A Test Particle Approach to Flow Classification for Viscoelastic Fluids

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The formulation of the Deborah and Weissenberg dimensionless numbers for viscoelastic fluids by means of a scheme based on the response of hypothetical tracer test particles having the form of elastic dumbbells is presented. By considering the dynamics of these test particles, we can classify flows as being either strong or weak. Consideration of the amount of stretching of a swarm of dumbbells in one relaxation time also provides a definition of the Weissenberg number. A simple workable definition of the Deborah number is given, and, finally, the ideas are applied to a viscoelastic lubrication case, to an inlet flow, and to the flow through porous media.

SCOPE

When we deal with viscoelastic flows, it is necessary to augment the usual range of dimensionless numbers used in fluid mechanics (N_{Re} , N_{Fr} , etc.) because the fluid itself has a characteristic time. This must give rise to the introduction of one more dimensionless parameter. Among the usual Newtonian fluid dimensionless numbers is the Strouhal number, which usually gives the ratio of unsteady inertia forces to steady inertia forces in the flow. Because inertia forces are often neglected in non-Newtonian fluids, it is usual to leave out the Strouhal number and use two new groups, the Deborah and Weissenberg numbers, to make up the requisite numbers of dimensionless groups. The definition of these numbers is considered. A basic idea is that the behavior of viscoelastic

fluids often seems markedly different in steady shearing flows and in flows where an elongational or stretching motion predominates; this is because the latter type possesses much greater power to reorganize the fluid microstructure. Hence it is convenient to introduce the idea of strong and weak flow classifications describing flows which are elongationlike and shearlike, respectively, together with relevant dimensionless numbers. The object of this paper is to introduce kinematic criteria and definitions which are useful in the preliminary stages of viscoelastic flow classification, correlation, or analysis. Some examples are given; these are drawn from lubrication theory and the flow through porous media.

CONCLUSIONS AND SIGNIFICANCE

The method of obtaining the results was to study the dynamics of a set of hypothetical test particles. The form of the test particles was that of an elastic dumbbell, that is, two small spheres joined by a linear elastic spring whose rest length is zero. The spheres are supposed to interact with the viscoelastic fluid through Stokes law. Thus, the test particles are a crude representation of a single macromolecule, and the relaxation time of the test particles is assumed to be the same as the characteristic time of the fluid under study (θ) . It is found that the behavior of the dumbbell, as measured by the distance between the beads, differs markedly according to whether the flow field is of a shearing type or a stretching type. Basically, this occurs because neighboring particles separate at an exponentially increasing rate in elongational types of flows and only linearly in shear flows; our tracer particles try to follow these separation rates. Thus, the elongation (a strong flow) is able to deform the dumbbell much more than the shear (a weak flow). Given a steady spatially uniform flow, we show how to classify all such flows as

weak or strong, and a diagram showing the regions of weak and strong flow is constructed. This classification tells one when to look for large deviations from correlations based on inelastic fluid theories. The adjectives weak and strong describe whether the structure is little or greatly disturbed, respectively, after a long time in the flow. Having computed the amount of distortion of the dumbbells when defining our weak and strong flows, we can consider the amount of stretching of the dumbbells in one relaxation time and so define a Weissenberg number N_{Wi} for both weak and strong flows. In a steady shearing flow, our definition gives the result $N_{Wi} = \sqrt{\theta}$, while in a very strong steady elongational flow this gives the result $N_{Wi} = e^{G'\theta}$. The Weissenberg number does not discuss flow unsteadiness; for this we need the Deborah number. Various forms of Deborah number definitions are considered, and ultimately the simplest is adopted; it is recommended that the characteristic time of the fluid/ residence time of particle in a given region of fluid be used to define the Deborah number. To illustrate the concepts,

we consider the flow of a viscoelastic fluid through packed beds, through a sudden contraction, and in a lubrication type of flow. In the latter case, there is not much structural distortion, contrary to some previously expressed ideas; by contrast, packed beds deform structures efficiently. It is expected that the ideas presented here will be useful in preliminary analysis of viscoelastic flows before highly complex analyses are proceeded with.

In Newtonian fluid mechanics, the characterization of flows via various dimensionless numbers (Reynolds number, Froude number, and so on) has proved to be a very fruitful idea, and one expects that similar schemes may be applicable to viscoelastic fluid flows. Hence one is looking for a further set of dimensionless groups which can be used to correlate viscoelastic phenomena and classify viscoelastic flows. The present position seems to be that two numbers, the Deborah number N_{De} (Reiner, 1964; Metzner et al., 1966a, b; Astarita and Marrucci, 1974) and the Weissenberg number N_{Wi} (White, 1964; Bogue and White, 1970), have been proposed for flow characterization purposes. Both of these numbers depend on the introduction of a characteristic time θ for the fluid; we also accept this idea because there is ample evidence that a characteristic time may be defined, for example in smallstrain relaxation or creep tests, for the types of materials considered here. Then, the Deborah number (Reiner, 1964) may be defined as

$$N_{De} = \omega \theta \tag{1}$$

where ω is a characteristic frequency which measures the unsteadiness of the flow. In certain flows, for example starting-up of a shear flow, ω may take the form (Metzner et al., 1966a)

$$\omega = 1/(\text{time of flow})$$
 (2)

instead of a frequency. The Deborah number is a dimensionless measure of the rate of change of flow conditions at a material particle, and it is clear that the concept is not solely confined to flows which are Eulerian unsteady. It is necessary to measure unsteadiness from a particle or Lagrangian viewpoint. Although the above discussion makes it intuitively clear what is required when ω is defined, it is perhaps not surprising that several definitions of N_{De} , including some unsatisfactory ones, have been proposed. Lodge (1974), working from the point of view of body tensor fields, demands that rheological steadiness be defined from convected coordinates. This point of view is very restrictive and, according to Lodge (1974), casts doubt on the very concept of a universal significant meaning for rheologically steady flow. We shall discuss these ideas again later. Astarita (1967) proposed a definition of N_{De} which is not zero in a rigid body motion. Subsequently, Marrucci and Astarita (1967) proposed another definition which is somewhat unattractive in that quantities defined in the future as well as in the past history of particle deformation are involved. Pipkin (1972) introduced an amplitude vs. dimensionless frequency curve in his classification of shearing flows. Although satisfactory for its original intended purpose, it is not completely clear how to proceed for other flow kinematics. Huilgol (1975) has proposed another definition of N_{De} equivalent to the absolute value of the maximum rate of change of the quantity T(t), where

