Oscillations of Drops Falling in a Liquid Field

R. R. SCHROEDER and R. C. KINTNER
Illinois Institute of Technology, Chicago, Illinois

Frequencies and amplitudes of oscillations for nineteen pure systems were measured photographically. The period of oscillation was longer than that predicted by Lamb. The discrepancy was not due to wall effects, viscosity, or velocity of fall but to amplitude of oscillation. Modification of previous expressions included an amplitude function which could be experimental or empirical. Oscillations began near the peak velocity, and a vortex trail was necessary for them to take place. Oblate-prolate oscillations did not cause drop breakup, as all systems ceased to oscillate and wobbled randomly below maximum drop size. Oscillations do not decay with time.

The problem of oscillations of drops has been of interest for approximately 100 yr. The early interest was mathematical and resulted in derivations by Lord Kelvin (*39*), Rayleigh (*32*), Webb (*40*), and Lamb (*25*), giving the relationship between frequency of oscillation and the physical properties of the system. All deal with small oscillations of a fluid sphere at rest in an infinite motionless medium. The fluids are considered to be inviscid. Later derivations by Gilliland (*20*), Chandrasekhar (*3*), and Reid (*33*) include viscosity or higher-ordered (nonlinear) terms.

It has been shown that oscillations greatly increase the rate of heat and mass transfer between the moving drop and its field fluid (*2, 9, 11, 21*). The present work is not directly concerned with mass and heat transfer but is directed at finding out more about the oscillations themselves.

Some of the proposed causes of drop oscillations include formation, wake turbulence, vortex shedding, mass transfer, heat transfer, turbulence in the continuous phase, and wall effects. Hughes and Gilliland (*20*) and Garner (*6*) feel that the method of formation may be the determining factor in whether oscillations are initiated. There has been considerable work on fluctuations of solid spheres as related to turbulence in the wake and vortex shedding. Gunn (*13*) studied rain drops falling in air and found that the natural frequency of oscillation of drops and the frequency of eddy detachment are approximately the same. Oscillations of the wake are also cited by Marty (*28*) and Hughes and Gilliland (*20*). Miyagi (*29*) and Blanchard (*1*) relate oscillations to the opposing action of surface tension and resistance to motion. Many authors (*4, 12, 16, 17, 26, 27, 31, 36, 37, 38*) feel that local variations in surface tension (Marangoni effect) cause oscillations. These local variations may be due to mass transfer across the interface, heat transfer, surface active agents, or local variations in surface temperature due to mass transfer. Oscillations due to local variations in the surface tension are not regular but vary randomly as to direction, period, and amplitude.

Other areas of interest concerning drop oscillations in-

R. R. Schroeder is with the Esso Research Laboratories, Baton Rouge, Louisiana.

TABLE 1. PHYSICAL PROPERTIES OF THE MUTUALLY SATURATED PHASES

System (dispersed phase-continuous phase)	ρ_D, g./cc.	ρ_c, g./cc.	σ, dynes/cm.	$\mu_D \times 10^2$, poise	$\mu_c \times 10^2$, poise	Temp., °C.
Benzyl alcohol-water	1.037	0.999	3.94	4.085	0.910	28.0
Nitromethane-water	1.112	1.013	9.61	0.560	0.875	27.5
Carbon tetrachloride-water	1.584	0.997	36.2	0.880	0.875	25.8
Ethylchloroacetate-water	1.146	1.002	15.36	1.089	0.970	23.5
Nitrobenzene-water	1.198	0.998	22.0	1.771	0.892	25.2
Tetrabromoethane-water	2.951	0.998	35.6	9.34	0.900	25.0
Chlorobenzene-water	1.101	0.997	35.5	0.746	0.890	25.0
o-Nitrotoluene-water	1.158	0.997	26.4	2.051	0.890	25.0
Bromobenzene-water	1.491	0.998	31.2	1.075	0.934	23.1
Ethylene bromide-water	2.168	0.999	29.3	1.603	0.909	24.0
Dibromomethane-water	2.480	1.001	31.7	0.888	0.975	25.8
Tetrachloroethane-water	1.592	1.000	29.3	1.618	0.975	26.5
Tetrachloroethylene-water	1.614	0.996	37.0	0.835	0.876	25.0
Tetrachloroethylene-23.3% glycerine sol'n	1.617	1.058	32.8	0.911	1.766	24.2
Tetrachloroethylene-40.7% glycerine sol'n	1.615	1.101	29.9	0.899	3.15	26.0
Water-toluene	0.999	0.865	34.6	0.935	0.562	23.0
49.5% glycerine sol'n-toluene	1.128	0.867	22.1	5.79	0.577	20.6
63.3% glycerine sol'n-toluene	1.162	0.864	23.9	11.78	0.558	23.8
71.4% glycerine sol'n-toluene	1.192	0.867	19.9	25.91	0.578	24.0

