THE FALL OF SINGLE LIQUID DROPS THROUGH WATER

SHENGEN HU AND R. C. KINTNER
Illinois Institute of Technology, Chicago, Illinois

The steady motion of single drops of ten organic liquids falling through a stationary water field is discussed. A correlation is presented for nine systems with the exception of the aniline-water system, in the form of a single curve relating the drag coefficient, Weber number, Reynolds number, and a physical property group. The curve can be used directly to predict the terminal velocity, drag coefficient, Reynolds number, and Weber number for any given equivalent drop size.

A break point in the curve serves to predict the peak velocity and its related quantities. The critical drop size is predicted from the pertinent physical properties alone. All these estimations were accurate within 10% for the systems used. The interfacial tensions ranged from 24 to 45 dynes/cm. and the drop densities from 1.100 to 2.947g./ml., the latter resulting in a twentyfold range of density differences. The drop viscosities had no apparent effect.

With the exception of extended-surface units and packed towers, which operate by the film-contact mechanism, nearly all types of liquid-liquid–extraction equipment used today secure contact through the dispersion of one phase as drops (*23*). Even in packed towers, drop formation has been found to prevail when the dispersed phase does not wet the packing (*4*). It is thus apparent that a study of the motion of liquid drops in a second liquid medium should provide basic information for the design of most liquid-liquid extractors, as drop size is directly related to stage efficiency and terminal velocity to the capacity of the equipment.

The hydrodynamic aspect of the problem is so complex that even for the more simple cases analyzed (*15*) many assumptions have to be made in order to render any mathematical solution possible. It is generally assumed, for instance, that the drop is of a simple spherical or ellipsoidal shape regardless of its size and velocity or of the physical properties of the system. From the fragmentary experimental results obtained by previous workers it has been found that a liquid drop does not always behave like a rigid sphere or ellipsoid. This discrepancy should be easily anticipated inasmuch as the nature of a liquid-liquid interface is not the same as that of a liquid-solid boundary and the shape of the drop is not always rigid and spherical or ellipsoidal. With these considerations the present investigation has been initiated with the simple objective of experimentally studying the behavior of single liquid drops falling under steady-state conditions in a stationary water field as governed by the pertinent physical properties of the system, i.e., densities, viscosities, and interfacial tensions. No mass transfer was involved.

PREVIOUS WORK

The translation of a solid sphere in an infinite mass of a perfect liquid was first analyzed by Stokes (*30*) in 1845 and was later reduced to the case of viscous flow by neglecting the inertial-force terms in the Navier-Stokes equations of motion (*15*). The simple Stokes' law of resistance thus obtained is $F = 3\pi\mu du$, which holds only at Reynolds numbers of less than two (*3, 24, 32*).

The question of the existence of slipping at the boundary surface of a solid sphere moving in a liquid medium within the Stokes'-law range has been treated by Basset (*6*), but experimental evidence obtained by various workers seems to indicate that there is no slipping at the solid-liquid boundary (*2, 3, 13*).

Shengen Hu is with the Institute of Gas Technology, Chicago, Illinois.

TABLE 1.—PHYSICAL PROPERTIES, C. G. S. UNITS

	System	ρ_o	ρ	$\frac{\Delta\rho}{\rho}$	$\mu_o \times 10^2$	$\mu \times 10^2$	σ_i	P	T
1	Aniline	1.0176	0.9979	0.0197	3.5416	0.9471	2.8*	1.41×10^8	25.0
2	Tetrabromoethane	2.9474	0.9973	1.9553	9.2888	0.8968	35.9	3.73×10^9	24.9
3	Dibromoethane	2.1541	0.9966	1.1614	1.5852	0.8968	31.9	4.39×10^9	25.0
4	Ethyl bromide	1.4478	0.9977	0.4511	0.4908	0.8814	30.0	1.01×10^{10}	26.2
5	Nitrobenzene	1.1947	0.9972	0.1981	1.7379	0.8835	24.1	1.18×10^{10}	25.6
6	Bromobenzene	1.4881	0.9971	0.4924	1.0719	0.8958	37.9	1.75×10^{10}	25.0
7	*o*-Nitrotoluene	1.1576	0.9970	0.1609	2.0360	0.8996	26.5	1.80×10^{10}	24.7
8	Tetrachloroethylene	1.6143	0.9970	0.6192	0.8903	0.8946	44.4	2.25×10^{10}	25.1
9	Carbon tetrachloride	1.5770	0.9957	0.5838	0.8702	0.7797	40.6	3.14×10^{10}	30.4
10	Chlorobenzene	1.0995	0.9969	0.1029	0.7606	0.9036	36.7	7.30×10^{10}	24.8