$$T(t) \equiv \int_0^\infty g(s) [tr C_t(t-s)] ds; \quad g(0) = 1 \quad (3)$$

Here, $C_t(\tau)$ is the right relative Cauchy-Green strain tensor at time τ (Astarita and Marrucci, 1974), and g is a weighting function which decreases with s. Thus, according to Huilgol (1975), we may define

$$N_{De} = \max_{t > \tau > \infty} \left| \frac{dT(\tau)}{d\tau} \right|$$
 (4)

While this definition might be reasonable, it is not obvious how this definition is motivated in physical terms. Thus, there appears to be room for improvement in the definition of N_{De} .

The Weissenberg number was named by White (1964). Here we are concerned with the quantity that Pipkin (1972) has termed the amplitude of shearing in his diagram for shear flows; for steady viscometric flows, it is the rate of shear multiplied by the characteristic time which is important, that is, the amount of shear experienced in one characteristic time. Thus, N_{Wi} may be defined in this case as

$$N_{Wi} = \gamma'\theta \tag{5}$$

This quantity is independent of the Deborah number, which must be zero in a viscometric motion (or any other motion with constant stretch history). Again, the problem of how to extend this definition to other kinematics appears. The reader must wonder at this point why two new dimensionless groups appear when we have only introduced one new dimensional quantity (the characteristic time θ). Slattery (1968) has considered this point and has shown that one may use any two of the Deborah, Weissenberg, and Strouhal numbers in a viscoelastic flow. Let us consider an unsteady (sinusoidal in time) shearing flow with a characteristic frequency ω . In this case, the Strouhal number is $\omega L/V$, where L and V are a characteristic length and a characteristic speed, respectively. Attempts to define a characteristic speed will always come out with a result of the form length \times ω , and so the Strouhal number in this case is merely the ratio of two lengths. Here the Deborah number appears to give a more relevant description. It is believed that this is often true, and hence we shall not use the Strouhal number here. Starting from this premise, one sees from the foregoing discussion that there are a number of puzzles associated with the Deborah and Weissenberg numbers concepts, and it was felt that it would be useful to reconsider the matter from a new viewpoint which was suggested by some molecular model calculations (Tanner and Stehrenberger, 1971).

Suppose we consider the injection of some (possibly hypothetical) tracer particles into our flow field. If they are simply small spheres or other compact particles, they will trace out the fluid particle path lines but will not tell us anything about the expected response of a viscoelastic material element in such a flow. If, however, we consider injecting dumbbell models (Figure 1), consisting of two very small spheres linked by a linear-law spring whose rest length is zero, then the response of these models in a given

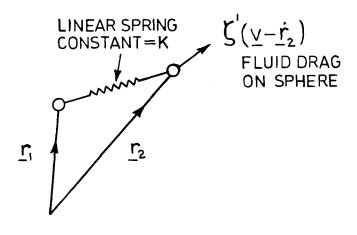


Fig. 1. Dumbbell test particle.

flow will resemble, to a certain extent, the response of viscoelastic fluids in the same flow. The resemblance in behavior is obviously not exact, but it has been demonstrated that a dilute solution of dumbbell models and a plausible network model of a concentrated solution (Lodge, 1970) behave in an identical manner (that is, have an identical constitutive equation for the polymer contribution to the stress tensor) and that this constitutive law is useful for at least qualitative discussion of real viscoelastic flows of concentrated polymer fluids (Lodge, 1964). Unlike the papers cited above, we propose to ignore Brownian motion here. It is known that this must give rise to inaccuracies in shearing types (weak) flows, but it models accurately the transition between weak and strong flows (Tanner, 1975). Furthermore, there is no need of Brownian motion to prevent total collapse of the hypothetical test particles; at time zero they are inserted into the fluid with a definite spacing between the beads. This contrasts with the random excitation existing in a polymer solution. Thus, it seems reasonable to use our hypothetical dumbbell behavior to characterize the kinematics of a flow field. It is found that behavior must mean something related to the length of the vector joining the beads for a single dumbbell; it will actually turn out to be more convenient to consider a swarm of dumbbells and to compute ensemble averages of the quantities of interest. By studying the dumbbell tracer response, we hope to demonstrate a rational method of flow classification and characterization.

DUMBBELL RESPONSE

We consider a set of linear dumbbells (Figure 1) inserted in a noninteracting manner in the fluid under consideration. If the bead friction factor is ζ' and the spring constant is K, then the equation of motion for bead 2 (coordinate r_2) is

$$-K(r_2-r_1)+\zeta'(V-\dot{r}_2)=0$$
 (6)

while that for bead 1 is

$$K(\mathbf{r}_2 - \mathbf{r}_1) + \zeta'(\mathbf{V} - \dot{\mathbf{r}}_1) = \mathbf{0}$$
 (7)

