Part I)) and then verified by experiments (Beavers (9, Part II)). The effects are not small in any way and they may be verified by experiments even with crude, inexpensive equipment.

The equations governing the problem of the free surface on a viscoelastic fluid outside of a rod undergoing torsional oscillations are given in Fig. 3.1 with  $f(t) = \epsilon \sin \omega t$ . The analysis is based on the perturbation theory described in Section 3. It is a small amplitude theory with amplitude

$$\epsilon = \frac{\omega\theta}{2}$$

where  $\theta$  is the angle of twist (see Fig. 5.1)

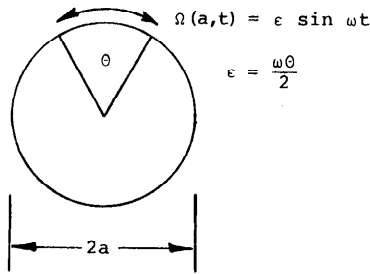


Fig. 5.1 Torsional oscillations of a rod

A moment's reflection will convince the reader that symmetry properties which follow from a change in the sign of  $\epsilon = \Omega(a)$  in the steady case also apply in the time-periodic case being considered here. Of course, one could not have an unsteady rise at second order in  $\epsilon$  without secondary motions. But this possibility exists and is realized for the mean-value of the height rise over a period of oscillation. The analysis shows that the first deviation of the free surface occurs at second order in  $\epsilon$  as a superposition of a mean climb, independent of  $t$ , and an oscillating climb which oscillates with frequency  $2\omega$ . It is very easy to compute the mean rise but hard to compute the time-periodic part. (The time periodic part of the rise of the free surface on a fluid between oscillating planes has been computed by Sturges and Joseph (18) and is discussed in Section 6.2). The experiments of Beavers (9, II) show that the mean climb completely dominates the total climb at each and every instant and at all oscillation frequencies within the operating range of the experimental equipment. This happy circumstance allows one to make fairly simple, repeatable measurements of the free surface which can be compared

with the simple theoretical expressions which give the mean rise at second order.

### 5.2 Brief Review of Theory

A summary of the theoretical expressions derived by Joseph (9, I) is listed below.

$$r\Omega(r, z, t, \epsilon) = r(\bar{\omega}(r)e^{i\omega t} + \bar{\omega}(r)e^{-i\omega t})\epsilon + O(\epsilon^3),$$

$$\psi(r, z, t, \epsilon) = \frac{1}{4}[e^{2i\omega t}\bar{\psi}_1(r, z) + e^{-2i\omega t}\bar{\psi}_1(r, z)]\epsilon^2 + O(\epsilon^4)$$

$$\phi(r, z, t, \epsilon) = \frac{1}{4}[2\bar{\phi}(r) + e^{2i\omega t}\bar{\phi}_1(r, z) + e^{-2i\omega t}\bar{\phi}_1(r, z)]\epsilon^2$$

$$h(r, t, \epsilon) = \frac{1}{4}[2\bar{h}(r) + e^{2i\omega t}\bar{h}_1(r) + e^{-2i\omega t}\bar{h}_1(r)]\epsilon^2 + O(\epsilon^4)$$

where quantities with two bars are average values over an oscillation period  $2\pi/\omega$ , single bars signify complex conjugate and  $\psi(r, z, t, \epsilon)$  is the stream function for axisymmetric flow. The function

$$v(r, t) = r(\bar{\omega}(r)e^{i\omega t} + \bar{\omega}(r)e^{-i\omega t}) \text{ satisfies}$$

$$\rho \frac{\partial v}{\partial t} = \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial v}{\partial r} \right) - \frac{1}{r^2} v \right] \int_0^\infty G(s) v(r, t-s) ds \text{ in } \mathcal{V}_0 \quad (5)$$

and

$$v(a, t) = a \sin \omega t, \quad v(\infty, t) = 0.$$

$\bar{\phi}(r)$  and  $\bar{h}(r)$  satisfy

$$\begin{aligned} \frac{1}{2} \frac{d\bar{\phi}}{dr} &= -\frac{4}{r} [r^3 |\bar{\omega}, r|^2], r \int_0^\infty G(s) \frac{\sin \omega s}{\omega} ds \\ &+ 2[r^2 |\bar{\omega}, r|^2], r \int_0^\infty \int_0^\infty \gamma(s_1, s_2) \cos \omega(s_1 - s_2) ds_1 ds_2 \\ &+ 2\rho r |\bar{\omega}|^2 \text{ in } \mathcal{V}_0 \end{aligned}$$

