The viscosity of a suspension of spheres

By A. D. MAUDE
City of Liverpool College of Technology

(Received 6 May 1959)

It is shown that in Stokes's flow the perturbation field, due to the addition of one more sphere to a shear flow of a fluid containing a number of non-interacting spheres, has the property that the total additional shearing force, acting on any plane normal to the direction of velocity change, is zero. However, the perturbation velocity, integrated over such a plane, takes a constant value, positive if the plane lies on one side of the sphere and negative if it lies on the other side. It follows that the effect of all the spheres is not to alter the shearing stress at all, but to reduce the mean shear by a factor $1-2{\cdot}5c$, where c is the concentration. This suggests that Einstein's viscosity law should be altered to $\eta = \eta_0/(1-2{\cdot}5c)$ when c is not small.

1. Introduction

If a number of spheres is added to a fluid the viscosity of the fluid appears to increase. Einstein (1906, 1911) has shown that if the density of the fluid is the same as that of the spheres, if the Reynolds number is sufficiently low for all inertial effects to be neglected, if the spheres are much smaller than the vessel containing the liquid, and if the spheres do not exert an attractive or a repulsive force between themselves, then for very low concentrations

$$\eta = \eta_0(1+2{\cdot}5c), \tag{1}$$

where η and η_0 are the viscosities of the suspension and fluid respectively, and c is the volume of spheres in unit volume of suspension. Many attempts have been made to extend this equation to higher powers of c for the case of equi-sized spheres. In most previous work, simplifying assumptions have had to be made, and the present work is an attempt to extend (1) without the use of such assumptions.

2. General considerations

The liquid is considered as being sheared in an idealized viscometer consisting of two infinite planes $y = 0$ and $y = H$, the latter being moved with a velocity U_x in the positive direction of x by a force F per unit area. The viscosity is then given by

$$\eta_0 = FH/U_x.$$

The introduction of spheres into this liquid will disturb the flow pattern, and it is convenient to consider the disturbance such that it vanishes at infinity. It is shown below that this implies that the average value of F is unaffected, although the average value of U_x is reduced. The perturbation velocity in the x-direction, v_x, will cause the velocity to vary over any plane y = const., but when the plane considered is far from the spheres, and when the perturbations of many spheres are superimposed, the variation over the plane will vanish. This variation over the planes $y = 0$ and $y = H$ is called a wall effect, and will be neglected in the present work.

The flow pattern may be calculated in theory by the method of successive reflexions. The first-order perturbation velocity caused by each sphere placed in the original flow is first calculated. The sum of all these perturbation velocities is then taken as the initial velocity field, and the second perturbation field due to each sphere placed in it is calculated. Third and higher-order perturbation velocity fields can be calculated similarly, leading to an infinite series of terms. It will be shown below that second and higher-order perturbation velocity fields cannot affect the viscosity of the suspension.

It appears to be difficult to prove the convergence of the above series, and, while it is expected to hold for low concentrations, there may well be a critical concentration above which it diverges.

Lamb (1945) gives a general solution of the linearized Navier Stokes equation as follows:

$$v_x = \frac{1}{\eta}\Sigma\left\{\frac{r^2}{2(2n+1)}\frac{\partial p_n}{\partial x} + \frac{nr^{2n+3}}{(n+1)(2n+1)(2n+3)}\frac{\partial}{\partial x}\frac{p_n}{r^{2n+1}}\right\} + \Sigma\left\{\frac{\partial \phi_n}{\partial x} + z\frac{\partial X_n}{\partial y} - y\frac{\partial X_n}{\partial z}\right\} \quad (2)$$

with two similar expressions for v_y and v_z obtained by cyclic permutation of x, y, z. Here $r^2 = x^2+y^2+z^2$, and p_n, ϕ_n, X_n are arbitrary solid harmonics of degree n, and the pressure $p = \Sigma p_n$.

If the origin is now taken at the centre of the added sphere, the restriction that the perturbation velocity and pressure must vanish at infinity implies that $n < 0$. For mathematical rigour, a proof is needed showing that it is always possible to satisfy the required boundary condition at the sphere surface; such a proof could easily be constructed using the method of § 7, but will not be given here.

3. Lemma 1

The force in the x-direction on any plane y = const. $\neq 0$ due to a perturbation velocity field is zero.

The force on a plane y = const. will be given by

$$\eta\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty}\left(\frac{\partial v_y}{\partial x} + \frac{\partial v_x}{\partial y}\right)dx\,dz;$$

and as this must be independent of y for there to be no accelerating force on an infinite slab between two planes y = const., only harmonics which give rise to v_x and v_y of degree -1 need be considered.

