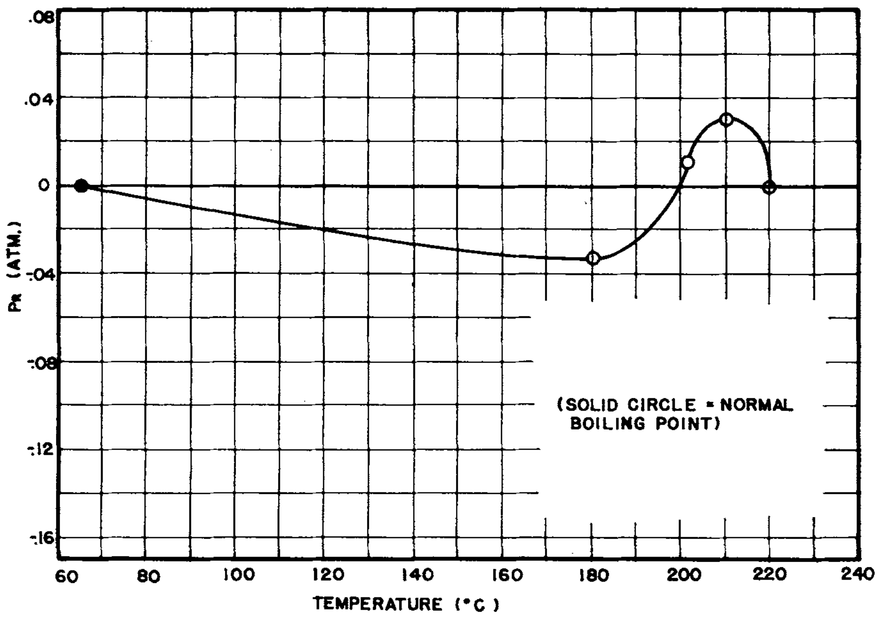

Fig. 1. Vapor pressure residual P_R vs. temperature.

edict-Webb-Ruben equation of state is 0.64%.

The critical temperature is estimated to be 228.5° ± 0.5°C. and the critical pressure 35.4 ± 0.5 atm.

ACKNOWLEDGMENT

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The Motion of Two Spheres Following Each Other in a Viscous Fluid

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The purpose of this investigation was to determine the interaction effect of one spherical particle upon another when both are falling in a viscous fluid. The velocities of two identical spheres, falling along the axis of a cylinder in a direction parallel to their line of centers, were measured experimentally as a function of the center-to-center distance between them at very low Reynolds numbers. The experimental results compared very well with theoretical studies found in the literature which predicted that two spheres will fall faster than one sphere.

At Reynolds numbers greater than 0.25 the influence of inertial effects were studied for one and two spheres. The experimental results qualitatively confirmed the Oseen equations. A definite attraction between two spheres falling one above the other was observed; the inertial forces acted to slow down the lower sphere without affecting the upper one.

The behavior of spherical particles settling in a viscous fluid is of fundamental importance in solving problems involving sedimentation, flow through packed beds, and fluidization. However both mathematical analysis and experimental observation are extremely difficult when dealing with assemblages of particles because of the many boundary conditions and interaction effects encountered. In addition to these complications, as soon as the particle Reynolds number becomes as high as 0.25, the inertial terms can no longer be neglected in the solution of the Navier-Stokes equations of motion.

For these reasons a logical start in attempting to solve problems involving many particles is to carry out a complete study of the important variables affecting the motion of one particle at very low Reynolds numbers. Then these observations can be used to study the motion of two particles and so on, until eventually enough theoretical and experimental information is available to predict the motion of the complex particle systems actually encountered in practice. The same technique can be utilized in studying the inertial effects which become important at higher Reynolds numbers.

The purpose of the investigation described in this paper was to continue to add to the information available concerning the motion of two particles settling one above the other in a viscous fluid. As an idealization of the numerous particle shapes conceivable, identical, smooth, and rigid spheres were chosen to work with.

Experimental measurements were made at low Reynolds numbers (much less than 0.25) of two spheres settling along the axis of a cylindrical tube in a direction parallel to their line of centers. This was done to determine how well the theoretical equations of motion, and the pertinent correction factors which take into account the boundary and interaction effects, could predict the actual motion. The influence of inertial effects at higher Reynolds numbers on the motion of two spheres was also determined experimentally and compared with the corresponding effects on a single sphere. A qualitative analysis of the motion of three spheres will also be discussed.