Symbols:
ρ = density
μ = viscosity
σ_i = interfacial tension
Sub o = organic phase
Others = water phase

$$P = \frac{\sigma_i^3 \rho^2}{g \mu^4 \Delta\rho}$$

T = temperature, °C.
*System unstable.

The case of a liquid sphere was analyzed by Rybczynski(*25*) in 1911 and independently by Hadamard(*11*) in the same year. Lamb (*15*) has given a detailed discussion by considering the internal as well as the external motions and has given the following correction factor for Stokes' law when steady-state conditions prevail:

$$F = K_1 \cdot 3\pi\mu\, a\, U_o$$

and

$$U_o = (1/K_1)\, g\, d^2 \Delta\rho / 18\mu$$

$$= \text{terminal velocity}$$

where

$$K_1 = (3\mu_o + 2\mu)/(3\mu_o + 3\mu)$$

Bond (*7*) and Bond and Newton (*8*) made further investigations on this subject. By taking into account the interfacial tension at the drop-medium boundary, they speculated at a critical drop size at which the drop motion would start to deviate from that of a rigid sphere. Unfortunately their results were too few and too erratic to substantiate their postulations.

Allen(*2*) proposed to explore relatively high Reynolds numbers by means of dimensional analysis. By analogy with the flow of liquids through pipes(*26*) he assumed that the resistance to the motion of a sphere was proportional to the *n*th power of the velocity:

$$U_o \propto d^{\,(3/n)\,-\,1} \Big/ \nu^{\,(2/n)-1}$$

Thus if n is 1.0 and K is 3π, Stokes' law is obtained and U_o varies as the square of the drop diameter. If n is 1.5, U_o is proportional to d, and

$$F = k\rho^{\frac{1}{2}}\, \mu^{\frac{1}{2}}\, d^{3/2}\, u^{3/2}$$

If n is 2.0, U_o varies as $d^{\frac{1}{2}}$, and the resistance is independent of viscosity. This is the so-called "Newton's law of motion":

$$F = k\rho a^2 u^2$$

Allen has verified these equations with his own data for all three ranges.

Characterization of drop motion by Reynolds-number ranges was also done by Smirnov and Ruban (*28*), who worked on eleven organic liquid-water systems, using water as the field liquid in most of their experiments. The highest *Re* they reached was less than 1,000.

THEORETICAL CONSIDERATIONS

Deformation and Oscillation. The main reasons for the differences between the motion of liquid drops and that of rigid spheres are believed to be the deformation and oscillation of the drops as well as the flow on the drop surface and the circulation inside the drops. The effects of the last two factors on the motion of liquid drops have been the subjects of many experimental investigations and theoretical analyses in the past(*2, 3, 7, 8, 13, 15*), covering *Re* ranges both within and beyond the Stokes'-law region. Unfortunately no definite conclusions have been reached to date.

Previous workers (*15, 24, 29, 31*) have also analyzed the deformation of moving drops without oscillation and the oscillation of deformed drops without gross drop motion. Solutions for such ideal cases are expressed in terms of the eccentricity of the drops, the amplitude and frequency of their oscillations, and the time of oscillation decay, as related to drop size and velocity and the physical properties of the system. Despite these mathematical formulations, the exact nature of drop motion still remains to be determined by experimental methods because of the inseparableness of the two interfering factors in the actual situation.