Neither ϕ nor X can give rise to terms in v of degree -1, and if the sphere moves so that it experiences no resultant force, the significant term in p_n—that is, p_{-2}—must be zero.

4. Lemma 2

The value of $\iint v_x\,dx\,dz$ in a perturbation field is constant for all positive and negative values of y.

From lemma 1 the total force in the x-direction on any plane $y = \text{const.}$ is zero. That is,

$$\eta\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty}\left(\frac{\partial v_y}{\partial x}+\frac{\partial v_x}{\partial y}\right)dx\,dz = 0.$$

But
$$\eta\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty}\frac{\partial v_y}{\partial x}\,dx\,dz = \eta\int_{z=-\infty}^{\infty}[v_y]_{x=-\infty}^{\infty}\,dz = 0.$$

Therefore
$$\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty}\frac{\partial v_x}{\partial y}\,dx\,dz = \frac{\partial}{\partial y}\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty}v_x\,dx\,dz = 0,$$

and
$$\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty}v_x\,dx\,dz = \text{const.} \tag{3}$$

5. Lemma 3

The average value of $(\partial v_y/\partial x+\partial v_x/\partial y)$ and of $\partial^2 p/\partial x\,\partial y$ over the surface of any sphere $r = \text{const.}$ is zero.

If expressions for $\partial v_x/\partial x$, $\partial v_y/\partial y$ and $\partial^2 p/\partial x\,\partial y$ are found from equations (2), and are integrated over the surface of a sphere, only a very limited number of values of n will be found to give a non-zero value, and in each case $n > 0$, so that this term will not occur in the perturbation velocity.

For example, consider the contribution of ϕ_n to the integral of $\partial v_x/\partial y$ over the surface of a sphere, which is

$$\int\frac{\partial^2\phi_n}{\partial x\,\partial y}\,dA.$$

As ϕ_n is a solid harmonic, $\partial^2\phi_n/\partial x\,\partial y$ is also a solid harmonic, and by the orthogonal properties of harmonic functions this integral will be zero unless $\partial^2\phi_n/\partial x\,\partial y$ is of degree 0 or -1. Therefore, $v_x = \partial\phi_n/\partial x$ will be of degree 1 (the degree zero not giving $\partial^2\phi_n/\partial x\,\partial y$ of degree -1). But as v_x must vanish at infinity this term cannot be included in the perturbation velocity field, so that no velocity due to the term ϕ_n can contribute to the integral.

A similar argument applied to each possible harmonic proves the proposition.

6. Expression of the significant parts of the integral (3) in terms of p_n, ϕ_n, X_n

As the integral (3) is a constant, only terms in v_x of degree -2 can contribute to it, so that we are restricted to p_{-3}, ϕ_{-1}, X_{-2}. Let

$$v_{x1} = -\frac{r^2}{10}\frac{\partial p_{-3}}{\partial x}+\frac{3}{30r^3}\frac{\partial}{\partial x}(r^5 p_{-3}) = \tfrac{1}{2}p_{-3}x,$$

$$v_{x2} = \frac{\partial\phi_{-1}}{\partial x},$$

$$v_{x3} = z\frac{\partial X_{-2}}{\partial y}-y\frac{\partial X_{-2}}{\partial z}.$$

If we express p_{-3}, ϕ_{-1}, X_{-2} in spherical polar co-ordinates, using the axis of y as the line $\theta = 0$ and the positive (y, z)-plane as the plane $\omega = 0$, considerations of symmetry show that the only terms which contribute to the integral and give a value which is different for positive and negative values of y are as follows. First, there is

$$p_{-3} = Ar^{-3}\cos\omega P_2^1(\cos\theta),$$

where $P_2^1(\cos\theta)$ denotes the associated Legendre polynomial $\cos\theta\sin\theta$. This gives

$$\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty} v_{x1}\,dx\,dz = \begin{cases} \pi A/3\eta & \text{for positive } y, \\ -\pi A/3\eta & \text{for negative } y. \end{cases}$$

Secondly, there is

$$X_{-2} = Br^{-2}\sin\omega\, P_1^1(\cos\omega),$$

which gives

$$\int_{z=-\infty}^{\infty}\int_{x=-\infty}^{\infty} v_{x1}\,dx\,dz = \begin{cases} -2\pi B & \text{for positive } y, \\ 2\pi B & \text{for negative } y. \end{cases}$$

It should be noted that terms which make the integral give the same value for positive and negative y may be neglected since they do not affect the rate of shear, and hence the viscosity, of the suspension.