We have assumed that the small spheres have a drag force proportional to their speed relative to the fluid and ignore disturbance of the flow by the test particles. We shall assume the velocity field is homogeneous in space around a given dumbbell, so that at point x the velocity is

$$\mathbf{V} = \mathbf{L}'(t') \mathbf{x} \tag{8}$$

where the components of the velocity gradient matrix L' may in general be functions of time (t') (but not of space, x). To justify this simplification, we assume that the scale of length appropriate to our test particles is much smaller than any length scale appropriate to the flow field. It is also assumed that the centroids of the test particles are convected at the local fluid velocity without slip. It then follows that L' may depend on time even in a Eulerian steady flow and that the variations of the velocity gradients at the particle due separately to unsteadiness and transportation in such a fixed frame are irrelevant; it is only the material or particle-following rate of change that is significant. This point is emphasized here because it seems to be largely independent of the details of the rheology of the fluid. The above formulation is, of course, identical, except for the omission of Brownian motion, to the usual formulation for a dilute solution of linear-spring dumbbell molecules in dilute solution when hydrodynamic interaction between the spheres and internal viscosity are neglected (Tanner and Stehrenberger, 1971). In most cases, the main functions of the Brownian motion are to maintain the molecules in a noncollapsed condition in a quiescent fluid and to maintain a random orientation in the same condition. In many so-called strong flows (Tanner, 1975), its contribution to the force balance on the molecule is minimal except for very small velocity gradients, and except in these conditions, our test dumbbells show molecular behavior, as we will discuss below. Combining Equations (6) to (8) and using the difference coordinate

$$\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1 \tag{9}$$

we find an equation of motion

$$\frac{d\mathbf{r}}{dt'} = \mathbf{L}'\mathbf{r} - \left(\frac{1}{2\theta}\right)\mathbf{r} \tag{10}$$

where $\theta = \zeta'/4K$ is the relaxation time for the test particle. We shall assume that the value of θ chosen for our test particles is the same as the characteristic time scale relevant to the fluid of interest. (Note the interplay of fluid and flow characteristics.) We make Equation (10) dimensionless by defining

$$t = t'/\theta; \quad \mathbf{L} = \theta \mathbf{L}'$$
 (11)

so that (10) becomes

$$\frac{d\mathbf{r}}{dt} = \mathbf{L}(t)\mathbf{r} - \frac{1}{2}\mathbf{r} \tag{12}$$

For any given dumbbell, we suppose that an initial value of r is given at t = 0:

$$\mathbf{r} = \mathbf{r}_0 \quad \text{at} \quad t = 0 \tag{13}$$

Thus, since $\mathbf{r}(t)$ depends only on \mathbf{L} when \mathbf{r}_o is fixed, if we can solve (12) and compute the end-to-end vector \mathbf{r} as a function of time, we can characterize our flow field.

The solution of (12) is (Bellman, 1960)

$$\mathbf{r} = \left\{ \exp \int_0^t (\mathbf{L} - \frac{1}{2}\mathbf{I}) dt \right\} \mathbf{r}_0 \equiv e^{-t/2} \mathbf{F}_0(t) \mathbf{r}_0$$
(14)

where $\mathbf{F}_0(t)$ is the displacement gradient (Lodge, 1974) of \mathbf{r} referred to the initial configuration at t=0. So, for any given dumbbell, the value of $r^2(t)$ may be computed formally as

$$r^2 = e^{-t} \mathbf{r}_o^T \mathbf{F}_o^T \mathbf{F}_o^T \mathbf{r}_o = e^{-t} \mathbf{r}_o^T \mathbf{C}_o(t) \mathbf{r}_o$$
 (15)

where C_o is the Cauchy-Green tensor of continuum mechanics (Lodge, 1974). We now suppose that our seeding of dumbbells was initially random in orientation and that all dumbbells have the same initial length h_o .

If we average over the orientation space, we find the average $\langle r^2(t) \rangle$ as a measure of dumbbell distortion by the flow field. Carrying out this process, we find

$$h^2 = \langle r^2 \rangle = \frac{h_o^2}{3} e^{-t} \operatorname{tr} C_o(t)$$
 (16)

where $h_o = |\mathbf{r}_o|$. The suggestion is that

$$N_{sg} \equiv \left(\frac{h}{h_o}\right)^2 = \frac{1}{3} e^{-t} \{ tr \, C_o(t) \}$$
 (17)

is a suitable criterion for judging the severity of structural distortion in a time t of a structure being convected by a flow field. We shall call N_{sg} the dimensionless stretching parameter; the main difficulty in finding N_{sg} is the computation of $tr C_o(t)$.

While the above results are exact, they are not very useful owing to the need to compute tr C_0 ; this is not a trivial operation in a general flow field. Considering that we are looking for rough guides at the moment, we now propose to simplify the problem of computing C_0 by dividing up the flow history for a particle into segments so that within a given segment, L can be considered constant. This simplifies the problem considerably. If we have a succession of kinematic states L_1 , L_2 , -- L_n and the particle spends times t_1 , t_2 , -- t_n in each segment, then we can compute matrices F_n , giving the strain at the end of the n^{th} time interval relative to a reference state at the beginning of that time interval. Then we can compute by successive matrix multiplication

$$\mathbf{F}_o = \mathbf{F}_{n-1} - \mathbf{F}_2 \cdot \mathbf{F}_1 \tag{18}$$

and hence the total strain matrix C_o may be found for the whole flow history. We will now concentrate on a single time segment extending from time zero to time t', with a constant velocity gradient matrix L'.

In that case, $\dot{\mathbf{F}}_o$, from (14), becomes

$$\mathbf{F}_o = \exp \mathbf{L}t \tag{19}$$

where L is constant in the dimensionless time range 0 to t. For practical use, it is necessary to relate N_{sg} to the elements of L; we expect to have a rough idea of the duration t and the elements of L in our problem. To compute \mathbf{F}_o , we need to find the eigenvalues of L. In general, we will have four cases: L has three distinct real eigenvalues; L has three distinct eigenvalues, two of which are complex conjugates and one of which is real; L has two coalescent real eigenvalues and a third separate eigenvalue; L has all three eigenvalues equal. (In this case, the eigenvalues must all be zero since tr L, which is the sum of the eigenvalues, is zero in an incompressible fluid.)

Except in the fourth case, at least one of the eigenvalues must have a positive real part, since their sum is zero. This approach has been used to classify flows (Tanner and Huilgol, 1975) into strong and weak flows. A slightly different approach is presented below, but the notion of strong and weak flows is again an important idea.