DESCRIPTION OF APPARATUS

The apparatus used for taking velocity measurements consisted of a cylindrical Pyrex glass column 32 in. long with an

I.D. of 5 11/32 in. (Figure 1). The column was equipped with a closed nipple set between two 1¼-in. gate valves which was mounted directly below the axis of the cylinder. This equipment was used to recover the spheres and also, when necessary, to empty the column. A dropping mechanism, consisting of a thin-walled brass bushing slightly larger in diameter than the spheres which were dropped, was carefully mounted on the top of the column so that the axis of the bushing coincided with the axis of the cylindrical column. This allowed the spheres to fall freely into the fluid.

Two thermometers were inserted into the glass column 8 in. from the top and bottom and ¾ in. from the glass wall so that the temperature of the fluid might be read at both of these points. Since it was impractical to surround the column by a constant-temperature bath, the experiments were performed in a relatively constant-temperature room which was maintained at a temperature between 24.5° and 26.5°C. by a Fenwall thermoswitch connected through a relay to a ¾-ton room air conditioner. The temperature of the room never varied more than ±½°C. in any 24 hr. period; temperature gradients across the room were minimized by the use of an electric fan to circulate the air.

To obtain different operating conditions two polyalkylene glycol types of fluids having different viscosities were used in the experiments. The fluids show a comparatively small change in viscosity with temperature and are also extremely stable. They have a negligible vapor pressure, low hygroscopicity, and a low specific heat; they are also water soluble which is very helpful in cleaning the apparatus. The viscosity and density of the two fluids were determined over a temperature range of 23° to 28°C. with a calibrated Ostwald type of viscometer and pycnometer.

The spheres which were used in the experiments consisted of plastic balls approximately ¼ in. in diameter. The balls, made of marbelette, Lucite, and nylon, were perfectly smooth and rigid and completely insoluble in the fluids. The density of the spheres was measured to five significant figures by the use of a constant-volume pycnometer; the diameters of the spheres were measured with a micrometer.

Some of the pertinent properties of the spheres and fluids that were used are listed below.

Spheres

Material	Color	Diameter, in.	Density, g./cc.
Nylon	White	0.250	1.1586
Lucite	Red	0.234	1.1919
Marbelette	Black	0.241	1.3249

Fluids

Ucon fluid	Viscosity at 25°C., centipoises	Density at 25°C., g./cc.
1	1,337	1.050
2	242	1.043

EXPERIMENTAL METHOD

The experimental runs consisted of taking velocity measurements of one sphere falling along the axis of the cylindrical glass column and of two spheres falling along the axis, one above the other.

For one sphere falling alone the time required for the sphere to fall between two strips of tape 12 in. apart was recorded with a stop watch which could be read to 0.1 sec. Before each run the temperature of both thermometers was read to 0.1°C. and the average value recorded. This was justified because an error as large as 0.2°C. in the average temperature would amount to an error of only 1% in the viscosity of the fluid and a completely negligible error in the density. No runs were made, however, if the temperature gradient was more than 0.4°C.

The 12-in. length of fall was located in the central section of the 32 in.-long glass column so that the possibility of errors due to unsteady motion was eliminated. This also minimized any errors due to end effects.

For two spheres falling one above the other, at very low Reynolds numbers ($N_{Re} << 0.25$), the velocities of the two spheres were found to be equal and were measured by recording the time (to 0.1 sec.) required for the bottom sphere to fall 12 in. The center-to-center distance between the spheres was taken both at the top and bottom of the 12-in. length of fall by measuring the time difference between the two spheres as they both crossed each of the tape strips; another stop watch which could be read to 0.1 sec. was used for this purpose. The observed time between the spheres at the top and bottom of the fall was converted to distance units by multiplying the time by the velocity of the spheres. The two results were averaged; if they differed by more than 1% of the total 12-in. length of fall, the run was discarded.