and

$$-\bar{\phi}(r) + \rho g \bar{h}(r) = \frac{\pi}{r} (r \bar{h}, r), r$$

$$\bar{h}, r(a) = 0, \quad \bar{h}(r) = 0.$$

It is not hard to solve (5.1) and (5.2) in terms of Bessel functions. But the properties of the solution are easier to see from a good approximate solution which leads to the following expressions:

$$v(r,t) = a \left(\frac{a}{r}\right)^{\Lambda} \sin(\omega t + \Lambda_i \log \frac{a}{r}) \quad (5.3)$$

where

$$\Lambda = \Lambda_r + i\Lambda_i = re\sqrt{1+a^2m^2} + i im \sqrt{1+a^2m^2}$$

and

$$m^2 = i\rho\omega \int_0^{\infty} G(s) e^{-i\omega s} ds$$

The height rise at  $r = a$  may be expressed in the same approximation

$$\bar{h}(a) = \frac{a}{\sqrt{\rho g T}} \left\{ \frac{|\Lambda+1|^2 \hat{\beta}_\Lambda}{(\Lambda_r+1)[2(\Lambda_r+1)+\lambda]} - \frac{\rho a^2}{2\Lambda_r(2\Lambda_r+\lambda)} \right\} \quad (5.4)$$

where  $\lambda = \rho g a^2 / T$  and

$$\hat{\beta}_\Lambda = - (2\Lambda_r+1) \int_0^{\infty} G(s) \frac{\sin \omega s}{\omega} ds + (\Lambda_r+1) \int_0^{\infty} \int_0^{\infty} \gamma(s_1, s_2) \cos \omega(s_1 - s_2) ds_1 ds_2 \quad (5.5)$$

The solutions for the steady climb which were discussed in Section 4 may be recovered from those just given in the limit  $\omega \rightarrow 0$ . In the limit  $\Lambda = \Lambda_r = 1$ , and using (3.9)

$$\hat{\beta}_1 = \hat{\beta} = 3\alpha_1 + 2\alpha_2 \quad \text{and} \quad \bar{h}(r) = \frac{1}{2} h^{[2]}(r)$$

Equation (5.3) shows that the azimuthal component of velocity is a decaying wave as might be expected in a viscoelastic fluid.

### 5.3 Experimental Results

Equation (5.4) shows that for a fixed value of  $\epsilon^2 = \omega^2 a^2 / 4$  the height rise at  $r = a$  is a monotone decreasing function of the frequency  $\omega$  having a maximum value for steady flow ( $\theta \rightarrow \infty, \omega \rightarrow 0$ ). This property predicted by analysis is in good agreement with experiments (see Fig. 5.2).

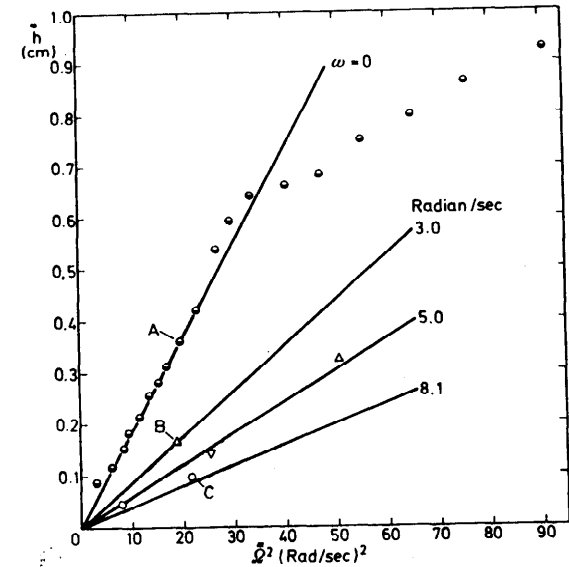


Fig. 5.2 Graph of  $\bar{h} = \frac{\epsilon^2}{2} \bar{h}(a)$ , where  $\frac{\epsilon^2}{2} = \Omega^2$ , for a rod of radii 0.636 cm. The line  $\omega = 0$  gives the height rise when the rotation is steady. (Joseph and Beavers (9)).