CONSTANT VELOCITY GRADIENTS: STRONG AND WEAK FLOWS

Before going to the finite time case, we can consider the steady state case when $t \to \infty$, and the test particles spend a very long time in a region of constant flow gradi-

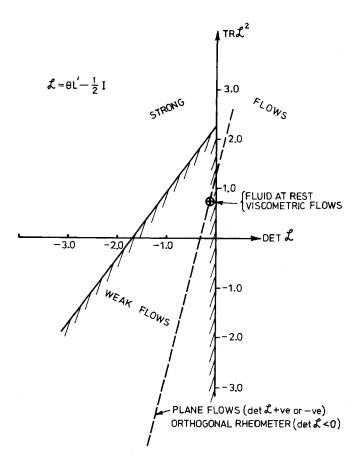


Fig. 2. Regions of strong and weak flow in terms of the invariants of the modified velocity gradient matrix \mathcal{L}_r where $\mathcal{L} = \theta \mathbf{L}' - \mathcal{V}_2 \mathbf{l}$.

ent. From (19) and (15), the value of N_{sg} in (17) will depend only on whether the largest real part of any of the eigenvalues of \mathbf{L} exceeds, equals, or is less than 0.5. In the first case, N_{sg} is infinite, and we have a strong flow; in the two latter cases it is not. Thus, all the flows may be divided into strong and weak flow classes. A convenient way to consider this is to form the matrix $\mathcal{L} = \mathbf{L} - \frac{1}{2}\mathbf{I}$ [see Equation (14)], where \mathbf{I} is the unit matrix. Then, if any eigenvalue of \mathcal{L} has a positive real part, we have a strong flow. Forming the determinantal equation explicitly, noting that $tr \mathbf{L} = 0$ and $tr \mathcal{L} = -3/2$, we find that the eigenvalues of \mathcal{L} satisfy

$$\lambda^3 + \frac{3}{2} \lambda^2 + \frac{\lambda}{2} \left[\frac{9}{2} - tr \mathcal{L}^2 \right] - det \mathcal{L} = 0 \quad (20)$$

Thus, they depend only on the two parameters $tr \mathcal{L}^2$ and $det \mathcal{L}$. We can use the Hurwitz-Routh (Bellman, 1960) criterion to obtain the result that weak flows exist if

$$\det \mathcal{L} < 0 \tag{21}$$

and

$$\frac{27}{16} - \frac{3}{4} \operatorname{tr} \mathcal{L}^2 + \det \mathcal{L} > 0$$
 (22)

Figure 2 shows a chart where these regions of strong and weak flows are delineated in terms of the two parameters $\det \mathcal{L}$ and $\operatorname{tr} \mathcal{L}^2$. We may note that the above criterion gives precisely the same results for strong and weak flows as the criteria previously given (Tanner, 1975), in which Brownian motion was considered. (I am indebted to A. C. Pipkin, Brown University, for producing a proof of this fact, and to E. J. Hinch, Cambridge University, for clarifying discussions on this point.) Since the present criteria are much simpler and easier to evaluate, they are to be preferred to our previous formulation (Tanner, 1975) and are recommended for use. To see that Brownian motion

cannot affect the results, we note that inclusion of it amounts to putting in a random excitation for our models. This excitation would appear on the right-hand side of (10) as a forcing function which does not affect the eigenvalues of the problem. In our previous formulation, we considered the expectation $\langle rr_j \rangle$ and defined a strong flow as one in which at least one of these expectations became infinite. The resulting criteria for this to happen are usually more complex than the present criteria, but they are equivalent, as may be seen directly in the cases of plane flows and simple elongational flows. Our previous result for plane flows with an L matrix of the form

$$\mathbf{L} = \left[\begin{array}{ccc} a & b & 0 \\ c & -a & 0 \\ 0 & 0 & 0 \end{array} \right] \tag{23}$$

was that the flow is weak if

$$a^2 + bc < \frac{1}{4} \tag{24}$$

Computing (21) and (22), we find

$$\det \mathcal{L} = \frac{1}{2}(a^2 + bc - \frac{1}{4}) < 0 \tag{25}$$

and

$$\frac{27}{16} - \frac{3}{4} \operatorname{tr} \mathcal{L}^2 + \det \mathcal{L} = 1 - a^2 - bc > 0 \quad (26)$$

In this case, (26) is included in (25) and gives no new result; (24) and (25) are identical. In the case of a simple elongational flow, where L = diag(G, -G/2, -G/2), it is known (Lodge, 1964) that the flow is weak if

$$-1 < G < \frac{1}{2} \tag{27}$$

If we compute (21) and (22), we find, for a weak flow

$$\frac{1}{8}(G+1)^2(2G-1) < 0$$
 (28)

and

$$\frac{1}{4}G^3 - \frac{3}{4}G^2 + 1 = \left(\frac{G}{2} - 1\right)^2 (G + 1) > 0 (29)$$

Thus, both criteria are significant here. In summary, we see that in these simple flows, in which the Deborah number N_{De} is zero, we find from the present approach that there are two significant dimensionless numbers which are related to the Weissenberg number N_{Wi} ; both $\det \mathcal{L}$ and $\operatorname{tr} \mathcal{L}^2$ play the part of significant dimensionless numbers in classifying steady homogeneous flows.

A DEFINITION OF THE WEISSENBERG NUMBER

Neither $\det \mathcal{L}$ nor $\operatorname{tr} \mathcal{L}^2$ can be regarded as a Weissenberg number. This may be seen by looking at simple shearing or at any other viscometric flow, where we find

$$\det \mathcal{L} = -\frac{1}{8} \quad \text{and} \quad \operatorname{tr} \mathcal{L}^2 = \frac{3}{4} \tag{30}$$

which are independent of shear rate. To define a Weissenberg number, we may go back to the idea of Pipkin (1972) for shear flows, where a significant parameter is the amount of shear in one relaxation time. Instead of the amount of shear, we will take the value of the stretching parameter after one relaxation time, which may be found from (17) to be

$$N_{sg}(1) = \frac{1}{3e} tr C_o(1)$$
 (31)

