

## THE RHEOLOGICAL ASPECT OF HYDRODYNAMICS\*

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1. When the rheologist considers liquids, he is interested in them as *materials* having certain material properties. Now, when he turns in his investigations to hydrodynamics for help, what she offers is the mathematics of *two* sorts of liquids:—One is called the *ideal* liquid and has no material property besides elasticity of volume determined by the bulk modulus  $\kappa$ , connecting a hydrostatic pressure  $p$  with a volume expansion  $e$ , by

$$p = -\kappa e, \quad (1)$$

the other, the *viscous* liquid, has, in addition to volume-elasticity, a property called viscosity ( $\mu$ ),<sup>1</sup> defined quantitatively since Newton as the ratio of shearing, or tangential, stress ( $p_t$ ) to the velocity gradient ( $G$ ), or

$$\mu = p_t/G. \quad (2)$$

The latter is better replaced by the tangential component of the rate of strain ( $\dot{e}_t$ ) which is half the velocity gradient<sup>2</sup>

$$\dot{e}_t = G/2; \quad (3)$$

so that

$$\dot{e}_t = p_t/2\mu. \quad (4)$$

The rheologist is at once confronted with the question: Is this enough? Can he for the description of the rheological behaviour of liquids manage with these two “coefficients”:  $\kappa$  which refers to *all* materials; and  $\mu$  which refers specifically to liquids? This question will presently be examined.

2. Rheologists very often have to deal with highly viscous substances, of the order  $10^6$  to  $10^{10}$  poises, such as tar and pitch, for which the standard “shear” methods of viscometry are very inconvenient. Since Trouton (1906) such substances are formed into cylindrical rods which are subject to pull and the rate of elongation is observed. Let  $p_{zz}$  be the tractional force per unit area of cross section and  $\dot{e}_{zz}$  the *rate* of elongation per unit length, with the  $z$ -coordinate in the direction of the axis of the cylinder, then Trouton defined a coefficient of *viscous traction* by the equation

$$\lambda_T = p_{zz}/\dot{e}_{zz}. \quad (5)$$

The coefficient  $\lambda_T$  will evidently depend in some manner upon  $\mu$ . As a matter of fact the Stokes-Navier differential equations, which are claimed by classical hydrodynamics to be all-embracing, should be sufficient to deal with the case.

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<sup>1</sup>For easy reference I shall use Lamb's notation. Rheologists usually denote the coefficient of viscosity by  $\eta$ .

<sup>2</sup> $\dot{e}_t$  is sometimes identified with  $G$ , but (3) is a better definition.

In order to find the relation between  $\lambda_T$  and  $\mu$  Trouton reasoned as follows:<sup>3</sup>

When the pull  $p_{zz}$  is applied, there will be an *initial* state, the *first* stage in the experiment, when the rod is not only elongated but at the same time also expands, the measure of the volume expansion being at every moment  $e_v$  and its rate  $\dot{e}_v = \theta$ . This volume expansion produces an elastic reaction of the nature of a hydrostatic tension in accordance with (1). The pull  $p_{zz}$  has an isotropic component  $p_{zz}/3$ .<sup>4</sup> When  $e_v$  has so much increased that  $\kappa e_v = p_{zz}/3$ , the elastic reaction balances the isotropic component of the pull. With this, the volume expansion ceases to increase and its rate of increase  $\dot{e}_v$  accordingly vanishes. A *second* stage then sets in, in which  $\dot{e}_v = \theta$  is permanently equal to zero. Let us forget about the first stage and start observations only after the second stage has been reached. We then can make use of the equations (4,L) Art. 328 on page 577 of the sixth edition of the classical work of Lamb (1932). In order to get rid of the action of gravity, we immerse the rod, as was done by Trouton, in a liquid of equal density. The movement being slow, we may also neglect inertia forces. Then the Stokes-Navier equations become

$$-\partial p/\partial x + \mu \nabla^2 u = 0 \quad (6)$$

and two others where  $x$  is replaced by  $y$  and  $z$  respectively and  $u$  by  $v$  and  $w$ .

