On the Velocity of Steady Fall of Spherical Particles through Fluid Medium.

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1. Recent investigations[*] have called attention to the question as to how closely the rate of fall of particles through a fluid when very small approximates to that given by the law obtained by Stokes from hydrodynamical reasoning. This paper deals theoretically with two of the main sources of divergence from that law.

Firstly, taking the particles to be smooth spheres moving through a gas, it is shown that the deviation to be expected on account of the diameter of the particle being small, compared with the molecular free path, is extremely small for such particles as have been experimented with; and a modified formula is given which may be taken to hold approximately for lower pressures or particles of smaller dimensions. Secondly, an approximate treatment is made of the effect of the simultaneous presence of a large number of particles moving with the same velocity through the fluid, and it is found that the force required to maintain the motion of one of them depends not only on the diameter, but on the ratio of the diameter to the distance between the particles, and in such a way as to increase rapidly as this ratio increases beyond the value 0·1.

With the figures obtained by J. J. Thomson,[†] it is shown that these corrections together would imply an increase of not more than $0{\cdot}07 \times 10^{-10}$ in the charge on the gaseous ion.

[*] See the preliminary experiments of Zeleny with spores (British Association, 1909), and of Buller ('Nature,' April, 1909); also the later concordant results of Zeleny with spherical particles ('Nature,' Dec., 1909); also Millikan ('Physical Review,' Dec., 1909). Since this paper was prepared, Prof. Larmor has informed the writer that Prof. Zeleny is pursuing the subject of fall in gases at diminished pressures, with a view to throwing light on the nature of a molecular collision with a solid body, such as is required for the theory of the radiometer and other purposes (*cf.* Maxwell, 'Phil. Trans.,' 1879). For convenience of reference, a short calculation on this point has been added. The formula obtained in the first part of this paper agrees with the general conclusion that experiments of this kind cannot lead to a determination of molecular magnitudes, as at first sight seemed not impossible.

[†] J. J. Thomson, 'Phil. Mag.,' vol. 5, 1903.

On the Rate of Diffusion of a Cloud of Particles through a Gas.

2. Suppose that a cloud of particles of mass M at a concentration of N particles per unit volume is diffusing through a gas, the mean velocity of the particles being V_0, the mass of the gas molecule being m, and the number of molecules per unit volume being n. It is desired to find the mean force per particle required to maintain the velocity V_0.

It will be assumed in the first instance that the collisions are of the nature of impulses between smooth elastic spheres, and that a collision takes place between a molecule and a particle when the distance apart of their centres is a.

Consider the circumstances of collisions of this type with a single particle, the centre of the molecule at collision being within a small area

$$d\omega = a^2 \sin\theta \, d\theta \, d\phi$$

of the sphere of radius a surrounding the particle, (r, θ, ϕ) being spherical polar co-ordinates having the direction of V_0 as axis.

Let the velocity of the molecule be (u, v, w) and that of the particle (V_0+U, V, W).

The probability of molecular velocity lying within a range $du\,dv\,dw$ about (u, v, w) is

$$Ae^{-hm(u^2+v^2+w^2)}\,du\,dv\,dw,$$

where $$A(\pi/hm)^{\frac{3}{2}} = 1;$$

and that of the velocity of the particle lying within a range $dU\,dV\,dW$ is

$$Be^{-hM(U^2+V^2+W^2)}\,dU\,dV\,dW,$$

where $$B(\pi/hM)^{\frac{3}{2}} = 1.$$

In order that a collision may take place within the given limits in a given interval of time δt, the centre of molecule must at the beginning of it lie within a cylindrical volume of base $d\omega$ and height $(U+V_0\cos\theta-u)\,\delta t$, with the restriction that $(U+V_0\cos\theta-u)$ must be positive.

The magnitude of the impulse I is $(2Mm/M+m)(U+V_0\cos\theta-u)$.