clude types of oscillation, effect of oscillation on drop breakup, and the relationship of internal circulation and oscillations. Oscillation types have been described by Garner and Lane (*8*) and Kintner (*23*). Both agree that there is an axially symmetric oblate—prolate oscillation with which present work is concerned. The relationship between internal circulation and oscillation has been discussed by Hughes and Gilliland (*20*) and Johnson and Braida (*22*).

Many observers (*2, 7, 11, 14, 15, 30, 34, 35*) have reported that oscillations begin at drop Reynolds numbers as low as 200 and not higher than 500. Hartunian and Sears (*15*) also include Weber number and the *M* group $\left(\frac{g\mu_c^4}{\rho_c\sigma^3}\right)$ in their correlations.

Some authors (*10, 11, 21, 26*) report that oscillations decay, while others (*18, 24*) found no such decay. Hendricksen (*18*) observed oscillation of drops in air after a fall of 20 ft.

EXPERIMENTAL

All organic liquids used were the highest grade available from laboratory chemical supply firms. The distilled water used was from the regular laboratory supply. All phases were mutually saturated so as to eliminate mass transfer across the interface. Heat transfer was also avoided by conducting all tests at room temperature. The use of pure materials and the avoidance of mass and heat transfer are important to eliminate the Marangoni effect mentioned above which would greatly complicate the study.

The physical properties of the materials used were measured immediately prior to the photographing of drop oscillations. Density was measured by use of a pycnometer; several determinations were made at 25.0°C. The densities of the high viscosity glycerine solutions were measured with a hydrometer. Viscosity was measured with a viscometer at approximately 20°, 25°, and 30°C. with several determinations at each temperature. To obtain the viscosity at the temperature of the run, the viscosity data were interpolated. The density data were also corrected to the temperature of the run. Interfacial tensions were measured with a ring tensiometer. All the physical data for the mutually saturated phases at the temperatures of the runs are given in Table 1.

All drops were formed at submerged nozzles made from brass and with edges bevelled free from burrs to insure consistency in drop size. The two smallest nozzles were hypodermic needles. There were twenty-one nozzles giving a drop size range from well below the peak velocity to the size where wobbling was taking place for most systems. It is felt that there is no point in reporting nozzle diameters since there are several other important variables involved in duplicating a drop size. Three of these variables are, of course, how far the nozzle is submerged in the field fluid, the head of liquid above the nozzle, and the rate of drop formation. In this work these variables were controlled as closely as possible to insure uniform drop size. Rate of drop formation was sufficiently slow that disturbances in the field fluid owing to the preceding drop were damped out, as indicated by a study of fall velocity and frequency of oscillation at varying drop formation rates. Drop volumes were determined by measuring the total volume of several hundred drops.

With the exception of wall effect runs, all drops were falling in a cylindrical glass tube 65 cm. long and 9.48 cm. in diameter. For the wall effect runs, with tetrachloroethylene used, tubes of 6.94-, 4.61-, and 2.20-cm. diameters were also used. A grid with the centimeter lines accented was placed behind the tube; this facilitated determination of the velocity and amplitude data from the films. An electric timer is in view in all films to obtain the necessary time intervals. All filming was done at 64 frames/sec., and this was sufficient to determine accurately the periods of oscillation for all systems used. The filming was done from 15 ft. away with a telescopic lens to eliminate parallax.