At higher Reynolds numbers ($N_{Re} > 0.25$) the two spheres falling one above the other were invariably observed to approach each other. Since the distance between the two spheres kept getting smaller during the run, the velocities of the spheres were neither equal nor constant. The experimental procedure in this case consisted only of measuring the velocity of each of the spheres over the 12-in. length of fall; two stop watches were used.

THE MOTION OF A SINGLE SPHERE AT $N_{Re} << 0.25$

The velocity of a single sphere falling along the axis of the cylindrical column in the high-viscosity fluid was measured at slightly different fluid temperatures (between 24° and 27°C.). The velocity measurements were taken for each of the three different density spheres and were found to be reproducible. Since the sphere-diameter–to–cylinder-diameter ratio for a ¼-in. sphere falling in a column having an inside diameter of 5 11/32 in. is less than 0.1, Stokes's Law (*10*) combined with the Faxen (*2*) correction factor, which accounts for the effect of the cylindrical boundary, can be used to predict the experimentally measured velocities:

$$U_{predicted} = \frac{(\rho_S - \rho) g\, d^2}{18 \mu K_I} \qquad (1)$$

$$K_I = \frac{1}{1 - 2.105 \dfrac{d}{D}} \qquad (2)$$

The predicted values of the velocity at each temperature were calculated by the use of Equation (1); since the Reynolds numbers were all much less than 0.25, the effect of inertial forces was neglected. The average error between the calculated and measured velocities was less than 1%; these results show that inertial and end effects were indeed negligible. The single-sphere velocity measurements were plotted as a function of the temperature of the fluid.

THE MOTION OF TWO SPHERES AT $N_{Re} << 0.25$

If two spheres are falling in an infinite viscous fluid, a mutual interaction effect between them is observed. Each sphere will have a tendency to move fluid down along with it, and therefore the velocity of each sphere will be increased owing to the motion of the other. For two spheres falling one above the other in an infinite fluid at very low Reynolds numbers the drag force on each sphere can be expressed by

$$W = \lambda 3 \pi \mu\, dU \qquad (3)$$

Equation (3) is Stokes's Law modified by a correction factor to account for the interaction effect.

Smoluchowski (*8*) in a theoretical study showed that

$$\lambda = 1 - \frac{3}{4}\frac{d}{l} + \left(\frac{3}{4}\frac{d}{l}\right)^2 \qquad (4)$$

Equation (4) is an approximate expression and can only be applied to two equal-sized spheres falling with a center to center distance greater than three diameters. The mutual interaction between two spheres of different size has recently been reported in a series of studies by Andersson (*1*) based on the creeping-motion equations.

A more rigorous solution of the problem of two spheres falling with equal and constant velocities parallel to their line of centers at negligible Reynolds numbers is that given by Stimson and Jeffrey (*9*). Mathematically their expression for λ is

$$\lambda = \frac{4}{3} \sin h\alpha \sum_{n=1}^{n=\infty} \frac{n(n+1)}{(2n-1)(2n+3)} \cdot \left[\frac{1 - 4 \sin h^2(n+\frac{1}{2})\alpha - (2n+1)^2 \sin h^2 \alpha}{2 \sin h(2n+1)\alpha + 2n + 1 \sin h^2 \alpha} \right] \quad (5)$$

where $\cosh \alpha = l/2a = l/d$. A numerical evaluation of this function is given below for different values of l/d

α	l/d	λ
0.5	1.128	0.663
1.0	1.543	0.702
1.5	2.352	0.768
2.0	3.762	0.836
2.5	6.132	0.892
3.0	10.068	0.931
∞	∞	1.00

Faxen (*2*) calculated the value of λ for the limiting case of the two spheres touching (that is, $\alpha = 0$, $l/d = 1$) and obtained $\lambda = 0.645$. This predicts that a doublet consisting of two equal spheres touching one above the other would fall 55% faster in an infinite fluid than if one sphere were falling alone.