Thus the probable impulse in the direction of V_0 on the particle per unit time is

$$\iiiint\!\!\int Ae^{-h(u^2+v^2+w^2)}\,I\cos\theta\,n\,(U+V_0\cos\theta-u)\,d\omega\,du\,dv\,dw,$$

the limits for v, w, θ, ϕ being unrestricted, but not so for u. This expression is equal to

$$An\frac{\pi}{hm}\,2\pi a^2\frac{2Mm}{M+m}\iint e^{-hmu^2}\,(U+V_0\cos\theta-u)^2\sin\theta\cos\theta\,du\,d\theta.$$

Multiplying by the probability of the given velocity (U, V, W) and integrating, the mean impulse per particle per unit time becomes

$$\left\{ \mathrm{AB}n\left(\frac{\pi}{h}\right)^2 4\pi \frac{a^2}{\mathrm{M}+m}\right\} \iiint e^{-h(mu^2+\mathrm{MU}^2)} (\mathrm{U}+\mathrm{V}_0\cos\theta-u)^2 \sin\theta\cos\theta\, d\theta\, du\, d\mathrm{U},$$

the limits for θ being 0 and π, U and u taking all values for which $u < \mathrm{U}+\mathrm{V}_0\cos\theta$.

The evaluation of the last integral is only possible on the assumption that V_0 is small compared with the mean relative velocity. Making this assumption, and calling the integral K, we have

$$\mathrm{K} = \iiint e^{-h[m\mathrm{M}\alpha^2+(m+\mathrm{M})^2\beta^2]/(m+\mathrm{M})} (\alpha+\mathrm{V}_0\cos\theta)^2 \sin\theta\cos\theta\, d\theta\, d\alpha\, d\beta,$$

where $\alpha = \mathrm{U}-u, \qquad \beta = (\mathrm{MU}+mu)/(\mathrm{M}+m)$.

Thus

$$\mathrm{K} = \left\{\frac{\pi}{h(\mathrm{M}+m)}\right\}^{\frac{1}{2}} \int_0^\pi \int_{-\mathrm{V}\cos\theta}^{\infty} e^{-hm\mathrm{M}\alpha^2/(\mathrm{M}+m)} (\alpha+\mathrm{V}_0\cos\theta)^2 \sin\theta\cos\theta\, d\theta\, d\alpha.$$

If the lower limit for α were 0, the value of this expression would be

$$\left\{\frac{\pi}{h(\mathrm{M}+m)}\right\}^{\frac{1}{2}} \int_0^\infty \tfrac{4}{3}\alpha\mathrm{V}_0 e^{-hm\mathrm{M}\alpha^2/(\mathrm{M}+m)}\, d\alpha = \tfrac{2}{3}\mathrm{V}_0 \sqrt{\{\pi(\mathrm{M}+m)/h^3m^2\mathrm{M}^2\}}.$$

The error in taking this value for K is

$$\left\{\frac{\pi}{h(\mathrm{M}+m)}\right\}^{\frac{1}{2}} \int_0^\pi \int_{-\mathrm{V}\cos\theta}^{0} e^{-hm\mathrm{M}\alpha^2/(\mathrm{M}+m)} (\alpha+\mathrm{V}_0\cos\theta)^2 \sin\theta\cos\theta\, d\theta\, d\alpha,$$

and is therefore of the order V_0/c times K, where c is the mean value of α, so that to the first order the above value may be taken for K.

Hence the mean impulse per particle per unit time is

$$\{\mathrm{AB}n(\pi/h)^2 4\pi\sigma^2/(\mathrm{M}+m)\}\, \tfrac{2}{3}\mathrm{V}_0 \sqrt{\{\pi(\mathrm{M}+m)/h^3m^2\mathrm{M}^2\}}$$
$$= \tfrac{8}{3}\sigma^2\mathrm{V}_0 n\sqrt{\{\pi\,\mathrm{M}m/(\mathrm{M}+m)^2\}}.*$$

This, then, is the force per unit particle required to maintain a drift V_0 of the particles relative to the gas. It represents the rate of transfer of momentum from a particle to the surrounding gas by reason of collisions.