The data obtained from the films included drop velocity and frequency and amplitude of oscillation. In general, five individual drops at each size were photographed. Drop velocities were measured from 28 to 46 cm. below the nozzle. It was found that at least 28 cm. of fall was required to reach terminal velocity for oscillating drops. The frequency of oscillation is taken as an average of twenty-five oscillations. The amplitudes of oscillation are an average of ten measured amplitudes with the maximum and minimum vertical diameters. The amplitude is defined here as

$$B_e = \frac{1}{2}\,(d_{\max} - d_{\min})/d_{\mathrm{avg}} \qquad (1)$$

The amplitude data are very scattered, and in all calculations a smoothed amplitude is used. It should be pointed out that whereas the frequency of oscillation is very consistent, the amplitudes are very erratic. From one oscillation to the next the amplitude may vary considerably, especially when the drop size is near the random wobble range.

RESULTS

The observable start of oscillations in every case took place at or shortly after the peak velocity. This tends to confirm the thoughts of others that the very fact that there is a decrease in velocity after a certain point is due to the oscillations themselves. The drop size at the onset of oscillations can therefore be gotten from the break in the Hu—Kintner (*19*) correlation or from a terminal velocity curve if the latter be available.

As stated earlier, there can be many causes for drops to behave in an oscillatory manner. In this work heat and mass transfer and surface active contaminants were avoided. Nozzle effects are considerable and can easily be observed near the nozzle. Under extreme conditions, nozzle-induced oscillations can be of sufficient violence to cause drop rupture (*23*). Even in the cases where oscillations are not sustained (below the peak velocity), there may be several oscillations before they are damped out. The concern here is not, however, with these nozzle-induced oscillations. The oscillations considered here are the steady state, nondamped oscillations taking place well below the point of drop formation. In most cases a definite change in oscillatory motion can be observed at some point below the nozzle which indicates that the transient behavior is damped out. Steady state conditions are not achieved until well below the nozzle. This is probably because a steady condition can not be reached until a vortex trail is fully developed.

The vortex trail, in the authors' opinion, is a necessary condition for oscillations to take place. In every case the drop Reynolds number at the onset of oscillation is above that which is observed for the onset of a vortex trial in experiments conducted on solid bodies. In the cases of solid bodies, the vortex shedding was considered to be responsible for fluctuations, and the frequency of these fluctuations could be correlated to the frequency of vortex discharge. A correlation between drop oscillation and frequency of vortex discharge was attempted, and although there was a pattern, the scatter was quite large. Some observers have felt that oscillations are caused by a combination of frontal drag which tends to flatten the drop and the interfacial tension which tends to retain the drop in a spherical shape. It would seem, however, that a steady state would be reached in which these opposing forces would balance one another. Consider the analogy of an undamped (or slightly damped) spring which is perturbed by a continuing outside force. In this case the spring will oscillate at or near its natural frequency and continue oscillating as long as a sustaining mechanism is

* All data, derivations, curves, etc. may be found in the Ph.D. Thesis of R. R. Schroeder, Illinois Institute of Technology, Chicago, Illinois, and are available in microfilm form.

present. Thus, a drop with a vortex trail has such a sustaining mechanism which causes the drop to oscillate at or near its natural frequency and explains the nondamping of the oscillations.

As stated earlier drop oscillations begin at or near the peak velocity. Shortly after the drop size at which oscillations begin, they are well developed and of large fractional amplitude. There must be a narrow drop size range over which the amplitude of oscillation increases from zero to the observed values, although this was noted in only a few systems. Most of the observed oscillations, over a certain size range, were very regular and well defined. These oscillations are commonly described as oblate—prolate, although in most cases the drop never reaches the prolate shape but rather oscillates between oblate to less oblate. This is why an average diameter is used in the amplitude calculations rather than the equivalent spherical diameter. Some systems exhibit a rippling or surface wave effect over the surface of the drop. In all systems at the larger drop sizes a random wobbling effect takes place which obscures the oblate—prolate oscillations. This random effect takes place well below the maximum drop size observed by others, and since oscillations are not present at or near the maximum drop size, they cannot cause drop breakup as postulated by Elzinga and Banchero (*5*). Large drops are torn apart by viscous shear or by uneven gross distortion.