The relationships of Ladenburg (*5*) and Faxen (*2*) which were developed for the effect of a cylindrical boundary on the motion of a single sphere can be directly applied to the study of two spheres. If two spheres *A* and *B* are falling one above the other along the axis of a cylinder, the drag on sphere *A* will be the result of adding four velocity fields. These include the initial undisturbed velocity field U_{A_1} and the first reflection of this field from the cylinder wall U_{A_2}; these would be the only contributions to the velocity field if sphere *A* were falling alone. In addition sphere *B* will reflect the motion of sphere *A* in two ways, by a direct reflection of its own velocity field U_{B_1} (interaction effect) and by the reflection of this velocity field at the cylinder wall U_{B_2}. U_{A_1} and U_{A_2} are given by Stokes's Law combined with the Faxen correction factor. Happel (*4*) has shown that U_{B_2} can be represented by the following series expansion is sphere *B* is close to sphere *A* and $a/R = d/D$ is less than 0.1:

$$U_{B_2} = \frac{Ua}{R}\left(2.105 - 1.139 \frac{l^2}{R^2} + 0.595 \frac{l^4}{R^4} + \ldots \right) \quad (6)$$

To convert the observed velocity for two spheres falling in a cylinder to that which would be obtained if they fell in an infinite medium, it is necessary to multiply the observed velocity by a correction factor obtained by adding unity to U_{A_2} and U_{B_2}. This factor K_{II} is found to be

$$K_{II} = 1 + 4.21 \frac{a}{R} - 1.139 \frac{al^2}{R^3} + 0.595 \frac{al^4}{R^5} + \ldots \quad (7)$$

If l/R is very small, Equation (7) reduces to

$$K_{II} = 1 + 4.21 \frac{a}{R} \quad (8)$$

and the coefficient of $a/R = d/D$ is just twice the value of the coefficient for a single sphere. If l/R is large, Equation (6) will not converge, and a numerical evaluation of U_{B_2} is required.

Equation (7) has been used to evaluate the boundary correction factor for values of $l/R < 0.3$. For the special case of $l/R = 1$, K_{II} has been evaluated numerically and was found equal to 3.45 a/R. The values of K_{II} were calculated for the three different density spheres which were used and plotted for convenience as a function of l.

If l/d is small (spheres very close together), it would be desirable to consider more than one reflection between them to completely evaluate their interaction. When the spheres are touching, for example, the constant in Equation (8) will be smaller than 4.21. This would tend to raise the values of λ determined experimentally for small values of l/d. If the drag force exerted on each sphere when two spheres fall in an infinite viscous fluid is compared with that exerted on a single sphere, it is clear that

$$\lambda = \frac{U_{I\infty}}{U_{II\infty}} \quad (9)$$

To correct the experimental velocities for the effect of the cylindrical boundary, the factors K_I and K_{II} are used. If U_{II} is the experimentally measured velocity of the two spheres at a known fluid temperature and U_I is the velocity of a single sphere at the same temperature, then

$$\lambda = \frac{K_I U_I}{K_{II} U_{II}} \quad (10)$$

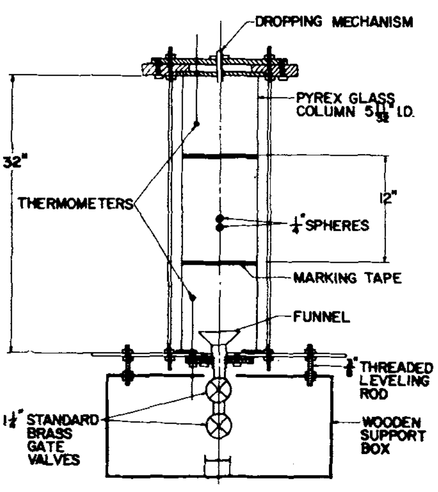

Fig. 1. Experimental apparatus.

The values of λ were calculated from Equation (10) by measuring the velocity of the two spheres. For each different run with two spheres the corresponding velocity of a single sphere at the same average temperature was obtained from the plots mentioned above, K_I was calculated by means of Equation (2), and K_{II} was obtained from the plots of K_{II} vs. l.

The experimental values of λ obtained for the three different-density ¼-in. spheres were plotted against the l/d ratio in Figure 2; the theoretical curve predicted by Stimson and Jeffrey has also been included. Figure 2 shows an excellent agreement between the observed values and the theoretical curve in the entire l/d range which was investigated. The Reynolds number range for these runs was between 0.008 and 0.03; at these low Reynolds numbers no attraction between the two spheres was observed, and inertial effects were taken to be negligible.