It may be expected that, in the case where the particles are few in number compared with, and of the same order of magnitude as, the molecules, the mass velocity of the gas in the neighbourhood of a particle will be very slightly affected by the motion of the particle; but if the diameter of the particle becomes comparable with the mean free path of the molecule, it will become

* This expression is also obtained as a particular case of a general formula by Langevin ('Annales de Chimie et de Physique,' vol. 5, 1905, p. 266), and is applied by him to the consideration of the mobility of ions.

impossible to neglect the mass velocity produced in the gas by the motion of the particle. It will be necessary to substitute for V_0 in the above expression for the transfer of momentum, the difference between the mean velocity of the particle and the mass velocity of the gas.

It may be remarked that though a cloud of particles has been spoken of above, the result will hold equally well for the mean velocity of a single particle taken over a time sufficiently long to include a very large number of collisions. This is the case that must be considered in examining the deviation from Stokes' Law, which is demonstrated only for a single particle in an infinite fluid.

3. We may obtain now a modification of the law, taking into account the machinery by which the transfer of momentum from particle to fluid takes place. This is not considered in the ordinary hydrodynamical investigation. Instead, a kinematic assumption is made, namely, that the mass velocity in the neighbourhood of the particle is V. Let us assume, in place of this, that the mass velocity is kV. Then the rate of dissipation of momentum on the hydrodynamical theory is $6\pi\mu a k V$, a being used now for the effective radius of the particle, inasmuch as it is assumed to be large compared with the molecule. In the steady motion this diffusion of momentum must just balance the momentum communicated by the particle to the gas. Taking M large compared with m, this is, from the above formula,

$$\tfrac{8}{3}\sqrt{\left(\frac{\pi m}{h}\right)}\, a^2 n\,(V - kV).$$

Hence
$$6\pi\mu a k = \tfrac{8}{3}\sqrt{\left(\frac{\pi m}{h}\right)}\, a^2 n\,(V - kV),$$

or
$$k = \frac{4\,an\,(\pi m/h)^{\frac{1}{2}}}{4\,an\,(\pi m/h)^{\frac{1}{2}} + \rho\pi\mu} = \left(1 + \frac{3\,l}{4\,a}\sqrt{\frac{3\,\pi}{2}}\right)^{-1},$$

where l* is the mean free path of the molecules in the air. Hence the force required to maintain the velocity V is

$$X = 6\,\pi\mu a k V = 6\,\pi\mu a V\,(1 + 1{\cdot}63\,l/a)^{-1};$$

that is, the limiting velocity of a falling particle will exceed that given by the ordinary formula in the ratio of $(1 + 1{\cdot}63l/a)$ to unity. This ratio for a given gas should therefore differ from unity by an amount which varies inversely as the density.

For air at ordinary pressure and temperature l may be taken to be about $0{\cdot}9 \times 10^{-5}$, so that the limiting velocity would, on the present calculation, be double the usual value for a particle of radius about $1{\cdot}5 \times 10^{-5}$. For particles

* l is the mean free path obtained from the viscosity by the formula $\mu = \frac{1}{3}\rho\bar{c}l$, not the value corrected for persistence of velocity (*vide* Jeans, 'Kinetic Theory of Gases,' p. 250).

of the magnitude of the smallest spores in Zeleny's earlier experiments the ratio would be about 1·07.

The nature of the collision between molecule and particle has, so far, been assumed to be that which occurs between smooth elastic spheres. If this is replaced by supposing that each molecule became entangled in the particle, so that on re-emission there was no correlation between its velocity and that with which it collided, a similar calculation can be easily made. The impulses by which the molecules are thrown out from the particle have no resultant, if all directions of re-emission are supposed equally probable. The impulse at collision in the direction of the velocity V_0 is

$$\frac{Mm}{M+m}\{V_0+(U-u)\cos\theta+(V-v)\sin\theta\}.$$

If the integration be effected as before, the mean impulse is found to bear to that previously found the ratio of $\int_0^\pi(1+\cos^2\theta)\sin\theta\,d\theta$ to $\int_0^\pi 2\cos^2\theta\sin\theta\,d\theta$, that is, it is exactly double.