A solution of (6) is given by

$$u = qx, \quad v = qy, \quad w = rz \quad (7)$$

in which case  $p$ , the hydrostatic pressure, is constant throughout the rod.

In order to determine  $q$  and  $r$ , we go back to Lamb's equations (5, L), and (6, L), Art. 326 on page 574, from which we find the stresses

$$p_{zz} = -p + 2\mu \partial u/\partial x; \quad p_{vz} = \mu(\partial w/\partial y + \partial v/\partial z) \quad (8)$$

and similarly  $p_{vv}$ ,  $p_{zz}$ ,  $p_{zz}$ ,  $p_{zv}$ .

Now from our (7),

$$\partial u/\partial x = \partial v/\partial y = q; \quad \partial w/\partial z = r; \quad \partial w/\partial y + \partial v/\partial z = 0 \quad (9)$$

and therefore

$$p_{zz} = p_{vv} = -p - \frac{2\mu}{3}(r - q), \quad (10)$$

$$p_{zz} = -p + \frac{4\mu}{3}(r - q).$$

We know  $p_{zz}$  and as no forces act on the sides of the bar,  $p_{zx}$  and  $p_{vv}$  must vanish. This gives us two equations

$$p + \frac{2\mu}{3}(r - q) = 0, \quad (11)$$

$$p - \frac{4\mu}{3}(r - q) = -p_{zz},$$

<sup>3</sup>In what follows Trouton's arguments are not reproduced literally, but in their essence.

<sup>4</sup>The isotropic component of a stress is the mean normal traction  $p_m = (p_{xx} + p_{yy} + p_{zz})/3$ . In our case  $p_{xx} = p_{yy} = 0$ .

which together with the third

$$\dot{e}_v = \theta = \partial u / \partial x + \partial v / \partial y + \partial w / \partial z = 2q + r = 0 \quad (12)$$

yield

$$p = -p_{zz}/3 \quad (13)$$

and

$$r = p_{zz}/3\mu, \quad (14)$$

$$q = -p_{zz}/6\mu,$$

or [compare (9)]

$$\dot{e}_{zz} = \partial w / \partial z = r = p_{zz}/3\mu \quad (15)$$

from which [compare (5)]

$$\lambda_T = 3\mu. \quad (16)$$

3. However, this reasoning of Trouton's begs the problem. How is one to know when the first stage has come to an end? In materials of such high viscosity this may, for all we know, take quite a long time. One cannot rest satisfied with such a state. The rheologist often has a practical interest in the first stage. He has the feeling that Stokes conjured away another coefficient of viscosity. Actually Lamb's equations (5, L) and (6, L) which we used above, are gained by specialization of more general equations, namely (2, L) and (3, L), page 574 which are<sup>5</sup>

$$p_{zz} = -p + \lambda\theta + 2\mu \partial u / \partial x, \quad (17)$$

$$p_{vv} = \mu(\partial w / \partial y + \partial v / \partial z).$$

By defining (compare Art. 325, p. 573)

$$-p = (p_{zz} + p_{vv} + p_{zz})/3 \quad (18)$$

and introducing this definition, Lamb finds

$$3\lambda + 2\mu = 0 \quad (19)$$

from which

$$\lambda = -2\mu/3. \quad (20)$$

This must cause confusion. As  $p_{zz}$ ,  $p_{vv}$ ,  $p_{zz}$  may, on principle, depend "also on the rate of expansion at the point  $(x, y, z)$ "<sup>6</sup>,  $p$ , accordingly, is in general not the hydrostatic pressure, i.e. not that pressure which is independent of the rate of strain. If we, as most workers will be inclined to assume without question, use  $p$  to denote the *hydrostatic* pressure, we shall not find (19), but will in general have

$$3\lambda + 2\mu = 3\mu, \quad (21)$$

<sup>5</sup>This  $\lambda$  should not be confused with Trouton's coefficient of viscous traction  $\lambda_T$ .