7. Expression of A and B in terms of the original flow

To avoid the complication of the movement of the sphere we may impress a translational and a rotational velocity upon our axes, and thus upon the original flow, such that

$$[4\pi a^3 \operatorname{curl} \mathbf{v}]_{r=0} = 0,$$
$$[6\pi\eta a\mathbf{v} + \pi a^3 \operatorname{grad} p]_{r=0} = 0.$$

The bracket [ ] is used to indicate that the function is to be calculated for the original flow, and the suffix $r = 0$ to indicate that its value at the origin is to be taken. Here a is the radius of the sphere.

By Faxen's theorem (proved simply by Pérès in 1929), this implies the absence of movement or rotation of the sphere if no external force or couple is exerted on it. This choice of axes will not affect the perturbation velocity field.

Lamb gives

$$rv_r = \frac{1}{\eta}\Sigma\frac{nr^2}{2(2n+3)}p_n + \Sigma n\phi_n, \tag{4}$$

$$r\operatorname{curl}_r \mathbf{v} = \Sigma n(n+1)X_n. \tag{5}$$

The left-hand side of both these is determined by the original flow at $r = a$.

In both the original flow and in the perturbation flow

$$\operatorname{div}\mathbf{v} = \frac{1}{r^2}\frac{\partial}{\partial r}(r^2 v_r) + \frac{1}{r\sin\theta}\frac{\partial}{\partial\theta}(v_\theta\sin\theta) + \frac{1}{r\sin\theta}\frac{\partial v_\omega}{\partial\omega} = 0.$$

As v_θ and v_ω are equal and opposite in the original flow and in the perturbation flow at $r = a$, $\partial(r^2 v_r)/\partial r$ must also be equal and opposite in the two flows at $r = a$. But from (4) we have

$$\frac{\partial}{\partial r}(r^2 v_r) = \frac{1}{\eta}\Sigma\frac{n(n+3)r^2}{2(2n+3)}p_n + \Sigma n(n+1)\phi_n, \tag{6}$$

and the left-hand side of this is determined by the original flow at $r = a$.

If both sides of (4) and (5) are multiplied by $\cos\omega P_2^1(\cos\theta)$ and are integrated over the surface of a sphere, the orthogonal property of surface harmonics allows us to write

$$I_1 = -\int_{-1}^{1}\int_{0}^{2\pi}[rv_r]_{r=a}\cos\omega P_2^1(\cos\theta)\,d\omega\,d\cos\theta$$
$$= \frac{6\pi}{5}\frac{1}{\eta}\frac{A}{a} - \frac{36\pi}{5}\frac{C}{a^3},$$
$$I_2 = -\int_{-1}^{1}\int_{0}^{2\pi}\left[\frac{\partial}{\partial r}r^2v_r\right]_{r=a}\cos\omega\, P_2^1(\cos\theta)\,d\omega\,d\cos\theta$$
$$= \frac{72\pi}{5}\frac{C}{a^3},$$

where C is the constant of the harmonic

$$\phi_{-3} = Cr^{-3}\cos\omega P_2^1(\cos\theta).$$

Similarly,

$$I_3 = -\int_{-1}^{1}\int_{0}^{2\pi}[r\,\mathrm{curl}_r\,\mathbf{v}]_{r=a}\sin\omega P_1^1(\cos\theta)\,d\omega\,d\cos\theta$$
$$= \frac{8\pi}{3}\frac{B}{a^3}.$$

This gives

$$A = \frac{5}{12}\frac{\eta a}{\pi}(2I_1+I_2), \quad B = \frac{3}{8}\frac{a^3}{\pi}I_3.$$

As I_1, I_2 and I_3 are determined by the original flow, A and B may be found for an arbitrary original flow. A similar procedure could be made to yield the constant for each of the harmonics in the perturbation velocity field, and thus to solve the problem of a sphere in an arbitrary velocity field.

8. Expression of I_1 and I_3 in terms of the original flow at the origin

Let the original flow be expressed as a series of spherical harmonics in the manner of equation (2). As it contains no singularities within $r = a$, only harmonics with $n \geqslant 0$ are included.