THE EFFECT OF INERTIAL FORCES ON THE MOTION OF ONE AND TWO SPHERES

In order to study some of the important effects on the motion of one and two spheres which are caused by inertial forces the less viscous fluid was used as the fluid medium. This resulted in the spheres falling at greater velocities and higher Reynolds numbers.

The experimentally measured velocities of the ¼-in. spheres were again compared to the predicted values given by Equation (1). The deviations were appreciably larger, varying from about 2% at a Reynolds number of 0.25 to 3% at a Reynolds number of 0.5. To obtain even higher Reynolds numbers spheres ½ in. in diameter were dropped, and velocity readings were taken. The corresponding velocities calculated from Equation (1) deviated by as much as 17% at a Reynolds number of 3.5. Fayon's (*3*) experiments have shown that the boundary correction factor K_I remains relatively constant up to Reynolds numbers of 20; therefore in this range Stokes's Law no longer applies for a single sphere in an infinite medium.

Oseen (*6*) included some of the quadratic inertial terms in his solution of the Navier-Stokes equations for a single sphere falling in an infinite fluid and derived theoretically that the drag force can be approximated at higher

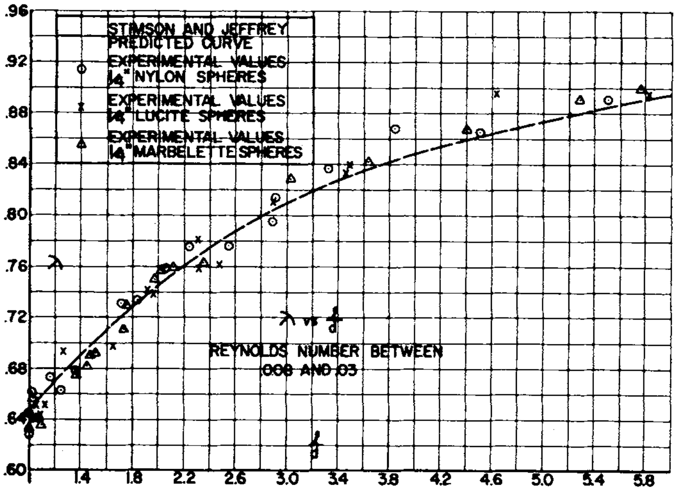

Fig. 2. Interaction effect between two spheres.

Reynolds numbers by a modification of Stokes's Law:

$$W = 3\pi\mu\, dU(1 + 3/16\, N_{Re}) \qquad (11)$$

Oseen also extended Smoluchowski's solution [Equation (4)] for the motion of two spheres so that it might be applied to higher Reynolds numbers. Oseen's equations for one sphere falling above another can be approximated when the spheres are not too far apart by

$$W_1 = 3\pi\mu\, dU \left(1 - 3/4\frac{d}{l} + 3/8\, N_{Re}\right) \qquad (12)$$

$$W_2 = 3\pi\mu\, dU\left(1 - 3/4\frac{d}{l}\right) \qquad (13)$$

These equations point out that the bottom sphere is slowed more than a single sphere owing to inertial forces [compare Equation (12) with Equation (11)] and that the slowing is a function of Reynolds number only. The top sphere, however, is not at all affected and will fall according to the Smoluchowski theory. Since the drag on two equal spheres must be equal, the velocity of the top sphere will be greater than that of the bottom sphere, and they will tend to move toward each other.

If the two spheres fall side by side with the same velocity, Oseen's equations show that the inertial force on the right sphere is

$$F = 3\pi\mu\, dU(3/32\, N_{Re}) \qquad (14)$$

and acts to the right; the corresponding force on the left sphere is equal in magnitude but opposite in direction, and the two spheres will tend to move away from each other, a phenomenon exactly opposite to that predicted for two spheres following each other.