If, now, it is assumed, following Maxwell, that a fraction "f" of the colliding molecules experience collisions of the earlier type, and the remainder of the latter type, the equation giving the limiting velocity becomes

$$X\left\{1+\frac{1{\cdot}63\,l/a}{f+2(1-f)}\right\} = 6\,\pi\mu a V.$$

Maxwell (*loc. cit.*) deduces from experiments of Kundt and Warburg that for impacts of air molecules on glass the fraction f is approximately half. This would reduce the divergence from Stokes' Law to two-thirds of that obtained by putting $f = 1$. If no molecules experience perfect reflexion, f would be zero, and the divergence would be reduced to one-half. If it be assumed that for the majority of material particles f lies between the values 0 and $\frac{1}{2}$, the result is that Stokes' formula will yield a velocity equal to half the actual for particles whose radii lie between $0{\cdot}75\times10^{-5}$ and 1×10^{-5}, while for particles of diameter 2×10^{-4}, such as those used in Zeleny's earlier experiments, it will yield a velocity between 1·05 and 1·036 too small. Experiments at lower pressures should show greater deviations, and may throw light on the value of f.

On the Mutual Influence of the Particles in a Cloud.

4. A possible source of deviation from Stokes' Law for a cloud of particles of dimensions for which the hydrodynamical condition of no surface slipping is satisfied, is the mutual influence of the particles, and the effect of the boundaries. The exact solution of this problem is clearly beyond analysis. But an approximate method of attack suggests itself, based on the fact

that if a cloud of particles is uniformly distributed throughout an enclosed volume of fluid and are moving with equal parallel velocities, the drift of the fluid with each particle in the direction of its motion in the immediate vicinity of the particle must be compensated by a drift in the opposite direction between the particles. In fact, if the distribution is uniform the mean flux of fluid across any area is zero if the fluid is contained within fixed boundaries. In Stokes' problem this condition is absent.

The method employed to obtain an idea of the magnitude of the influence of this consideration is to imagine that each particle is effectively limited to moving within a concentric spherical mass of fluid whose boundary is fixed. If, for instance, supposing the particles to be of radius a, and surrounded by concentric spheres of radius b, the arrangement of particles being such that these latter spheres are packed in the closest possible arrangement, the fluid in the spaces between them being supposed occupied by fluid at rest, the conditions of the problem are to some extent realised; but the velocity of each particle in its own sphere will be somewhat less than the actual limiting velocity owing to the restriction imposed on the fluid between the spheres. We may therefore expect by this method to obtain a lower limit for the velocity sought in terms of the radius of each particle and the number of particles per unit volume.

The problem is therefore to be solved of the drag due to viscosity on a particle moving with velocity V at the centre of a spherical envelope. This can be effected exactly, the method adopted being that given by Lamb* for the solution of Stokes' problem.

Taking the axis of x in the direction of motion, the velocity at a point can be expressed in terms of spherical harmonics of the first order.

Putting $p_1 = \alpha x, \quad p_{-2} = \mathrm{A}x/r^3,$
$\phi_1 = \beta x, \quad \phi_{-2} = \mathrm{B}x/r^3,$

and using the formula of type

$$u = \frac{1}{\mu}\Sigma\left\{\frac{r^2}{2(2n+1)}\frac{\partial p_n}{\partial x} + \frac{nr^{2n+3}}{(n+1)(2n+1)(2n+3)}\frac{\partial}{\partial x}\frac{p_n}{r^{2n+1}}\right\} + \Sigma\frac{\partial \phi_n}{\partial x},$$

we obtain

$$u = \frac{\alpha}{6\mu}\left\{r^2 + \frac{r^2-3x^2}{5}\right\} + \frac{2\mathrm{A}}{3\mu}\left\{\frac{1}{r} - \frac{r^2-3x^2}{4r^3}\right\} + \mathrm{B}\frac{r^2-3x^2}{r^5} + \beta,$$