Terminal Velocity. Under steady-state conditions, the gravity force on a falling spherical drop is exactly balanced by the resistance it encounters:

$$\frac{\pi d^3}{6} \Delta\rho g = C_D \frac{\pi d^2}{4} \frac{U_o^2 \rho}{2}$$

from which

$$C_D = (4/3)\,(\Delta\rho/\rho)\,(dg/U_o^{\,2}$$

$$= (4/3)\,(\Delta\rho/\rho)\,(1/Fr)$$

If the drop starts to fall from rest, it will accelerate for a short distance before reaching its terminal velocity. It can be shown that (*15*),

$$\frac{\pi d^3}{6}\left(\rho_o + \frac{\rho}{2}\right)\frac{du}{dt} =$$

$$\frac{\pi d^3}{6}\Delta\rho g - C_D \frac{\pi d^2 u^2 \rho}{4 \times 2}$$

When solved

$$u = U_o \tanh\left(\frac{U_o t}{\propto}\right),$$

$$S = \propto \ln\cosh\left(\frac{U_o t}{\propto}\right)$$

where

$$U_o = \sqrt{(4/3)\ (\Delta\rho/\rho)\ (dg/C_D)}$$

$$\propto = (2/3)\ (d/C_D)\ (2\rho_o + \rho)/\rho$$

hence

$$u = U_o\left[1 - \exp\left(-\frac{3C_D \rho S}{(2\rho_o + \rho)d}\right)\right]^{\frac{1}{2}}$$

The relationship between C_D and U_o has been experimentally established for various rigid shapes (*10*), but to date no generalized correlation has been obtained for liquid drops.

Dimensional Analysis. As the objective of the present work is to characterize drop motion by means of the pertinent physical properties of the systems concerned, it would appear that the best way to correlate the experimental results is by dimensional analysis in a manner similar in many respects to that used by Peebles and Garber (*23a*) for gas bubbles. One may consider

$$f(\mu_o, \mu, \rho, \Delta\rho, \sigma_i, d, U_o, g) = 0$$

or

$$K.\ \mu_o^a.\ \mu^b.\ \rho^c.\ \Delta\rho^e.\ \sigma_i^f.\ d^h.\ U_o^i.\ g^k = 1$$

If the equation is solved dimensionally,

$$\left(\frac{\mu_o}{\mu}\right)^a \left(\frac{\Delta\rho}{\rho}\right)^b (Re)^c\, (We)^d\, (M)^e = 1$$

where

$$M = \frac{g\mu^4}{\rho\sigma^3} = \frac{3\rho C_D\, We^3}{4\Delta\rho Re^4}$$

A short history of the *M* group as well as a resumé of the attempts made by previous workers to use it for correlating experimental data can be found in the literature (*1*, *27*). In the data analysis of the present work, however, a modified form of the *M* group, called the *P* group, has been found to give better correlations than the *M* group:

$$P = \frac{1}{M} \frac{\rho}{\Delta\rho}$$

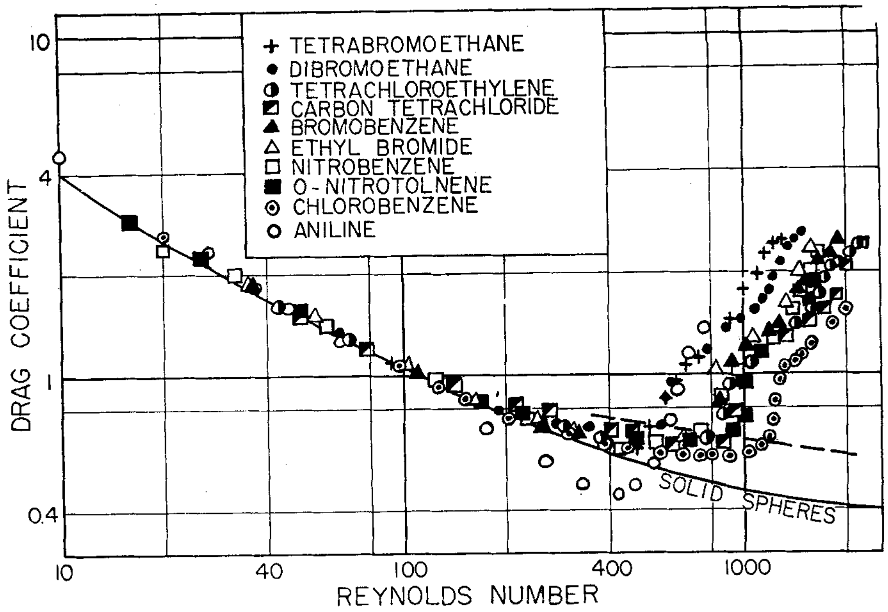

Fig. 1. Variation of Drag Coefficient with Reynolds Number.