Consider the harmonics

$$[p_2] = [A'r^2\cos\omega P_2^1(\cos\theta)],$$
$$[X_1] = [B'r\sin\omega P_1^1(\cos\theta)],$$
$$[\phi_2] = [C'r^2\cos\omega P_2^1(\cos\theta)].$$

Then in a similar manner to the last section we get

$$I_1 = -\frac{12\pi}{35}\frac{a^4}{\eta}A' - \frac{24\pi}{5}a^2C',$$
$$I_2 = -\frac{12\pi}{7}\frac{a^4}{\eta}A' - \frac{72\pi}{5}a^2C',$$
$$I_3 = -\frac{8\pi}{3}aB'.$$

Therefore

$$A = -a^5A' - 10a^3\eta C',$$

and

$$B = -aB'.$$

Consider now $[\partial^2 p/\partial x\,\partial y]_{r=0}$. Clearly, only harmonics of degree 2 can contribute to its value, as higher degrees will vanish when r is put as zero. Application of the double differentiation to all possible harmonics of degree 2 shows that

$$\left[\frac{\partial^2 p}{\partial x\,\partial y}\right]_{r=0} = A';$$

similarly,

$$\left[\frac{\partial v_y}{\partial x}+\frac{\partial v_x}{\partial y}\right]_{r=0} = 2C',$$

and

$$[\operatorname{curl}_z \mathbf{v}]_{r=0} = B',$$

which is zero by the hypothesis at the beginning of §7.

It follows that the difference between the values of $\iint v_x\,dx\,dz$ over each viscometer plate is altered by an amount

$$S = \tfrac{4}{3}\pi a^3\left\{\frac{5}{2}\left[\frac{\partial v_y}{\partial x}+\frac{\partial v_x}{\partial y}\right]_{r=0}+\frac{a^2}{2\eta}\left[\frac{\partial^2 p}{\partial x\,\partial y}\right]_{r=0}\right\}.$$

9. Viscosity of a suspension

If there are n spheres in unit volume, the value of U_x is reduced by an amount $nH\bar{S}$, where $\bar{S}$ is the average of S averaging over all the sphere centres. But from lemma 3, the perturbation velocity cannot affect the average value of S, so that it is determined solely by the original flow. That is,

$$\bar{S} = \frac{u_x}{H}\tfrac{5}{2}\cdot\tfrac{4}{3}\pi a^3.$$

Therefore U_x is reduced to

$$U_x(1-\tfrac{5}{2}n\,\tfrac{4}{3}\pi a^3) = U_x(1-2\cdot 5c).$$

The viscosity is thus

$$\eta = \eta_0/(1-2\cdot 5c). \tag{7}$$

10. Limitations of the theory

It has been implicity assumed in the last section that no ordering has taken place. That is, any one sphere will be situated in perturbation fields which arise evenly throughout space. In fact, some ordering will occur. It is well known that the probability of finding a second sphere centre a distance r from a given sphere centre is zero up to $r = 2a$, rises to a maximum and falls again, giving an oscillatory function of r of rapidly decreasing amplitude. The maxima become sharper as the concentration increases. However, except near to a wall, no ordering of direction occurs, and as the average values of $\partial v_y/\partial x+\partial v_x/\partial y$ and $\partial^2 p/\partial x\,\partial y$ are zero for any value of r, the ordering with respect to distance will not affect the result.

The possible divergence of the series mentioned in §2 might produce a change of the law at some concentration. Experiments by Higginbotham, Oliver & Ward (1958) show that the viscosity of a suspension of spheres obeys the law

$$\eta = \eta_0/(1-Kc)$$

up to a concentration of 28 %, above which concentration there is a marked departure from this relation. The value of K obtained was slightly less than 2·5. It is suggested that this change of the law at $c = 0{\cdot}28$ is due to the divergence of the series at higher concentrations.

Equation (7) above agrees with Einstein's result (equation (1)) if the square and higher powers of c are neglected. It also agrees with a theoretical expression derived by Kynch (1956) on what he calls a 'rigid envelope' model. Kynch's expression is

$$\eta = \eta_0(1 + 2{\cdot}5c + 6{\cdot}25c^2 + \ldots).$$

However, equation (7) does not agree with most of the other theoretical expressions which have been published.

The author wishes to express his thanks to his wife and to Mr H. Mulholland for help in preparation of the manuscript of this paper.

REFERENCES

Einstein, A. 1906 *Ann. Phys., Lpz.*, **19**, 289.
Einstein, A. 1911 *Ann. Phys., Lpz.*, **34**, 591.
Higginbotham, G. H., Oliver, D. R. & Ward, S. G. 1958 *Brit. J. Appl. Phys.* **9**, 372.
Kynch, G. J. 1956 *Proc. Roy. Soc.* A, **239**, 90.
Lamb, H. 1945 *Hydrodynamics*, 6th ed. New York: Dover Publications.
Pérès, J. 1929 *C.R. Acad. Sci., Paris*, **188**, 310.