In simple shearing, with shear rate γ , we find that

$$tr C_o(t) = 3 + \gamma^2 t^2 \tag{32}$$

Hence, we may construct the Weissenberg number or Pipkin amplitude parameter by computing, whenever L is a constant matrix

$$N_{Wi} = \sqrt{3[eN_{sg}(1) - 1]} = \sqrt{tr C_o(1) - 3}$$
 (33)

In a shearing flow, this is just equal to γ (or $\gamma'\theta$ in dimensional terms). For a fluid at rest, $eN_s(1) = 1$, so $N_{Wi} = 0$; for a uniaxial elongational flow (with dimensionless elongational rate G), we find

$$N_{Wi} = \sqrt{e^{2G} + 2e^{-G} - 3} \tag{34}$$

For small rates of deformation, we have

$$tr \mathbf{C}_o(1) = 3 + tr \mathbf{L}^2 + tr \mathbf{L}^T \mathbf{L} + \frac{1}{3} tr \mathbf{L}^3 + tr (\mathbf{L}^T \mathbf{L}^2)$$

$$+\frac{1}{12} tr \mathbf{L}^{4} + \frac{1}{3} tr (\mathbf{L}^{T} \mathbf{L}^{3}) + \frac{1}{4} tr (\mathbf{L}^{2T} \mathbf{L}^{2}) + 0 |\mathbf{L}|^{5}$$
(35)

and hence, to a first approximation

$$N_{Wi} \sim \sqrt{tr \, \mathbf{L}^2 + tr \, \mathbf{L}^T \mathbf{L}} \simeq \sqrt{tr \, \mathbf{D}^2}$$
 (36)

This result is exact [that is, agrees with Equation (33)] in a shear flow; in an elongation flow it gives $N_{Wi} \sim \sqrt{3G}$. For large values of the norm of L in strong flows, an approximation to the magnitude of $tr C_0(1)$ is [compare with Equation (34)]

$$tr C_o(1) \sim e^2 \lambda_m \tag{37}$$

where λ_m is the largest positive eigenvalue of L. Then, an approximate value of the Weissenberg number would be

$$N_{Wi} \sim e \lambda_m$$
 (38)

which shows how the eigenvalues of L enter the problem and how our classification by eigenvalue type at the end of the section on dumbbell response is relevant

THE DEBORAH NUMBER

The concept of the Deborah number has been discussed. We now approach the definition of such a number through the microstructural ideas discussed above. As we have mentioned, in some early uses of the Deborah number a flow time was compared to the fluid characteristic time (Metzner et al., 1966a) in problems involving starting-up from a state of rest. Similarly, in sinusoidal motions it is easy to define a characteristic frequency (Pipkin, 1972) when the Deborah number is formed. In general, we need to identify a characteristic rate of change of flow conditions to define the Deborah number. It is natural that the stress state be taken as the most relevant flow quantity. According to Lodge (1974), who considers convected coordinates and computes the stress state relative to the particle trajectory, there are scarcely any rheologically steady states. Even spatially steady homogeneous flows are not in general deemed rheologically steady from his point of view because the axes of principal stress rotate relative to the particle path framework. Thus, elongational flow is not considered steady in this sense. From the microscopic view, this definition appears to be too restrictive, because the stresses are not changing in magnitude along the particle trajectory. Thus, here we propose only to consider measures of rheological unsteadiness depending on the invariants of the stress states; in a homogeneous flow field these quantities are constant.

Following the work above, it is known that the stresses due to the dumbbells are proportional to the averages $\langle r_i r_j \rangle$ (Tanner and Stehrenberger, 1971; Lodge, 1970), and we can write

$$\langle r_i r_j \rangle \propto \int_{-\infty}^t e^{-(t-\tau)} C_{ij}^{-1}(\tau) d\tau$$
 (39)

where $C_{ij}^{-1}(\tau)$ is the inverse of the right relative Cauchy-Green strain tensor (Astarita and Marrucci, 1974). We can then form the three invariants of the left-hand side of (39) and describe changes in flow conditions by their rates of change when following a particle. The third invariant of (39) is not useful, since $\det C^{-1} = 1$ always. Therefore, only two possible measures of unsteadiness may be formed this way:

(a)
$$\frac{D}{Dt} \int_{-\infty}^{t} e^{-(t-\tau)} tr \mathbf{C}^{-1}(\tau) d\tau$$
 (40)

and

(b)
$$\frac{D}{Dt} \int_{-\infty}^{t} e^{-(t-\tau)} tr C(\tau) d\tau$$
 (41)

In (41) we have used the fact that the second invariant of \mathbf{C}^{-1} is the first invariant of \mathbf{C} for incompressible materials. Note that the proposal (41) is a special case of part of the proposal of Huilgol (1975). We shall adopt (41) here so as to agree with Huilgol (1975) and write

$$N_{De}(t) = \left| \begin{array}{c} D \\ \overline{Dt} \end{array} \int_0^{\infty} e^{-s} tr C_t (t-s) ds \right| \qquad (42)$$

In Huilgol's (1975) definition, given above in Equations (3) and (4), we may, as he does, pick out the largest value of N_{De} over the particle history as the Deborah number. This definition is cumbersome, but it does satisfy the following desirable criteria: in rigid body motions N_{De} is zero; in homogeneous stress fields it gives $N_{De} = 0$; the definition is objective (not dependent on any special frame of reference); and the Deborah number has been shown to be physically motivated. However, definition (42) of the Deborah number has certain drawbacks. It is cumbersome, which may not be too serious, and it is unsatisfactory in an unsteady strong flow. For example, if we consider a suddenly started strong elongational flow, the integral in (42) becomes larger and larger as time goes on, and so does the Deborah number; ultimately $N_{De} \rightarrow \infty$. This does not seem satisfactory. To get over this problem, one idea is to replace the memory function e^{-s} in (42) by another function g(s), as envisaged by Huilgol (1975). The simplest choice which guarantees convergence of the integrals is to take

$$g(s) = 1 \quad 1 > s > 0 g(s) = 0 \quad s > 1$$
 (43)

Then, the definition (42) becomes

$$N_{De}(t) = \left| \frac{D}{Dt} \int_0^1 tr \, C_t(t-s) ds \right| \qquad (44)$$

While the above analyses may be conceptually satisfying, they are not very useful in practice, as it is usually impossible to form useful estimates of all the quantities involved. Therefore, an alternative approach will be suggested which is much simpler.