<sup>6</sup>Compare footnote on page 573 l.c.

where  $\mu_v$  is another coefficient which may be named "volume viscosity". It therefore appears that Stokes had assumed the volume viscosity to vanish. This is borne out by the following quotation from Stokes, to which Tisza (1942) has drawn attention: ". . . of course we may at once put  $\mu_v = 0$ , if we assume that in the case of a uniform motion of dilatation the pressure at any instant depends only on the actual density and temperature at that instant and not on the rate at which the former changes with the time. In most cases to which it would be interesting to apply the theory of the friction of fluids, the density of the fluid is either constant or may without sensible error be regarded as constant, or else changes slowly with time. In the first two cases, the results would be the same and in the third nearly the same, whether  $\mu_v$  were equal to zero or not. Consequently, if theory and experiments should in such cases agree, the experiments must not be regarded as confirming that part of the theory which relates to supposing  $\mu_v$  to be equal to zero". Tisza has objected to this assumption, which, according to him, is valid in monoatomic gases only while in polyatomic gases and liquids  $\mu_v \gg \mu$ . His arguments are based on considerations of supersonic absorption. We shall presently show that at the other extreme in very viscous liquids Stokes assumption also leads to unacceptable results.

4. We now start from

$$p_{xx} = -p + (\mu_v - 2\mu/3)\theta + 2\mu\partial u/\partial x \quad (21)$$

Only  $p_{zz}$ , the "viscous traction" acts in our case and both  $p_{xx}$  and  $p_{yy}$  are absent  
Therefore

$$\begin{aligned} -p + (\mu_v - 2\mu/3)\theta + 2\mu\partial u/\partial x &= 0, \\ -p + (\mu_v - 2\mu/3)\theta + 2\mu\partial v/\partial y &= 0, \\ -p + (\mu_v - 2\mu/3)\theta + 2\mu\partial w/\partial z &= p_{zz} \end{aligned} \quad (22)$$

from which, by adding up,

$$\theta = (p_{zz}/3 + p)/\mu_v. \quad (23)$$

Introducing this expression into the third of (22) we get

$$e_{zz}^{\cdot} = \partial w/\partial z = [p_{zz} + p - (\mu_v - 2\mu/3)\theta]/2\mu \quad (24)$$

which on re-arrangement becomes

$$e_{zz}^{\cdot} = [(p_{zz} + 3p)\mu + 3p_{zz}\mu_v]/g\mu\mu_v. \quad (25)$$

Introducing the expression for the hydrostatic pressure  $p$  from (1) we now get for (5)

$$\lambda_T^{-1} = e_{zz}^{\cdot}/p_{zz} = [(1 - 3\kappa e_v/p_{zz})\mu + 3\mu_v]/9\mu\mu_v \quad (26)$$

and therefore

$$\lambda_T = [g\mu\mu_v(1 - 3\kappa e_v/p_{zz})^{-1}]/[\mu + 3\mu_v(1 - 3\kappa e_v/p_{zz})^{-1}]. \quad (27)$$

Equation (27) is the viscous analogy to the classical relation between Young's modulus ( $E$ ), the modulus of rigidity ( $G$ ) and the bulk modulus ( $\kappa$ ), viz.  $E = 9G\kappa/(G + 3\kappa)$ . It should, however, be noted that the analogy is not exact. While  $E = 3G$  for  $\kappa = \infty$

and *only* in this case, the situation is very different with regard to  $\lambda_T$  as will presently be discussed.

5. In interpreting (27), one should keep in mind that  $1 - 3\kappa e_v/p_{zz} \geq 0$ , where the sign of inequality is valid in the first, the sign of equality in the second stage of Trouton's experiment. For the volume viscosity we have  $0 \leq \mu_v \leq \infty$ . It is clear that the volume viscosity cannot be negative as "otherwise the more alternate expansion and compression, alike in all directions, of a fluid, instead of demanding the exertion of work upon it, would cause to give work out" (Stokes). As can be seen, in the first stage, if  $\mu_v$  vanishes,  $\lambda_T$  also vanishes. This would mean that in the first stage of Trouton's experiment the viscous resistance of a very viscous liquid against extension vanishes, *no matter how high the ordinary viscosity*  $\mu$  of the liquid, a result at variance with our ideas of viscous flow. On the other hand  $\lambda_T/\mu = 3$  in both stages if  $\mu_v = \infty$  and also in the second stage whatever the magnitude of  $\mu_v$ . Because  $\mu_v$  cannot be negative,  $\lambda_T$  cannot exceed the value 3. Therefore  $0 \leq \lambda_T \leq 3$ .