Wall effects were studied by using a range of tube diameters, and it was found that the velocity of fall varied as predicted by others but that the frequency of oscillation was not at all affected. This indicates that the velocity of fall does not affect the frequency of oscillation and tends to support the vortex shedding—natural drop frequency concept.

Glycerine was added to the various phases to study the effects of viscosity on oscillations. As previously noted, the continuous-phase viscosity does greatly affect the velocity of fall, especially at viscosities of over 10 centipoises. The continuous-phase viscosity also greatly affected oscillations, and when the increased viscosity and resultant decreased velocity caused the Reynolds number to fall below 200, the oscillations ceased. It is interesting to note that a Reynolds number of 200 is the minimum necessary for vortex shedding to take place. In general the frequency of oscillation is not greatly affected by the low range of continuous-phase viscosity provided the requirement of vortex shedding is satisfied. Dispersed-phase viscosity has less effect on both velocity of fall and oscillations. As the drop viscosity goes up, larger and larger drops are required for the onset of oscillations. In this case Reynolds numbers considerably over 200 are reached, and the lack of oscillations may simply be due to viscous damping in the drop. As long as oscillations take place, however, the frequency is not greatly affected by drop-phase viscosity.

THEORY

As mentioned earlier, the problem of oscillating drops was first solved mathematically by Rayleigh (*32*) and by Webb (*40*) using different techniques. The original solutions were for drops in a gas, and these were modified by Lamb (*25*) for the general case of a field fluid of any density. Here an additional modification will be made to include an amplitude factor which helps to describe real systems more accurately.

Let an oscillating drop have a surface which may be represented by

$$r = a + b\,\frac{V_n}{a^n} \tag{2}$$

If the same series of mathematical manipulations (*25, 40*) be performed, the amplitude function will appear in the final equation so that

$$\omega^2 = \frac{\sigma b}{a^3}\cdot\frac{n(n+1)(n-1)(n+2)}{(n+1)\,\rho_D + n\rho_c} \tag{3}$$

which is identical with the Lamb equation except for the factor b. A similar approach was actually started in the Webb derivation but was not followed through, probably because it would not lead to a result identical with that of Rayleigh.

The first mode of oscillation ($n = 2$) is the only one observed experimentally. The factor b should approach unity for small oscillations. Hence, when one takes into account the experimental findings, b is defined as

$$b = 1 - \frac{d_{max} - d_{min}}{2d_{avg}} = 1 - B, \tag{4}$$

COMPARISON WITH THE RAYLEIGH-WEBB (LAMB) EQUATION

To predict a frequency of oscillation from the above equation it would be necessary to know the amplitude of

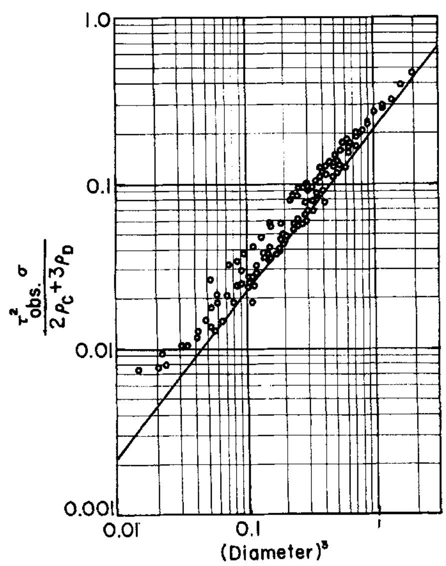

Fig. 1. Experimental data compared with Lamb's equation.