Let us suppose the flow field has been divided up into regimes in time and space which one is prepared to consider as regions of homogeneous steady flow [see Equation (18)]. Then we may consider whether or not each regime is strong or weak and define a Weissenberg number for each regime as outlined above. It is natural to define the Deborah number for a regime simply as

$$N_{De} = \theta / \text{(residence time in regime)}$$
 (45)

It is believed that (45) is a workable definition of N_{De} , and we now give some examples of its use. This definition of N_{De} is close to that of Metzner et al. (1966a).

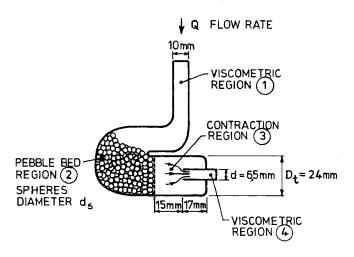


Fig. 3. Nonviscometric flow experiment. This shows the apparatus used by Smith et al. (1975) in their experiments. The sketch is adapted from that paper.

APPLICATION OF THE DEFINITIONS

We will set out for clarity the definitions that will be used in this section:

1. Test for weak flow. Flow is weak if (see Figure 2)

$$det(\mathbf{L} - \frac{1}{2}\mathbf{I}) < 0 \tag{21}$$

and

$$\frac{27}{16} - \frac{3}{4} tr(\mathbf{L} - \frac{1}{2}\mathbf{I})^2 + det(\mathbf{L} - \frac{1}{2}\mathbf{I}) > 0 \quad (22)$$

2. Definition of the Weissenberg Number

$$N_{Wi} = \sqrt{\operatorname{tr} C_o(1) - 3} \tag{32}$$

3. Complicated definition of the Deborah Number at

$$N_{De}(t) = \left| \frac{D}{Dt} \int_0^1 tr \, C_t(t-s) ds \right| \qquad (44)$$

4. Simple definition of N_{De} in a steady homogeneous flow regime

$$N_{De} = \theta / \text{(time of dwell in regime)}$$
 (45)

To begin with, consider a suddenly started shearing flow. This is a weak flow, and the Weissenberg number N_{Wi} is $\gamma H(t)$, while from (44)

$$N_{De}(t) = 2\gamma^2 t(1-t) [1 - H(t-1)]$$
 (46)

The maximum value of this expression occurs at $t=\frac{1}{2}$ and is $\frac{1}{2}\gamma^2$. In the simplified method, one has to divide the time of flow somewhat arbitrarily in this case. Upon switching on the flow, the structure will tend initially to be moved at a rate proportional to γ' . Hence, it is natural to divide the time by taking one segment of length $1/\gamma'$ and one of infinite length. In the initial segment

$$N_{De} = \gamma = \gamma'\theta \tag{47}$$

which is a reasonable result from the structural point of view. For a sinusoidally varying shear flow, the flow is again weak, and the result from (44) is very complex and unwieldy (see Huilgol, 1975). In the simplified approach, if one considers that the sinusoidal motion is approximated by a square wave of the same frequency, the residence time is π/ω , and hence the Deborah number is

$$N_{De} = \omega \theta / \pi \tag{48}$$

This agrees, except for the unimportant π factor, with the Pipkin diagram definition.

Similarly, we could construct further examples in basic simple flows, but instead we will now consider some less artificial examples. First, we take the flow used in the experiment by Smith et al. (1975). Their device is depicted in Figure 3. Four flow regions can be discerned:

1. A weak nearly viscometric flow upstream of the pebble bed.

2. The flow through the pebble bed.

3. The region of contraction.

4. The region deep inside the exit tube, again a viscometric flow.

The weak flow regimes 1 and 2 can be readily handled and are of little interest. Regions 2 and 3 are not viscometric flows. In region 2, a particle undergoes a very complex motion in the interstices between the pebbles. It is hopeless to apply the complex result (44) for the Deborah number. Let us consider the pebble-bed settling region and apply the usual concepts (Bird et al., 1964) for packed beds. Then, if Vo is the (empty bed) macroscopic speed, and R_h is the hydraulic radius, then a reasonable estimate of the elements of L is that they are of order $2V_o$ / ϵR_h , where ϵ is the void ratio. (This calculation merely assumes that the porous medium is made up of a set of identical tubes, each with a Poiseuille flow. Alternatively, one can consider the dissipation of energy and obtain similar results.) Substituting for R_h from the formula given by Bird, Stewart, and Lightfoot (1964), we find an estimate of the Weissenberg number for a packed bed of spheres to be

$$N_{Wi} \simeq \frac{12(1-\epsilon)}{\epsilon^2} \left(\frac{V_o \theta}{d_s}\right)$$
 (49)

if the flow is not very strong. If we consider a close-packed bed with a porosity of about 0.27, then the factor $12(1-\epsilon)/\epsilon^2$ is about 120; James and McLaren (1975) in a more detailed computation find maximum stretching rates of about $150(V_o/d_s)$, and hence (49) is a reasonable estimate which will be used here. The bead beds form a repeating pattern, and one can estimate the Deborah number from the time taken to traverse a distance of the order of a sphere diameter. Hence, we take

$$N_{De} \simeq \frac{V_o \theta}{\epsilon d_s} \tag{50}$$

If $N_{De} \sim 0(1)$, then N_{Wi} , from (49), will be of order $12(1-\epsilon)/\epsilon$; for $\epsilon=0.37$ (random packing), this ratio is about 20. Thus, a packed bed is likely to be an efficient stretching device for molecules.