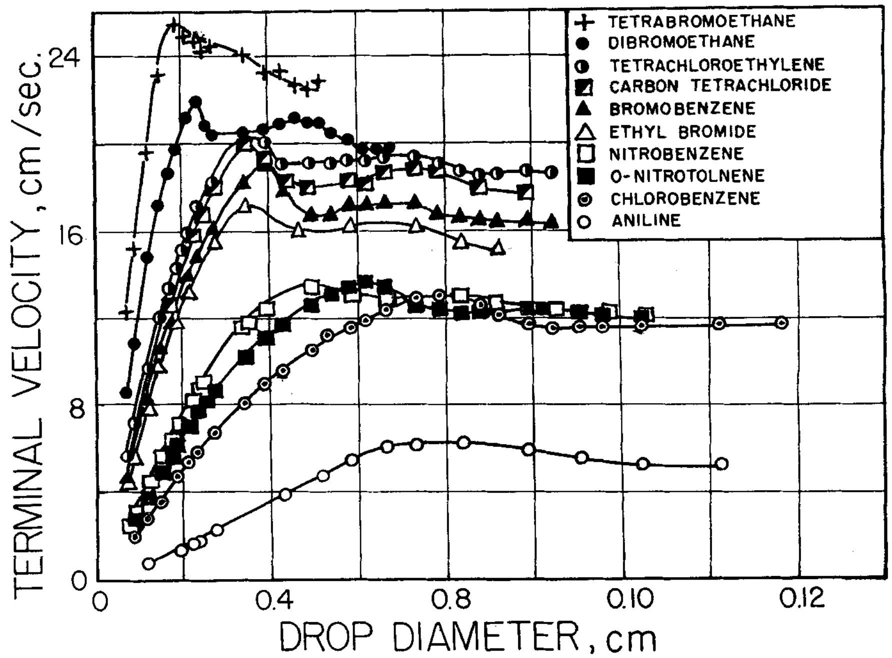

Fig. 2. Variation of Terminal Velocity with Drop Size.

EXPERIMENTAL*

Ten technical-grade organic liquids, with densities greater than the density of water, were purchased from Mathieson Chemical Corp. for forming the drops. Before being used, each compound was saturated with distilled water by being shaken with it in a separatory funnel. Distilled water, to be used as the field liquid, was also saturated with the organic compound, the two being stirred with a small mixer in a storage tank.

The densities and viscosities of the two phases and the interfacial tension of each system were determined both before and after each run in order to make sure that no appreciable changes in these constants had taken place during the course of the run. Densities were determined by the conventional weighing method by means of a fine balance and a glass bob. An Ostwald viscometer was used for the viscosity measurements. The capillary method for surface-tension measurements was modified to determine the interfacial tension of the liquid-liquid-systems (*5*). Table 1 gives the average results.

Drops of from 1.6 × 10^{-4} ml. to 8.0 × 10^{-3} ml. volume were delivered directly from a capillary tube into the test tank for velocity measurements. The capillary was made from a thermometer stem with a tip drawn out at one end and a rubber bulb fitted on the other end. The capillary bore, calibrated with a mercury bead of known weight, was found to be 0.226 mm. in radius. With the tip immersed under the water surface in the test tank, the desired amount of organic liquid was squeezed out by slightly pressing the rubber bulb and shaking the drops off from the tip. Care was taken to see that the tiny droplet formed in this way did not stick back to the side of the tip or break up when coming off.