Equation 5.4 shows that when the amplitude  $\epsilon^2$  is small the mean climb at  $r = a$

$$\frac{\bar{h}(a, \epsilon)}{\epsilon^2} = H(a, \omega)$$

is independent of the angle of twist  $\theta$ . The universal function frequency  $H(\omega)$  is shown as a dark line in Fig. 5.3.

### 5.4 Validity of Second Order Theory

To study the limits of validity of the second order theory the unsteady case we first used the same criterion (4.13) as in steady flow except that we replace  $\hat{\beta}$  with  $\hat{\beta}_\Lambda$  and  $h^{[2]}(a)$  with  $1/2 \bar{h}(a)$ . Then the condition  $F < 1$  becomes

$$\frac{\omega^2 a^2}{8g} \left[ \hat{\beta}_\Lambda(\omega) a \left(\frac{a}{\rho T}\right)^{1/2} \right]^{1/2} < 1,$$

that is,

$$\omega^2 < \frac{8g}{a^2} \left[ \frac{1}{a \hat{\beta}_\Lambda(\omega)} \left(\frac{\rho T}{g}\right)^{1/2} \right]^{1/2}$$



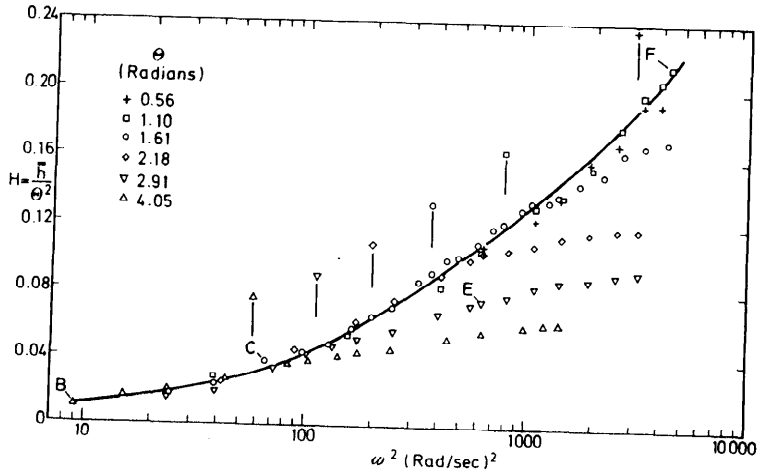


Fig. 5.3 The normalized height rise  $\bar{h}/\theta^2$  as a function of the frequency  $\omega$  of oscillation. Experimental points belonging to six different angles of twist are shown. The experimental normalized rise at second-order is shown as a solid line. The vertical bars are values  $\omega_c^2$  associated with the criterion (5.7). (Joseph and Beavers (9)).

The experiments show that  $\hat{\beta}_\Lambda(\omega)$  is a decreasing function of  $\omega$  with a maximum value  $\hat{\beta}_\Lambda(0) = 3\alpha_1 + 2\alpha_2$ . Hence the criterion

$$\omega^2 < \frac{8g}{\theta^2} \left[ \frac{1}{a(3\alpha_1 + 2\alpha_2)} \left( \frac{\rho T}{g} \right)^{1/2} \right]^{1/2} \quad (5.7)$$

is a conservative estimate of the region of validity of the second order theory. The discrepancy between the criterion (5.6) and the criterion (5.7) is an increasing function of  $\omega$ . Hence, if the criterion (5.6) is valid, then (5.7) will give increasingly conservative estimates of the  $\omega$  interval of validity of the second-order theory.