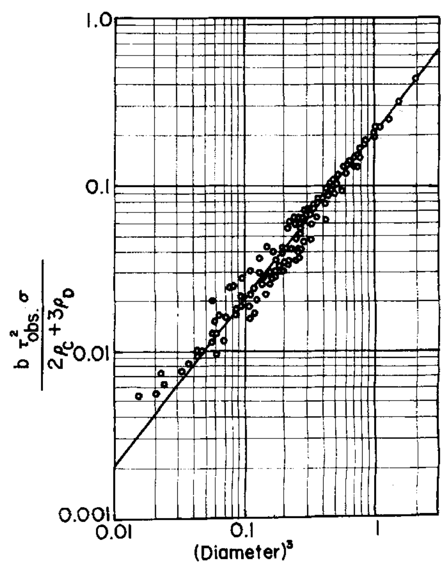

Fig. 2. Amplitude corrected data vs. drop size. Values of b from Equation (4).

oscillation. To avoid this the amplitude data were correlated empirically with a value

$$b = 0.805\, d_e^{0.225} \tag{5}$$

It should be cautioned that experimentally the amplitude data are very scattered, and the above correlation should not be used to predict amplitude but only to predict frequency. A comparison of the observed data with those predicted by Lamb (the Rayleigh-Webb derivation) shows that the average error in prediction of frequency of oscillation is 16.33%. The average error of prediction with the value of b from Equation (4) used is 9.24%, whereas when the empirical value of b from Equation (5) is used the average error is 9.01%. The data were for nineteen systems and a total of 132 points. Figures 1 and 2 show a plot of drop size vs. a time term. In Figure 1 the time term is based on observed frequencies of oscillation. In Figure 2 this time term is corrected for the experimental amplitudes in accordance with Equation (4). In both figures the straight line is the Lamb equation plotted in the same dimensional form as the ordinate.

CONCLUSIONS

Listed below are some of the conclusions of this work.

1. Oscillations begin at or near the drop size corresponding to the peak velocity.
2. A necessary condition for oscillations is the presence of a vortex trail which is the driving mechanism for the oscillations. This requires a Reynolds number of at least 200.
3. The oscillations discussed here (not nozzle induced) do not decay with time.
4. Drop breakup is not caused by normal oscillations.
5. The frequency of oscillation is best correlated by the equation presented here with the empirical amplitude correction of Equation (5) used.
6. Velocity of fall does not affect the frequency of oscillation provided the vortex trail is present to drive the drop oscillations at their natural frequency.
7. Dispersed-phase viscosity has very little effect on oscillations except at high viscosities where it damps them out.
8. Continuous-phase viscosity does not affect oscillations as long as a vortex trail is present. The vortex trail is damped out, however, at a fairly low continuous-phase viscosity.
9. The only type of oscillation observed here is the oblate—prolate type corresponding to the primary mode of the theoretical equation. This degenerates as the drops become large to random wobbling. Surface fluttering is observed in some systems but is not oscillatory.

NOTATION

a = radius of equilibrium sphere
b = amplitude dependent coefficient
B_s = fractional amplitude, defined by Equation (4)
d_e = equivalent spherical diameter
d_{max} = maximum vertical drop diameter
d_{min} = minimum vertical drop diameter
d_{avg} = average vertical drop diameter
g = gravitational acceleration
n = integer
r = radius
V_n = spherical solid harmonic

Greek Letters

μ_c, μ_D = viscosity of the continuous and dispersed phases, respectively
ρ_c, ρ_D = density of the continuous and dispersed phases, respectively
σ = interfacial tension
τ = period of oscillation
ω = angular frequency of oscillation