$$v = -\frac{\alpha}{10\mu}xy \qquad + \frac{\mathrm{A}}{2\mu}\frac{xy}{r^3} \qquad - 3\mathrm{B}\frac{xy}{r^5},$$

$$w = -\frac{\alpha}{10\mu}xz \qquad + \frac{\mathrm{A}}{2\mu}\frac{xz}{r^3} \qquad - 3\mathrm{B}\frac{xz}{r^5}.$$

* 'Hydrodynamics,' 3rd edit., § 324–5.

Now at the surface of the particle where $r = a$,

$$u = \mathrm{V}, \quad v = 0, \quad w = 0,$$

and at the sphere $r = b$ $\qquad u = v = w = 0.$

These conditions give the equations

$$\frac{a^2\alpha}{6\mu}+\frac{2\,\mathrm{A}}{3\,\mu a}+\beta = \mathrm{V}, \qquad -\frac{\alpha}{10\,\mu}+\frac{\mathrm{A}}{2\,\mu a^3}-\frac{3\,\mathrm{B}}{a^5} = 0,$$

$$\frac{b^2\alpha}{6\mu}+\frac{2\,\mathrm{A}}{3\,\mu b}+\beta = 0, \qquad -\frac{\alpha}{10\,\mu}+\frac{\mathrm{A}}{2\,\mu b^3}-\frac{3\,\mathrm{B}}{b^5} = 0.$$

The resultant traction on the sphere is found to be, exactly as in the simpler problem,*

$$\mathrm{X} = -4\,\pi\mathrm{A}.$$

The value of A obtained from the four equations above gives

$$\mathrm{X} = 6\,\pi\mu a\mathrm{V}\,\frac{4\,b\,(b^5-a^5)}{(b-a)^2\,(4\,b^4-b^3a-6\,b^2a^2-ba^3-4\,a^4)},$$

a result which reduces at once to Stokes' law when b/a is large.

Expanded in powers of (a/b), the result is

$$\mathrm{X} = 6\,\pi\mu a\mathrm{V}\left(1+\frac{9}{4}\frac{a}{b}+\frac{81}{16}\frac{a^2}{b^2}+\ldots\right).$$

It was said above that the value so found is a lower limit for the actual velocity under a given force, owing to the conditions imposed being possibly more rigid than the problem demanded.

5. If now round each particle be described a sphere of radius b', such that the new spheres overlap, but only by so much that they just include the whole of the fluid, each particle being again supposed to move in its own sphere, a new value for the velocity will be obtained. The arrangement of particles being as before, b' will be the distance from a corner to the centroid of a tetrahedron whose edge is $2\,b$, so that $b' = b\sqrt{(3/2)}$.

It is not clear, however, that in this way an upper limit to the velocity of the particles is obtained, though the following considerations point in that direction.

In either case the argument is that if the velocity system for the fluid be obtained when the fluid within one of the spheres is set in motion by the enclosed particle, the external fluid being at rest; and if the velocity systems thus obtained for all the particles are superposed, the resultant velocity system will approximate to the actual in the sense that in the neighbourhood of each particle the conditions imposed by the continuity of