Joseph, Beavers and Fosdick(11) showed that the criterion (5.7) was consistent with their observations of the height rise of STP in the steady case. This criterion is also applicable in the experiments summarized in Fig. 5.3. In that figure the observed normalized height rise is plotted for six different angles of twist  $\theta$ , and the vertical bars indicate the corresponding values of

$$\omega_c^2 = \frac{8g}{\theta^2} \left[ \frac{1}{a(3\alpha_1 + 2\alpha_2)} \left( \frac{\rho T}{g} \right)^{1/2} \right]^{1/2} = \frac{758}{\theta^2/a}$$

given below:

$\theta$	4.05	2.91	2.18	1.61	1.10	0.56	radians
symbol	$\Delta$	$\nabla$	$\diamond$	$\circ$	$\square$	$+$	
$\omega_c^2$	58	112	200	368	786	3045	(rad/sec) <sup>2</sup> .

The experiments support the argument of the previous paragraph and lead to the conclusion that (5.7) is a conservative estimate of the region of validity of the second-order theory, which becomes more and more conservative as  $\omega$  is increased (because  $\hat{\beta}_\Lambda(\omega)$  decreases).

To relate Fig. 5.3 to the second-order theory the reader should mentally delete all of the experimental points which violate the criterion (5.7). For example, all of the points marked  $\Delta$  for which  $\omega^2 > 58$  should be deleted. When this is done the remaining points define the curve shown as a solid line in Fig. 5.3. We interpret this curve as the experimental realization of the universal function

$$H(a, \omega) \sim \omega^2 \bar{h}(a, \omega) / 8$$

whose values are given theoretically by (5.4). Returning now to Fig. 5.3 we note that many of the deleted points also lie on the curve. This feature becomes increasingly pronounced as the angle of twist is increased. We interpret this feature as a demonstration that (5.6) is a more correct and less conservative estimate than (5.7).

Our conclusion then is that, as in the steady case, the points which break away from the universal function are a manifestation of effects which are induced by the terms of order higher than  $\epsilon^2$ .

#### 5.5 Measurements of the Shear Relaxation Moduli $G(s)$ and $\gamma(s_1, s_2)$ Using the Universal Function of Frequency $H(\omega)$ .

Joseph (9,I) and Beavers (9,II) proposed a method for obtaining approximations to the functions  $G(s)$  and  $\gamma(s_1, s_2)$  from the rise curve  $H(\omega)$  of Fig. 5.3. They approximated  $G(s)$  and  $\gamma(s_1, s_2)$  with generalized Maxwell models chosen for consistency with equations (3.9):

$$G(s) \sim G_N(s) = \frac{-\mu}{\alpha_1} \sum_{n=1}^N \frac{a_n^2}{1 + b_n s} e^{-\frac{\mu}{\alpha_1} \frac{a_n}{b_n} s}$$

$$\gamma(s_1, s_2) \sim \alpha_2 \sum_{n=1}^M c_n k_n^2 e^{-k_n(s_1 + s_2)}$$

where

$$\sum_{n=1}^N a_n = \sum_{n=1}^N b_n = \sum_{n=1}^M c_n = 1.$$

There are  $2N + 2M - 3$  unknown constants if the constants  $\mu$ ,  $\alpha_1$  and  $\alpha_2$  of the fluid of second grade be known. The simplest approximation is associated with the values  $(N,M) = (1,1)$ . We found that

$$\hat{\beta}_\lambda = \frac{(2\lambda_r + 1)\alpha_1}{1 + \left(\frac{\alpha_1 \omega}{\mu}\right)^2} + \frac{(\lambda_r + 1)\alpha_2}{1 + \left(\frac{\omega}{k_1}\right)^2}. \quad (5.8)$$

In the simplest approximation the response of the fluid is determined when the values of four constants,  $\mu$ ,  $\alpha_1$ ,  $\alpha_2$  and  $k_1$ , are known. Three of these constants,  $\mu$ ,  $\alpha_1$  and  $\alpha_2$ , may be regarded as known from experiments with slow steady flow. The constant  $k_1$  may be determined from  $H(\omega)$ .

The next approximation is associated with the values  $(N,M) = (1,2)$ . We found that

$$\hat{\beta}_\lambda = \frac{(2\lambda_r + 1)\alpha_1}{1 + \left(\frac{\alpha_1 \omega}{\mu}\right)^2} + (\lambda_r + 1)\alpha_2 \left[ \frac{c_1}{1 + \left(\frac{\omega}{k_1}\right)^2} + \frac{1-c_1}{1 + \left(\frac{\omega}{k_2}\right)^2} \right]. \quad (5.9)$$

In the  $(1,2)$  approximation the response of the fluid is determined when the values of six constants,  $\mu$ ,  $\alpha_1$ ,  $\alpha_2$ ,  $c_1$ ,  $k_1$  and  $k_2$ , are known. The constants  $c_1$ ,  $k_1$  and  $k_2$  may be determined from  $H(\omega)$ .