Examination of (27) now shows that Stokes was mistaken in equalizing the influence of either  $e_v = 0$  or  $\mu_v = 0$  on experimental results. We see that the same result follows from either  $e_v = 0$  or  $\mu_v = \infty$  and not  $\mu_v = 0$  and generally  $\mu_v \gg \mu$ . It may be mentioned that Tisza deduces from supersonic absorption in certain liquids for  $\mu_v/\mu$  a value of 2000. This, for all rheological purposes, is infinite.

6. Having discovered in this way another rheological coefficient besides  $\mu$ , namely  $\mu_v$ , we have become suspicious. There may be many other coefficients, concealed from us by the biased procedure followed in classical hydrodynamics. The rheologist therefore makes himself independent and starts *ab ovo*.

We first have to take account of the fact that even the simplest liquid is complex insofar as it has something in common with the solid, namely the property of *not flowing* under the action of a hydrostatic pressure (or tension). A hydrostatic stress will change the volume of the liquid in a reversible elastic manner. That stress is equal to  $\kappa e_v$ . In order to investigate the rheological properties of the liquid *qua liquid*, we must deduct from the applied stress  $p_{rs}^*$  the hydrostatic stress  $\kappa e_v \delta_{rs}$  where the stress tensor  $p_{rs}$  is defined by

$$p_{rs} = \begin{vmatrix} p_{xx} & p_{xy} & p_{xz} \\ p_{yx} & p_{yy} & p_{yz} \\ p_{zx} & p_{zy} & p_{zz} \end{vmatrix} \tag{28}$$

and the unit tensor  $\delta_{rs}$  by

$$\delta_{rs} = \begin{vmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{vmatrix} . \tag{29}$$

Only what remains, namely

$$p_{rs} = p_{rs}^* - \kappa e_v \delta_{rs} \tag{30}$$

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<sup>7</sup>The asterisk indicates the stress as applied, i.e. including the hydrostatic part.

will be the stress due to flow and depending upon the tensor of rate of strain  $\dot{e}_{rs}$ . Thus  $p_{rs}$  is the viscous resistance in the widest sense. Instead of the very special equation (4) we now write

$$\dot{e}_{rs} = f(p_{tu}) \tag{31}$$

and our task is to develop the function  $f$  in as general a way as possible. Reiner (1945) has shown how this can be done and the result is

$$\dot{e}_{rs} = \mathfrak{F}_0(P)\delta_{rs} + \mathfrak{F}_1(P)p_{rs} + \mathfrak{F}_2(P)p_{r\alpha}p_{\alpha s} \tag{32}$$

where the  $\mathfrak{F}$  are functions of the three invariants  $P_1, P_2, P_3$  of the stress tensor  $p_{rs}$  and  $p_{r\alpha}p_{\alpha s}$  is a tensor of the same rank as  $p_{rs}$ , the components of which can be written down if we remember the summary convention of tensor analysis and note that the suffix  $\alpha$  appears twice.<sup>8</sup>

All rheological coefficients are contained in the functions  $\mathfrak{F}_0, \mathfrak{F}_1$  and  $\mathfrak{F}_2$ . In order to extract them from there, we perform rheological experiments and adapt (32) to the experimental conditions. Let us first drop the last term on the right side of (32) and deal with the equation

$$\dot{e}_{rs} = \mathfrak{F}_0(p)\delta_{rs} + \mathfrak{F}_1(P)p_{rs} \tag{33}$$

where  $\mathfrak{F}_0$  and  $\mathfrak{F}_1$  are furthermore to be functions of the first and second invariant only.

It will facilitate our considerations if we resolve  $\dot{e}_{rs}$ , as can be done with every tensor, in an isotropic ( $\dot{e}_v$ ) and a deviatoric ( $\dot{e}'_{rs}$ ) part as follows

$$\dot{e}_v = \mathfrak{F}'_0(P_1, P'_2) \tag{34}$$

$$\dot{e}'_{rs} = \mathfrak{F}'_1(P'_2)p'_{rs}, \tag{35}$$

where  $P'_2$  is the second invariant of the deviator, its first invariant being equal to zero.