If Oseen's equation for a single sphere is rewritten in terms of the drag coefficient and is also modified to take into account the effect of a finite cylindrical boundary around the fluid, Equation (11) becomes

$$C_D = \frac{24}{Re} K_I(1 + 3/16\, Re) \qquad (15)$$

The measured velocity data for a single sphere did not correlate with Equation (15), and in order to obtain a correlation the constant 3/16 was replaced by an empirical constant so that Equation (15) read

$$C_D = \frac{24}{N_{Re}} K_I(1 + C_I N_{Re}) \qquad (15a)$$

C_I was calculated from Equation (15a) in the Reynolds number range of 0.25 to 3.5 by measuring the drag coefficient and the Reynolds number; the correction factor was assumed to remain constant. At Reynolds numbers greater than 0.5 C_I was found approximately equal to 0.05; the results at the lower Reynolds numbers were not too significant, since the deviations from the Stokes-Faxen linear approximation were less than 3%.

Preliminary experiments with two spheres falling one above the other showed, as predicted by Equations (12) and (13), that they invariably will come together during the length of fall. However, it was possible to establish the effect of inertia by observing their relative velocities, thus making a wall correction factor unnecessary. From Oseen's theory the following approximation may be developed (7):

$$U_2 - U_1 \cong 3/8\, \Psi N_{Re} \qquad (16)$$

where

$$\Psi = \frac{(\rho_S - \rho) g\, d^2}{18\mu} \qquad (17)$$

and $U_2 - U_1$ is the difference in velocity between the two spheres.

The constant, 3/8, in Equation (16) was again replaced by an empirical constant so that a correlation of the experimental data could be obtained; Equation (16) was rewritten as

$$U_2 - U_1 = C_{II} \Psi N_{Re} \qquad (16a)$$

The average values of U_1 and U_2 were measured for the three different density 1/4-in. spheres, independent of the distance l between them, during the 12-in. length of fall. For forty-two different runs in a Reynolds number range of 0.27 to 0.73 the average value of C_{II} was 0.11. Equations (12) and (13) can thus be rewritten as

$$W_1 = 3\pi\mu\, dU \cdot \left[1 - 3/4\frac{d}{l} + 0.11\, N_{Re}\right] \qquad (12a)$$

$$W_2 = 3\pi\mu\, dU \left[1 - 3/4\frac{d}{l}\right] \qquad (13a)$$

The bracketed terms correct Stokes's Law for the interaction effect between the spheres. The interaction effect, unlike the wall correction factor, is a function of the Reynolds number.

It is interesting to point out that the average value of C_{II} (0.11) over the Reynolds number range of 0.27 to 0.73 is a little more than twice the average value of C_I (0.05) obtained over the Reynolds number range of 0.5 to 3.5. This relative effect was predicted by Oseen, his constants being 3/8 and 3/16 respectively. Obviously the representation of the effect of Reynolds number in Equations (12) and (12a) will not be applicable at large distances between the spheres.

A QUALITATIVE ANALYSIS OF THE MOTION OF TWO AND THREE SPHERES AT HIGHER REYNOLDS NUMBERS

Before prediction of the complex interactions between assemblages of particles is tried, it is interesting to observe what happens with two or three particles settling at higher Reynolds numbers.

Figure 3 shows the effect of the inertial forces on the motion of two spheres falling one above the other along the axis of a cylindrical container in the Reynolds number range of 0.3 to 0.7. At time $t = 0$, the center to center distance between the two spheres is l_0. However the inertial forces will slow down sphere B [Equation (12)] without affecting the velocity of sphere A, so that l will become smaller until at some time $t = t_2$ the spheres will touch. Since the drag force exerted on sphere A is less than that exerted on sphere B, sphere A will tend to move over sphere B sidewise until the attractive force between the two spheres is just balanced by the repelling force given by Equation (14) ($t = t_3$ to $t = t_4$). The inertial forces will then cause the spheres to move away from the axis. This is shown at $t = t_5$ and $t = t_6$; the repelling force is about half as great as the attractive force when the spheres are falling one above the other.

It should be made clear that all the velocity measurements made to determine C_{II} were taken between $t = t_0$ and $t = t_1$ in Figure 3.

The motion of three spheres, as might be expected, is even more complex. For example (Figure 4) if spheres C, B, and A are falling in the same vertical plane so that the distance between the spheres A and B is less than the distance between spheres B and C, the two spheres A and B consitute a doublet and consequently move at a greater velocity than sphere C. At some time $t = t_1$, spheres A and B will have caught up to sphere C and so the distance between all three spheres is the same and a triplet is formed. However the triplet is not in stable equilibrium because the center sphere is influenced by interaction effects from both spheres A and C. These interaction effects cause sphere B to move closer to sphere C which produces a doublet between spheres B and C. At time $t = t_3$, the distance between sphere A and sphere B is greater than the distance between sphere B and sphere C because of the greater velocity of the doublet. These phenomena will occur even if inertial effects are negligible; the inertial forces act only to reduce the distance between the spheres in the doublet by slowing down the lower sphere during the length of fall.