In the  $(2,2)$  approximation the response of the fluid is determined when eight constants are known, and so on.

In Fig. 5.4 we compare the normalized mean height rise curves given by experiments (the dashed line) with the theoretical height rise curves for the generalized  $(N,M)$  Maxwell models when  $(N,M) = (1,1)$  and  $(1,2)$ . The values of the characterizing constants for these generalized models are determined by requiring that the theoretical and experimental rise curves should match over the largest possible interval  $[0, \omega]$  of oscillation frequencies. We did not compute values of the five parameters which appear in the next member  $(2,2)$  of the sequence. We expect that when the five parameters of the  $(2,2)$  member of the sequence of Maxwell models are chosen optimally, the agreement between theory and experiment will be extended to much larger values of  $\omega$ .

It is necessary here to explain what is meant by agreement between theory and experiment. The  $(N,M)$  Maxwell model has  $2N + 2M$  parameters. The values of three of these parameters are fixed from experiments with steady flow. There are therefore  $2N + 2M - 3$  parameters to be determined. The equations for the height rise is such that we may fit  $2N + 2M - 3$  points of the normalized rise curve by an appropriate selection of the  $2N + 2M - 3$  parameters. This does not guarantee that theoretical and experimental points between  $2N + 2M - 3$  fitted points will lie close together. If the fitted points are widely spaced, theoretical and experimental values at intermediate points will not lie close together. Now consider Fig. 5.4. For the  $(1,1)$  fluid there is just one disposable constant  $k_1^2$ . We fit the point at  $\omega^2 = 25$ . There is a close fit for  $I_{(1,1)}(\omega) = [\omega: 0 \leq \omega^2 < 30]$  (approximately) when

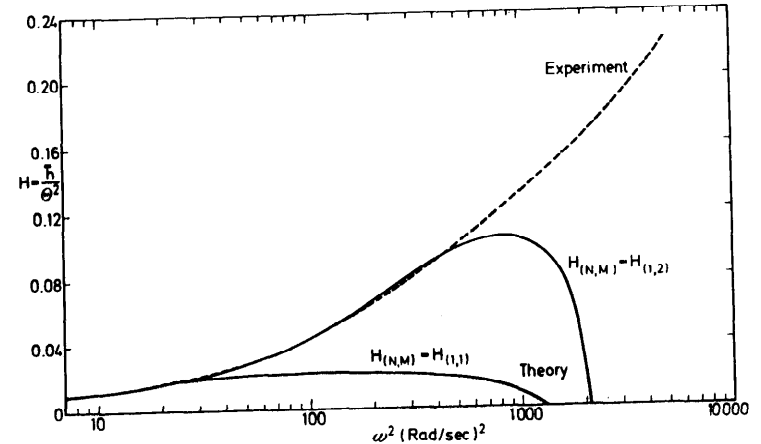


Fig. 5.4 Comparisons of the predictions of the generalized  $(N,M)$  Maxwell model with the observed normalized rise curve for  $(N,M) = (1,1)$  and  $(N,M) = (1,2)$ .

$$k_1^2 = 15.43.$$

For the  $(1,2)$  fluid there are three disposable constants,  $k_1^2$ ,  $k_2^2$  and  $c_1$ . We fit the points at  $\omega^2 = 25, 100$  and  $400$ . There is a close fit for  $I_{(1,2)}(\omega) = [\omega: 0 \leq \omega^2 \leq 450]$  when

$$k_1^2 = 14.50, \quad k_2^2 = 307.0, \quad c_1 = 0.9735.$$

It should be noted that  $k_1^2$  does not change by much. We expect to find a similar extension of the interval of good fit for the higher members of the  $(N,M)$  sequence.