Now let us consider the contraction portion of the apparatus. Here the inlet length is probably of the order of the small tube diameter d, and the stretch rates and shear rates are thus of order Q/d^3 . The particle residence time in this stretching region is of order d^3/Q , and hence we may again define Weissenberg and Deborah numbers as we did above for the pebble-bed portion. It is of interest to compare the two regions. Let $V_o = 4Q/\pi D_t^2$, where D_t is the diameter of the pebble bed. Then we see that the contraction is the stronger flow if

$$1 >> \frac{12(1-\epsilon)d^3}{\epsilon^2 D_t^2 d_s} \tag{51}$$

From the measurements given in Figure 3 (Smith et al., 1975), we find the right-hand side of (51) to be about 8.0. Thus, some effect of the pebble bed on the flow is to be expected. The observed values of molecular elongation ratio in the experiment were about 4. This is consistent with a residence time in a strongly elongational regime of slightly over one relaxation time, and this emphasizes the difficulty of creating large volumes of elongational flow.

We will now briefly reconsider the flow of polymer fluids through porous media. Much of the literature has been reviewed by James and McLaren (1975), and it is clear from their work that at a critical value of $\theta V_o/d_s$ of order 0.01, greatly increased flow resistance was observed with dilute polyethylene-oxide-in-water solutions. The critical value of $\theta V_o/d_s$, with geometrical factors taken into account [Equation (49)], was consistent with the local product of stretching rate \times relaxation time being of order unity, as we would expect from the above analysis. With this check on our ideas, we can now reconsider some of the results in the literature on flow through porous media which are puzzling. Sadowski (1965) and Marshall and Metzner (1967) showed data with an upturn in resistance to flow. Gaitonde and Middleman (1966), on the other hand, show no upturn in resistance. If we concentrate on the two latter papers, we find that some of the tests were done with a common polymer (L-100 polyisobutylene) in decalin and toluene, respectively The difference in their results for these solutions is most probably due to the different choices in time constants θ made by the two sets of investigators. Gaitonde and Middleman (1966) used the longest Bueche relaxation time. This is probably not relevant, and it would be preferable to use the Rouse relaxation time in the type of analysis considered in this paper. Marshall and Metzner (1967), on the other hand, used a time constant constructed from normal stress and viscosity measurements in simple shear flow. Roughly speaking, their value of θ decreases as shear rate increases; the decrease is proportional to the viscosity. Thus, they give a spread of relaxation times from 34 to 0.96 ms. From the point of view adopted here, only the highest value of θ is significant. When this is taken into account, the formerly predicted value of $\theta V_o/d_s$ of 1 now moves out to $\theta V_o/d_s \sim$ 25. The onset of increased pressure drop for this set of experiments occurs when $\theta V_o/d_s$ is about 0.05. Since the void ratio ε was 0.486 for this porous medium, application of Equation (49) shows that the onset Weissenberg number is about 1.3, which is consistent with our present ideas that it should be roughly 0.5. Equation (50) shows that the residence time is roughly ten relaxation times, so that a good deal of stretching can be expected in these flows.

Using their Bueche time constant, Gaitonde and Middleman (1966) found no increase in pressure drop up to $\theta V_o/d_s \sim 1.6$. If we use the data collected by Tanner (1973) to estimate the same type of time constant as that used by Marshall and Metzner (1967), then their values of $\theta V_o/d_s$ would probably only go up to about 0.4. However, one would still expect a large increase of resistance if their particular polymer solution shows a characteristic increase of stretching viscosity. Being much more concentrated than that of Marshall and Metzner, it is possible that this solution does not show a large stretching viscosity. Other data which do not show a large increase of pressure drop over the Newtonian correlation have been given by Siskovic et al. (1971), Jones and Maddock (1969), and McKinley et al. (1966). One must conclude provisionally that these fluids are less elastic than those used by the other investigators cited here, and that our dumbbell model does not apply here. Some further experimentation would be welcome.

The definition of θ to be used needs some consideration. The best situation would be if the spectrum of relaxation times was given for the fluid, and each component of the spectrum was considered separately. Normally, this information will not be available, and a more approximate measure is needed. It seems likely that the only accessible estimate of θ that can be used is that used by Marshall and Metzner (1967); however, it is necessary that the limit of θ , as stresses become small, be used. Thus, as a

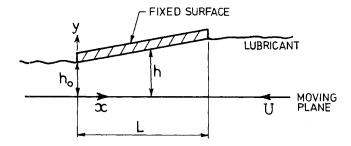


Fig. 4. Two-dimensional thrust bearing.

practical matter, it seems preferable that one should define θ from measurements of the first normal stress difference (N_1) and the zero shear rate viscosity (η_0) in the form

$$\theta = \lim_{\gamma \to 0} (N_1/\gamma^2)/2\eta_0 \tag{52}$$

where γ is the shear rate. (The first normal stress difference is defined by the difference in stresses $t_{xx} - t_{yy}$ in a shearing flow where the x component of velocity is γy .) For a discrete spectrum of relaxation time, it may be shown (Lodge, 1964) that if the relaxation times are θ_n , and if the fraction contributed to the zero shear viscosity by the relaxation process θ_n is p_n , then the value of θ given by Equation (51) is

$$\theta = \sum_{n=1}^{m} p_n \theta_n \tag{53}$$

Other weightings can be conceived, but (52) is the result of a fairly simple experimental procedure which gives a definite average. Provided the distributions are not too different, one should be able to compare two fluids with different spectra.