We require that the viscous resistance vanishes with vanishing rate of strain and vice versa. Equ. (35) conforms to this requirement, but in order to make (34) conform to it we re-write it as follows

$$\dot{e}_v = P_1\mathfrak{F}'_{01}(P_1, P'_2) + P'_2\mathfrak{F}'_{02}(P_1, P'_2), \tag{36}$$

vanishing stress carrying with it vanishing invariants  $P_1$  and  $P'_2$ . Analysis of (35) and (36) will yield us then at least three rheological coefficients. We know up to now two ( $\mu$  and  $\mu_v$ ) and we therefore look forward with interest of meeting a third one.

7. (i) The first of our experiments, carried out in imagination, will consist in the application of an isotropic stress  $p$ .<sup>\*</sup> Deducting  $\kappa e_v$ , we get

$$p = p^* - \kappa e_v \tag{37}$$

<sup>8</sup>

$$p_{r\alpha}p_{\alpha s} = \sum_{\alpha=x}^{\alpha=z} p_{r\alpha}p_{\alpha s} = p_{rx}p_{xs} + p_{ry}p_{ys} + p_{rz}p_{zs}.$$

The first component of this tensor is accordingly

$$p_{xx}p_{xx} + p_{xy}p_{yx} + p_{xz}p_{zx} = p_{xx}^2 + p_{xy}^2 + p_{xz}^2$$

and similarly the others.

while  $p'_{r,s} = 0$ . Equ. (35) gives  $e'_{r,s} = 0$  and the rate of strain will have an isotropic component only. The first invariant of the stress  $p\delta_{r,s}$  is

$$P_1 = 3p \quad (38)$$

while the second invariant of the deviator of stress, there being no deviator, is equal to zero. This makes (36)

$$e'_r = 3p\mathfrak{F}'_{01}(p, 0). \quad (39)$$

$3\mathfrak{F}'_{01}(p, 0)$  is our first rheological coefficient. We rewrite it in the form

$$[3\mathfrak{F}'_{01}(p, 0)]^{-1} = \mu_r \quad (40)$$

and have

$$p = \mu_r e'_r \quad (41)$$

and (compare (30))<sup>9</sup>

$$p^* = \kappa e_r + \mu_r e'_r \quad (42)$$

(ii) For the second experiment we apply a tangential stress  $p_t$

$$p^*_{r,s} = \begin{vmatrix} 0 & p_t & 0 \\ p_t & 0 & 0 \\ 0 & 0 & 0 \end{vmatrix} \quad (43)$$

from which (compare (30))

$$p_{r,s} = \begin{vmatrix} -\kappa e_r & p_t & 0 \\ p_t & -\kappa e_r & 0 \\ 0 & 0 & -\kappa e_r \end{vmatrix}. \quad (44)$$

The first invariant of  $p_{r,s}$  is

$$P_1 = -3\kappa e_r \quad (45)$$

the second of its deviator (compare (43))

$$P'_2 = -p_t^2 \quad (46)$$

Equ. (35) gives us

$$e'_{r,s} = \mathfrak{F}'_1(p_t^2) \begin{vmatrix} 0 & p_t & 0 \\ p_t & 0 & 0 \\ 0 & 0 & 0 \end{vmatrix} \quad (47)$$

$\mathfrak{F}'_1(p_t^2)$  is our second rheological coefficient. We re-write it in the form

$$[\mathfrak{F}'_1(p_t^2)]^{-1} = 2\mu \quad (48)$$

<sup>9</sup>Such a rheological equation is mentioned by Reiner (1943).

and have

$$p_i = 2\mu e_i \quad (49)$$

in accordance with (4), or generally

$$p'_{r_s} = 2\mu e'_{r_s} . \quad (50)$$

Equation (36) on the other hand gives

$$e_v = -3\kappa e_r \mathfrak{F}'_{01}(-3\kappa e_r, -p_i^2) - p_i^2 \mathfrak{F}'_{02}(-3\kappa e_r, -p_i^2). \quad (51)$$