The existence of increasing intervals  $I_{(N,M)}(\omega)$  over which we can fit the curves (and not just points) is one form of agreement between theory and experiment. This kind of agreement is already established for  $I_{(1,1)}(\omega)$  and  $I_{(1,2)}(\omega)$ . Assuming for discussion, that the intervals of agreement may be enlarged by marching up the  $(N,M)$  sequence, we come to the following concept: There is an increasing sequence of generalized  $(N,M)$  Maxwell fluids. Each fluid represents an approximation to a class of real simple fluids in small amplitude periodic motions over an increasing, but restricted, interval  $I_{(N,M)}(\omega)$  of frequencies. The constants which are required to completely specify each fluid in the sequence may be determined by comparison with a universal function similar to that shown in Fig. 5.3. If the concept is good, it will be possible to predict other periodic motions of the now completely determined  $(N,M)$  fluids over similar ranges frequency.

## 5.6 Breathing Instability of the Axisymmetric Steady Clin

The rise of a fluid on a rod in steady rotation is steady

axisymmetric when the angular velocity  $\epsilon$  of the rod is not too large. At a certain critical value of  $\epsilon$  the steady axisymmetric solution loses stability to a small amplitude axisymmetric time-periodic motion in which the rise is alternately filled and emptied of fluid by a pumping mechanism associated with secondary flow. The filling and emptying is analogous to the motion of the lung in breathing and we call the instability a breathing instability. This instability appears to lead to a time-periodic bifurcating solution of Hopf's type in which the amplitude of the breathing and its frequency change continuously with  $\epsilon$ .

The breathing instability may be a manifestation of Rayleigh's instability mechanism for rotating flows (see Joseph (4)). At low speeds, the distribution of angular momentum is the same as a potential vortex flow. In such a flow the angular momentum is constant and is stable by Rayleigh's criterion. The steady secondary motion is driven by normal stresses. It appears to take form as a single cell which carries the lower angular momentum from the outside of the cell near free surface toward the rod and the higher angular momentum away from the rod near the bottom of the cell (see Fig. 4.6). At higher speeds the secondary motion is more intense and it forces an accumulation of low angular momentum fluid into the climbing bubble. The adverse distribution of angular momentum in the bubble is unstable by Rayleigh's criterion; the higher momentum fluid near the rod is pushed outwards by centripetal acceleration. The result is a "bulge" of "thrown outward" fluid supported from below by normal stresses and from the side by increasingly strong surface tension forces generated by the high curvatures on the "bulge". The bulge buildup proceeds until the accumulated fluid is sufficiently heavy to be dragged down by gravity. The bulge then falls into the body of the fluid dragging most of the bubble with it. The depleted bubble is now back at "go" and the same sequence of bubble growth, accumulation of low momentum fluid, and overturning by centripetal acceleration can be initiated once again.

### 5.7 Flower Instability of the Axisymmetric Climb on the Oscillating Rod

There is also a critical value  $\epsilon$  when the rod oscillates with a frequency  $\Omega(a,t) = \epsilon \sin \omega t$ . In this case the basic time-periodic solution is axisymmetric but  $2\pi/\omega$ -periodic in  $t$ . At the critical  $\epsilon$  the axisymmetric configuration loses its stability to another  $2\pi/\omega$ -periodic flow with a different symmetry pattern (Beavers and Joseph, (15)). The new symmetry pattern has a certain integral number of lobes, formed like petals of a flower. The flow arising from the flower instability appears to be an example of supercritical bifurcation of a  $2\pi/\omega$ -periodic solution into another  $2\pi/\omega$ -periodic solution with a different symmetry pattern.

## 6. OTHER FREE SURFACE FLOWS

### 6.1 Second Normal Stresses in Steady and Time-periodic Free Surface Problems

The second normal stress is one of the three scalar functions of the rate of shear which are required to specify the stress in a simple fluid in viscometric flow. Of the three functions the second normal stress is by far the most difficult to measure and even its sign was for a time a matter of heated controversy.

Wineman and Pipkin (19) discovered that the shape of the free surface on a viscoelastic fluid flowing down a tilted open channel

would be proportional to the second normal stress at lowest  $O(\beta^4)$  where  $\beta$  is the angle of tilt. They found that the free surface bulges outward if the second normal stress is negative. Kuo and Tanner (20) reported experiments showing an outward bulge in the fluids studied by them. Sturges and Joseph (21) carried out the perturbation analysis through terms of  $O(\beta^4)$  and showed that secondary motion would not develop until  $O(\beta^6)$  when the channel is infinitely deep or has a semicircular cross-section.