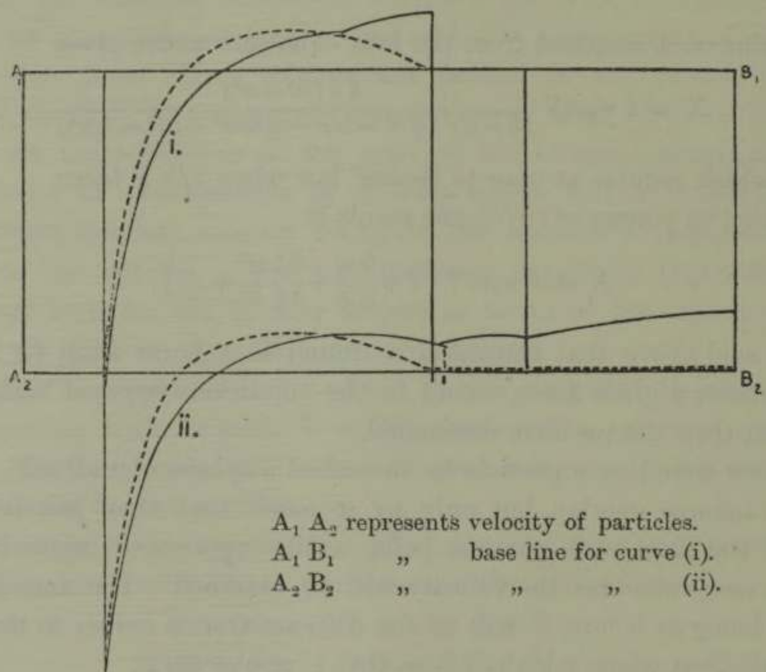
* *Loc. cit.*, p. 552, equations (5) and (13).

velocity is satisfied, and that the hydrodynamical equations, being linear, are also satisfied.

Thus, in the former case, the region external to all the spheres of radius b is kept at rest; while, in the latter, the motion in a region internal to two or more spheres is compounded of the contributions from the motions in the separate spheres.

But the difficulty is thus introduced, that, at the surfaces of the auxiliary spheres, discontinuity occurs in the velocity gradient. In the actual problem the discontinuity is smoothed out.

The diagram below shows, for $b/a = 5$, the variation of the vertical velocity of the fluid as obtained above: (i) along a line through the centres of two of



$A_1\ A_2$ represents velocity of particles.
$A_1\ B_1$ " base line for curve (i).
$A_2\ B_2$ " " " (ii).

the spheres supposed at distance $2b$ apart in a horizontal line; (ii) along a median of an equilateral triangle of side $2b$, there being supposed three spheres with their centres at the corners. The continuous line refers to the velocity found by the use of the larger spheres, the broken line to that given by the smaller. Comparison of these curves and of others not given here indicates fairly clearly that the former is more likely to be near to the true facts. Accordingly the values are tabulated below of the ratio of the forces required to maintain a velocity V according to the method adopted above, using the larger sphere, and, according to Stokes' Law, for different values of b'/a; the second row of the table gives the corresponding value of b/a.

b'/a	2	3	4	5	6	7	8	9	10	20	30	∞
b/a	1·63	2·45	3·26	4·08	4·90	5·71	6·50	7·35	8·16	16·3	24·5	∞
$X/6\pi\mu aV$	7·29	3·02	2·10	1·76	1·57	1·46	1·38	1·32	1·28	1·14	1·08	1

6. Assuming, now, that the modified laws obtained above are correct, we may see what correction they imply as necessary to J. J. Thomson's calculation of the charge on the gaseous ion.

The figures given by him fall into four groups corresponding to values of the charge on the ion—

$$10^{10}e = 3{\cdot}2,\ 3{\cdot}50,\ 3{\cdot}45,\ 3{\cdot}39,$$

respectively. Taking the figures given for the number of drops per cubic centimetre, we are able to calculate the corresponding values of b'/a, and thence the correction to be made as in § 5. The result is that e must be increased by 6·2, 7·4, 6, 5·9 per cent. in the four cases respectively. For the calculated values of the radii of the drops, however, the correction on account of the first cause (§ 3) amounts to about 4 per cent. and is in the opposite sense, and therefore corresponds to a diminution of 4 per cent. in e.

On account of both causes we may say that e is to be increased by about 2 per cent., that is by about $0{\cdot}07 \times 10^{-10}$.

It is clear, therefore, that neither of the causes discussed accounts for more than a small fraction of the discrepancy between the older and the more recent estimates.*

* *Vide* Rutherford, Presidential Address to British Association, Section A, 1909; also Millikan, 'Physical Review,' Dec., 1909.