As a second example, we will consider a lubricated thrust bearing, Figure 4. Several treatments have appeared in the literature (Tanner, 1963, 1969; Metzner, 1968; Walters, 1972); some use the argument that the flow is strong and should be treated as such (Metzner, 1968), while others deny this. Let us consider the Newtonian flow field. In the absence of side leakage, so that the flow is plane, this is given by

$$\frac{u}{U} = -1 + \frac{y}{h} - 3\left(\frac{y}{h}\right)\left(1 - \frac{h^*}{h}\right)\left(\frac{y}{h} - 1\right) \tag{54}$$

and

$$\frac{v}{U} = -\frac{dh}{dx} \left(\frac{y}{h}\right)^2 \left(2 - \frac{3h^*}{h}\right) \left(\frac{y}{h} - 1\right) \quad (55)$$

Here (Figure 4), the film thickness h=h(x), and h^{\bullet} is a constant; h^{\bullet} is the value of the film thickness where the pressure reaches a maximum. It is assumed in the development of (54) that h/L << 1; usually, $h/L \sim 10^{-3}$. We can now explore the strength of the flow using Equation (24); the flow is strong if

$$\left(\frac{\partial u}{\partial x}\right)^2 + \frac{\partial u}{\partial y} \frac{\partial v}{\partial x} > \frac{1}{4}\theta^2 \tag{56}$$

We shall now assume that the slider is straight, so that

$$h = h_o \left(1 + \frac{mx}{L} \right) \tag{57}$$

where h_o is the minimum film thickness, L is the bearing length, and m is a constant. Three planes have been investigated: the moving plane y = 0, the fixed plane y = h, and the intermediate plane y = h/2. In the first two, the flow is weak, since the left-hand side of expression

(56) vanishes. This is to be expected on a solid surface. In the third case, recognizing that d^2h/dx^2 is zero, and denoting dh/dx by h'(x), we find that the criterion (56) shows that the flow is strong if

$$\frac{9}{4} \theta U h' \frac{h^{\bullet}}{h^2} > 1 \tag{58}$$

For the straight slider, $h' = mh_o/L$, and if the smallest value of (58) is taken at the inlet plane, where $h = h_o$ (1 + m), then the flow is strong all along the plane y = h/2 if

$$\frac{9\theta Um}{4L(1+m)^2} \left(\frac{h^{\bullet}}{h_o}\right) > 1 \tag{59}$$

Typically, $m \approx 1$ (Fuller, 1956), and $h^{\circ}/h \sim 1 + m/2$. Using m = 1, we find the flow is strong on the plane y = h/2 if

$$\frac{\theta U}{L} > \frac{32}{27} \tag{60}$$

Note that the flow is still weak at the solid boundaries.

The Weissenberg number is not easy to work out from Equation (33), but since the flow is not far from viscometric, it is found from (36) that

$$N_{Wi} \sim 0 \left(U\theta/h_o \right) \tag{61}$$

for all stations in the bearing. Similarly, to work out the Deborah number, we shall consider each particle to follow a straight line trajectory through the bearing and to be subjected for the appropriate time to an average elongation field. At the fixed wall, clearly $N_{De}=0$, and on the moving boundary, the transit time is L/U, and $N_{De}=\theta U/L$. For the bearing as a whole, the mean transit time is 2L/U, and the appropriate Deborah number is thus

$$N_{De} = \theta U/2L \tag{62}$$

Now one can assess the flow dynamics. By looking at (14), we see that a large extension of the structure in a simple elongation will only occur if the product of $\partial u/\partial x$ and the residence time t_r exceeds unity. To secure a relative elon-

gation of (say) 100, it is necessary that $t_r \frac{\partial u}{\partial x} \sim \log_e 100$

= 4.6. It seems impossible to achieve this with the bearing analyzed because the product $t_r \frac{\partial u}{\partial x}$ is about 1, and thus

it is not expected that the structure will be much disturbed from the equilibrium configuration. This, in turn, implies that the nonviscometric terms will not be very important in this flow. This conclusion does not seem to be very sensitive to the exact flow field postulated; the mean residence time is not expected to change much from the magnitude calculated above, and, similarly, the rate of stretching must be of order U/L. Hence, no great structural changes are to be expected. Consequently, nonviscometric effects are not pronounced.

Further examples could be set out; for example, the flows in dies in injection moulding may be considered in the same spirit. It is hoped that the present discussion will help in the practical analysis and design of viscoelastic flow systems; wherever there is a flow which is a mixture of elongation and shearing, with a finite residence time in the "elongational" region, then I believe it should first be examined by using the simple approaches developed here, before more elaborate analyses are contemplated. Of course, it may happen that the fluid is not very elastic, in the sense that no great changes in behavior are observed around $N_{Wi} \sim 0(1)$, as in some of our examples in the flow through packed beds. Even in these cases, the cor-

relation will be useful. It may also help in the choice of constitutive models, as suggested by Huilgol (1975), if more complex analyses are to be proceeded with.

NOTATION

= Cauchy-Green strain tensor; $\Delta \mathbf{F}_o^T \mathbf{F}_o$, where \mathbf{F}_o^T is the transpose of \mathbf{F}_o

= right relative Cauchy-Green strain tensor

 d_s , D_t , d = diameters, Figure 3 = rate of deformation tensor

D/Dt = rate of change following a particle $= \partial r/\partial r_o$, a tensor; note $F_{ij} \triangle \partial r_i/\partial r_{oj}$

 \boldsymbol{G} = elongation rate

= film thickness, Figure 4, dumbbell end-to-end hlength at time t

= initial length of dumbbell, initial film thickness, h_o

 h^{\bullet} = film thickness where pressure gradient is zero

= identity matrix T

K = dumbbell spring stiffness = characteristic length

L'(L) = dimensional (dimensionless) velocity gradient matrices

 $= L - \frac{1}{2}I$ \mathcal{L} = constant

= first normal stress difference N_1

 N_{De} = Deborah number N_{sg} = stretching number = Weissenberg number N_{Wi}

= weighting factors for average time constant, Equation (53)

= flow rate

 $\mathbf{r_1}$, $\mathbf{r_2}$ = bead position vectors, Figure 1 = end-to-end dumbbell vector r

 R_h = hydraulic radius t'(t) = time, dimensionless= residence time

= velocity component along x direction

= sliding speed, Figure 4 U

= velocity component in y direction

V = characteristic speed

= empty bed mean velocity for porous medium V_o

= fluid velocity vector = position vector, x, y, z

Greek Letters

 $\gamma'(\gamma)$ = shear rate, dimensionless shear rate

= porosity of pebble bed = fluid characteristic time = bead friction factor

= largest positive eigenvalue of ${f L}$ λ_m

= zero shear rate viscosity

= characteristic frequency or rate

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