The second normal stress is, of course, a function which arises only in viscometric flows. However, Sturges and Joseph have shown that it is possible to obtain the limiting value of the second normal stress at zero shear from measurements of the free surface on a viscoelastic fluid between oscillating planes. The analysis of Sturges and Joseph (18) is like the analysis of the oscillating rod discussed under Section 5.2. In fact, Joseph (9,1) formulated the problem of the free surface on a simple fluid between cylinders undergoing torsional oscillations. The oscillating rod and oscillating planes arise as limiting cases of the problem between cylinders when the gap between the cylinders is, respectively, large or small. In the problem of oscillating planes it was possible to obtain exact solutions not only for the mean height rise but for the oscillating secondary motions and height rise as well. Among the results obtained by Sturges and Joseph (18) we emphasize the following two:

(i) The mean rise in height of the fluid between oscillating planes is proportional to an unsteady equivalent  $N_2(\omega)$  of the second normal stress. As  $\omega \rightarrow 0$ ,  $N_2(\omega) = 2a_1 + a_2$  where  $2a_1 + a_2$  is the limiting value of the ratio of the second normal stress upon shear rate squared. Observations of the elevation changes on the oscillating free surfaces give rheological data which includes information about the hard-to-measure second normal stress.

(ii) The mean rise between parallel planes is interesting because no rise occurs when planes are in steady motion and the shear rate is constant and uniform over the whole field of flow. In contrast, there is an appreciable steady rise on a rod rotating with even a small, steady, angular velocity. This contrast actually extends to the oscillating planes because the rise between planes is an order of magnitude smaller than the corresponding rise on the oscillating rod. The big mean rise on the rod dominates the whole rise, but between oscillating planes the mean rise is much smaller and steady and unsteady changes in elevation can be equally important.

### 6.2 Die Swell

Die swell is the enlargement of the diameter of a jet of viscoelastic fluid which is extruded from a capillary tube. The die swell phenomenon is not well understood. Even the case of low speed Newtonian jets leads to controversy about the limiting (no speed) value of the swell.

Joseph (22) has studied the problem of the change in diameter of a horizontal capillary jet of viscoelastic fluid with gravity neglected. A unique motionless jet in the form of a straight round cylinder held together by surface tension is an exact solution of the jet problem. Joseph uses this solution to construct a series solution, using the theory of domain perturbations, in powers of a speed parameter. He shows that the perturbation problem which governs at first order is, in the case of a plane

jet, exactly the same as a problem solved by Richardson (23). The problem at first order depends on the viscosity of the fluid; viscoelastic parameters do not enter at first order. The solution at first order has an integrable lip singularity, but the first order approximation to the shape of the jet is a smooth regular function of the axial coordinate.

The presence of an edge singularity in the perturbation analysis raised doubts about the legitimacy of the series solution for the jet problem; the rest solution has no singularity and though the velocity and acceleration tend to zero with  $\epsilon$ , the stresses and pressure at the lip are singular for any  $\epsilon > 0$  no matter how small  $\epsilon$ . We believe the singularity is there, in the physics, but the status of the mathematical solution at orders higher than one is in doubt. It would be of considerable interest to compute the solution of the perturbation problem which arises at second order where viscoelastic effects first appear. Such computations appear to be of more than ordinary difficulty.

Joseph (22) also gave an exact analysis of the momentum of the jet. This analysis was used by Huilgol (24) to obtain upper and lower bounds for die swell. The momentum analysis shows that it is not proper to neglect surface tension in the discussion of low speed jet.

Nickell, Tanner and Caswell (25) have introduced finite element numerical methods to compute the shape of free surfaces without perturbations. So far these methods seem not to have worked out for viscoelastic fluids but they are promising. A review of the finite element work can be found in the paper of Tanner (26).