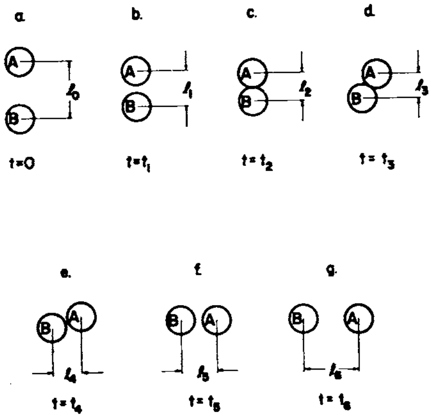

Fig. 3. Effect of inertial forces on the motion of two spheres.

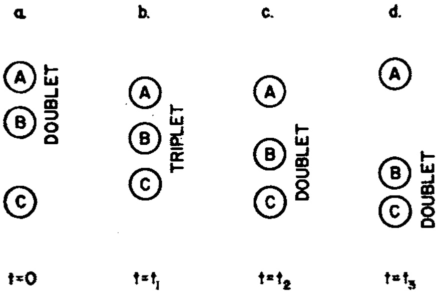

Fig. 4. The motion of three spheres.

CONCLUSIONS

The experimental values of λ obtained for the ¼-in. spheres agreed very closely with the values obtained by Stimson and Jeffrey from theoretical considerations over the l/d range of one to six diameters; no noticeable attraction between the spheres was observed. Since the Reynolds numbers were between 0.008 and 0.03, this would imply that inertial effects were negligible.

A definite attraction between two spheres falling one above the other was observed in the Reynolds number range of 0.25 to 0.7; the inertial forces acted to slow down the lower sphere without affecting the upper one.

Thus it is possible that spheres suspended in random orientation may not maintain their positions relative to each other. Perhaps the formation of doublets, and their corresponding higher velocities of fall, has been one of the causes for the wide discrepancies in the presently available fluidization data. Additional data will be required before the fundamental findings in this paper can be applied exactly to practical problems in fluidized or sedimenting systems of particles.

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NOTATION

(C.G.S. Gravitational Units employed unless otherwise indicated)

- a = radius of sphere, cm. or in.
- C_D = drag coefficient, dimensionless
- C_I = empirical constant (measures inertial effects for one sphere)
- C_{II} = empirical constant (measures inertial effects for two spheres)
- d = diameter of sphere, cm. or in.
- D = diameter of cylindrical column, cm. or in.
- g = acceleration of gravity
- K_I = Faxen correction factor for a cylindrical boundary on one sphere
- K_{II} = Happel correction factor for a cylindrical boundary on two spheres
- l = center to center distance between spheres, cm. or in.
- N_{Re} = Reynolds number = $dU\rho/\mu$, dimensionless
- R = radius of cylindrical column, cm. or in.
- U = terminal velocity
- U_{A_1} = initial undisturbed velocity field
- U_{A_2} = first reflection of initial undisturbed velocity field
- U_{B_1} = first reflection of velocity field at sphere A
- U_{B_2} = first reflection of velocity field at cylinder wall
- $U_{I\infty}$ = terminal velocity of a single sphere in an infinite fluid
- $U_{II\infty}$ = terminal velocity of two spheres in an infinite fluid
- U_I = terminal velocity of a single sphere in a bounded fluid
- U_{II} = terminal velocity of two spheres in a bounded fluid
- W = drag force; W_1 = drag force on bottom sphere, W_2 = drag force on top sphere

Greek Letters

- ρ = density of fluid
- ρ_S = density of sphere
- λ = correction factor for interaction effects between two spheres
- Ψ = $(\rho_S - \rho) g\, d^2/18\mu$, cm./sec.
- μ = viscosity of fluid
- $\cosh \alpha$ = l/d, dimensionless

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