This is something new. While in accordance with the assumptions of classical hydrodynamics a simple shearing stress produces a continuously increasing simple shearing strain—and nothing else (compare (4)), we now found that generally *it will*, in addition, *produce a volume expansion*, the rate of which is given by (51). This volume expansion raises an elastic resistance which ultimately stops it. When this is the case, we have  $e_v = 0$  and

$$e_v = -(p_i^2/3\kappa) \mathfrak{F}'_{02}/\mathfrak{F}'_{01} . \quad (52)$$

This gives us a third coefficient<sup>10</sup>

$$\delta = -3\mathfrak{F}'_{01}/\mathfrak{F}'_{02} \quad (53)$$

so that

$$e_v = p_i^2/\kappa\delta \quad (54)$$

or generally<sup>11</sup>

$$e_v = -P'_2/\kappa\delta \quad (55)$$

We have found a third coefficient by mathematical analysis and after we found it, we may first be puzzled about what it should signify—but not for long. The phenomenon is well known. An increase of volume caused by a shearing stress is what Reynolds first described in wet sand as dilatancy. We may, therefore, call  $\delta$  the coefficient of dilatancy. The phenomenon has more recently been noted in very concentrated suspensions.<sup>12</sup> Classical hydrodynamics could not account for it. Our analysis has shown that it may generally be present in any liquid.

8. If  $\mathfrak{F}_0$  is a function of the third invariant of stress as well, (36) will become

$$e_v = P_1 \mathfrak{F}'_{01}(P_1, P'_2, P'_3) + P'_2 \mathfrak{F}'_{02}(P_1, P'_2 P'_3) + P'_3 \mathfrak{F}'_{03}(P_1, P'_2 P'_3) \quad (56)$$

This implies a fourth rheological coefficient  $\mathfrak{F}'_{03}$ . If we furthermore take up  $\mathfrak{F}_2$  as well, we see that a viscous liquid will in general be characterised by 5 coefficients (in addition to its elastic bulk modulus) of which, so far, we have defined three only. This does, of course, not mean that every viscous liquid will possess rheological properties requiring for their description finite values of all five coefficients. Sometimes one and

<sup>10</sup>This  $\delta$  is not to be confused with the  $\delta$  of  $\delta_{r_s}$ .

<sup>11</sup>One should keep in mind that the second invariant of a deviator is always negative.

<sup>12</sup>This is the place for a warning against an indiscriminate use of the term dilatancy. It is sometimes identified with an increase of viscosity with shear—the opposite of “structural” viscosity. This is misleading. The so-called snow-plough effect in suspensions is *not* dilatancy.



sometimes another property will be more prominent, depending upon the conditions under which the liquid is observed. We have had a striking example with the coefficient  $\mu_v$ . From the times of Stokes to recent times viscous liquids were observed under conditions which concealed its presence. Only when the rate of strain was very great, as in ultrasonic waves, or the material observed very viscous, as in the case of tar, did it become necessary to take this neglected coefficient into account. It may well be that particular conditions of observations will require the consideration of other coefficients, to be derived by the method shown in this paper. This may be the case with the liquids which Weissenberg (1946) has called "general" liquids. As was shown by Reiner (1945), the presence of  $\mathfrak{F}_2$  implies that a simple shearing stress causes not only a continuous shearing of the liquid but also a "continuous lengthening (or shortening) in the direction normal to the plane of shear". Reiner adds that such a phenomenon "has never been observed". Actually the phenomenon had at that time been observed by Weissenberg and collaborators but published only later.

**Conclusions.** Tensor analysis of the most general relation between stress and rate of deformation leads to five coefficients of viscosity of which three are the ordinary (shear) viscosity  $\mu$ , the coefficient of volume-viscosity  $\mu_v$ , and the coefficient of dilatancy  $\delta$ , while one coefficient may be connected with the "Weissenberg-effect". The Stokes-Navier equations of hydrodynamics in which  $\mu_v$  is neglected, can be used consistently only when the volume of the flowing liquid is constant.

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