### 6.3 Edge Effects in Rheometers

Nearly all of the instruments used in laboratories to make rheometrical measurements are based on simplified theories in which edge effects are ignored. These effects are hard to analyze because the motions near the edge are complicated and the position of the free surface defining the edge is unknown. Analysis of the motion and the shape of free surfaces defining edges may be studied as a perturbation of the rest state. An example of such an analysis is Joseph's (22) study of the free surface at the edge of a torsion flow viscometer. Joseph solves the problem by perturbing the angular velocity  $\omega$  of the plates from zero and he computes the secondary motions and free surface which arise at  $O(\omega^2)$ . The solution is expressed in a rapidly convergent series of biorthogonal eigenfunctions whose effect is to turn the flow around at the edge. The secondary flow is driven, at second order, by the vertical stratification of inertia (see Fig. 6.1). The solution shows that turning at edges take place in a distance no more than about three times the plate separation. If the edge is extended to infinity a big inertial eddy remains. In the conventional theory, which assumes that the motion is viscometric, there is no inertial flow. At low speeds edge effects may be neglected when  $d/a < 1$ . Then, the normal stress on the top plate is given by

$$T_{xx} + \text{const} = r^2 \omega^2 \left\{ \frac{4\alpha_1 + 3\alpha_2}{d^2} - \frac{3\rho}{10} \right\}. \quad (6.1)$$

There is a critical gap distance

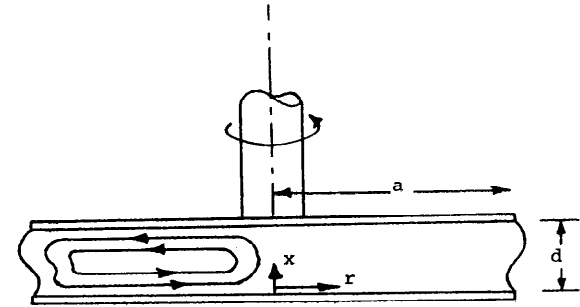


Fig. 6.1 Secondary flow in the torsion flow viscometer

$$d = d_c = \left[ \left( \frac{40}{3} \alpha_1 + 10\alpha_2 \right) / \rho \right]^{1/2}. \quad (6.2)$$

When  $d < d_c$  normal stresses are dominant. When  $d > d_c$  inertia dominates. For STP at room temperature  $d_c \sim 2$  cm; at  $50^\circ\text{C}$ ,  $d_c \sim 0.2$  cm.

It would be of interest to study the free surface on a cone and plate viscometer. The geometrical divergence of the gap between the cone and plate may amplify the effects of free edges.

### 7. CONCLUDING REMARKS

Unfortunately, the potential of perturbation theories is limited to local results. It is better to analyze problems of mechanics by global methods of analysis and numerical analysis. Such global methods are beginning to become available in some branches of mechanics. The value of perturbation theories in the study of the motion of viscoelastic fluids is enhanced by the fact that general, tractable expressions relating stress and deformation are unknown except for motion of a restricted type. We may therefore perturb the data and constitutive equation simultaneously under circumstances in which global representations of

<sup>6</sup> This formula may also be obtained from the formula

$$r \frac{d\bar{p}}{dr} = \frac{3\rho\Omega_1^2 r^2}{10} - [v_1(q) + v_2(q)] - q \frac{dv_2}{dq} \quad (6.3)$$

which has been given by Walters (27, Eq. 4.81). In (6.3)  $q = r \frac{d\omega}{dz}$  is the rate of shear and  $v_1$  and  $v_2$  are normal stress functions. Walters' formula is supposed to give the correction of viscometric flow due to inertia. It seems to us to be inconsistent to correct viscometric flow assumed to be valid at all orders of  $\Omega_1^2$  with an inertial correction valid only up to  $O(\Omega_1^2)$ . To get (6.2) from (6.3) it is necessary to expand the normal stress functions  $v_1 = -2\alpha_1 q^2$ ,  $v_2 = (2\alpha_1 + \alpha_2) q^2$ . Then, to keep a consistent approximation valid through  $O(\Omega_1^2)$  set  $q = r\Omega_1/d$  (use the shear rate at first order).

the constitutive equation, suitable for solving problems, are simply unknown. The study of free surfaces induced by motions of viscoelastic fluids which perturb the rest state is one example of this marriage of analytical convenience with a point of principle.

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