LITERATURE CITED

1. Blanchard, D. C., *Trans. Am. Geophys. Union*, **31**, 836 (1950).
2. Calderbank, P. H., and I. J. O. Korchinski, *Chem. Eng. Sci.*, **6**, 65 (1956).
3. Chandrasekhar, S., *Proc. London Math. Soc.*, **9**, No. 3, p. 141 (1959).
4. Davies, T. V., and D. A. Haydon, *Proc. Roy. Soc. (London)*, **243A**, 492 (1958).
5. Elzinga, J. C., and J. T. Banchero, *A.I.Ch.E. Journal*, **7**, 394 (1961).
6. Garner, F. H., *Trans. Inst. Chem. Engrs. (London)*, **28**, 88 (1950).
7. ———, V. G. Jenson, and R. B. Keey, *ibid.*, **37**, 191 (1959).
8. Garner, F. H., and J. J. Lane, *ibid.*, p. 162.
9. ———, and A. H. P. Skelland, *Ind. Eng. Chem.*, **46**, 1255 (1954).
10. ————, and P. J. Haycock, *Nature*, **173**, 1239 (1954).
11. Garner, F. H., and H. Tayeban, *Anales Real Soc. Espan. Fis. y Quim. (Madrid)*, **56B**, 479 (1960).
12. Groothuis, H., and F. J. Zuiderweg, *Chem. Eng. Sci.*, **12**, 288 (1960).
13. Gunn, R., *J. Geophysics Res.*, **54**, 383 (1949).
14. Harmathy, T. Z., *A.I.Ch.E. Journal*, **6**, 281 (1960).
15. Hartunian, R. A., and W. R. Sears, *Fluid Mech.*, **3**, 27 (1957).
16. Haydon, D. A., *Nature*, **176**, 839 (1955).
17. ———, *Proc. Roy. Soc. (London)*, **243A**, 483, 492 (1958).
18. Henrickson, F., M.S. thesis, Mass. Inst. Technol., Cambridge, Massachusetts (1941).
19. Hu, S., and R. C. Kintner, *A.I.Ch.E. Journal*, **1**, 42 (1955).
20. Hughes, R. R., and E. R. Gilliland, *Chem. Eng. Progr.*, **48**, 497 (1952).
21. ————, *Chem. Eng. Progr. Symposium Series No. 16*, **51**, 101 (1955).
22. Johnson, A. I., and L. Braida, *Can. J. Chem. Eng.*, **35**, 165 (1957).
23. Kintner, R. C. "Advances in Chemical Engineering," Vol. 4, p. 51, Academic Press, New York (1963).
24. Korchinski, I. J. O., Ph.D. thesis, Univ. of Toronto, Toronto, Canada (1956).
25. Lamb, H., "Hydrodynamics," 6 ed. (1945).
26. Lewis, J. B., and H. R. C. Pratt, *Nature*, **171**, 1155 (1950).
27. MacKay, G. D. M., and S. G. Mason, *ibid.*, **191**, 488 (1961).
28. Marty, L., *J. de Phys. et le Radium*, **6**, 373 (1935).
29. Miyagi, O., *Tokoyo Imp. Univ. Tech. Rept.* 3, No. 5, p. 1 (1925).
30. Moore, D. W., *Fluid Mech.*, **6**, 113 (1958).
31. Orell, A., and J. W. Westwater, *A.I.Ch.E. Journal*, **8**, 350 (1962).
32. Rayleigh, Lord, *Proc. Roy. Soc. (London)*, **29**, 71 (1879).
33. Reid, W. H., *Quart. J. Appl. Math.*, **18**, 1 (1960).
34. Saffman, P. G., *Fluid Mech.*, **1**, 249 (1956).
35. Schiller, L., and H. Schmiedel, *Z. fur Flugtechnile und Mot.*, **19**, 497 (1928).
36. Sherwood, T. K., and J. C. Wei, *Ind. Eng. Chem.*, **49**, 1030 (1957).
37. Sigwart, K., and H. Nassenstein, *Ver. Deutsch. Ing. Z.*, **98**, 453 (1956).
38. Sternling, C. V., and L. E. Scriven, *A.I.Ch.E. Journal*, **5**, 514 (1959).
39. Thomson, Sir W., *Phil. Trans.*, **153**, 583 (1863).
40. Webb, R. R., *Mess. of Math.*, **9**, 170 (1880).

Manuscript received May 8, 1964; revision received August 24, 1964; paper accepted August 24, 1964. Paper presented at A.I.Ch.E. Pittsburgh meeting.