Drops from 10^{-2} to 2.000 ml. in size were delivered into small weighing bottles from a microburette graduated to 10^{-2} ml. The tip of the burette was always kept immersed in a basin partly filled with the water phase so that the organic liquid would never come into contact with any free water

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*For Tables 5 to 14, giving the detailed diameter vs. velocity data, order document 4478 from American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D.C., remitting $1.25 for microfilm or photoprints.

surface. This precaution prevented part of the drop liquid from floating on the water surface, thus rendering the volume measurement inaccurate. The receiving vessels were always kept clean in order to avoid any sticking of the drop to the vessel walls. Twenty-five weighing bottles were used to hold twenty-five drops of the same size. The weighing bottles were then transferred one at a time to the test tank and the drop was carefully poured out under the water surface for the terminal-velocity measurement.

The test tank was made by cementing four 1/4- by 12- by 48-in. glass plates onto a framework of 1 1/2-in. angle irons bolted together. Permanite cement, made by Maurice A. Knight Co., was used for cementing the glass and Pliobond cement, made by Goodyear Rubber Co., for sealing the leaks. The tank bottom was a 1/16-in. stainless steel sheet bolted to the framework with a rubber gasket in between. A storage tank was connected with 1-in. iron pipes to a 4-in. stainless steel funnel soldered to the center of the stainless steel bottom of the test tank. A centrifugal pump installed in the pipe line served to pump the water through a two-way valve connection from one tank to the other as desired.

A thermometer was kept hanging half way down one corner of the test tank to keep a close check on the water temperature, which was maintained constant within ± 0.5°C.

The terminal velocities of the drops were measured by timing with an electric chronograph a distance of fall of 1 ft. between two marks near the bottom of the tank but sufficiently high to eliminate end effects. Twenty-five time measurements were made for each drop size, and the average was used for calculating the velocity.

In order to be certain that terminal velocity had been reached in all cases, the equations presented before were used to calculate the distance of fall required for attaining steady-state conditions, and the values obtained were found to be very much less than the actual distance of fall of about 70 cm. prior to the time measurements. Photographic measurements on three systems also substantiated the attainment of terminal velocity. Furthermore, these values check remarkably well with the results obtained by time measurements, indicating the timing equipment to be a reliable one.

The effects of rigid surface proximity on the motion of solid spheres in the Stokes'-law region have been studied rather exhaustively in the past(*14, 17, 19, 32*). For a solid sphere moving along the axis of a cylindrical container, the general form of the Stokes'-law correction factor is(*9*)

$$K_w = 1 + m\,(d/D)$$

where the constant m ranges from 2.1 to 2.4 according to different authors. Generalized correction for wall effects at higher Reynolds numbers has not yet been found possible(*18, 19*). Absence of wall effects in the test tank has been proved by comparing the data on three systems with those obtained in four glass cylinders with inside diameters of 2.14, 4.59, 6.99, and 9.46 cm. respectively after the method used by McNown, et al.(*19*).

OBSERVATIONS ON DROP MOTION

Although it has been speculated that any surface flow or internal circulation may cause a liquid drop to behave differently from a rigid sphere, yet the data obtained in the present work indicate that no such difference can be observed up to a *Re* of about 300. This fact is borne out by the coincidence of log C_D vs. log *Re* curves for the liquid drops with that for solid spheres below *Re* 300, as shown in Figure 1.

As the limiting *Re* range is passed, however, the log C_D vs. log *Re* curves for the liquid systems depart from the curve for solid spheres but still retain a similar general pattern. At first C_D decreases only very slightly with increasing *Re* for a rather large *Re* range, but when the *Re* reaches a certain value, C_D starts to increase rather abruptly within a narrow *Re* range. After the abrupt rise the curve proceeds at a more or less constant slope until the critical drop size is reached.

On plotting the terminal velocities against the equivalent drop diameters in rectangular coordinates for each system, as shown in Figure 2, one finds that there is a maximum or peak in each curve. This peak velocity occurs at correspondingly larger drop sizes and decreases in value as the density difference of the system becomes smaller. After the peak, the curve shows a dip and then flattens out, sometimes with a slight bump.

The general forms of the log C_D vs. log *Re* and U_o vs. d curves can be interpreted in the light of the change in behavior of the falling drops. It is observed that, as the

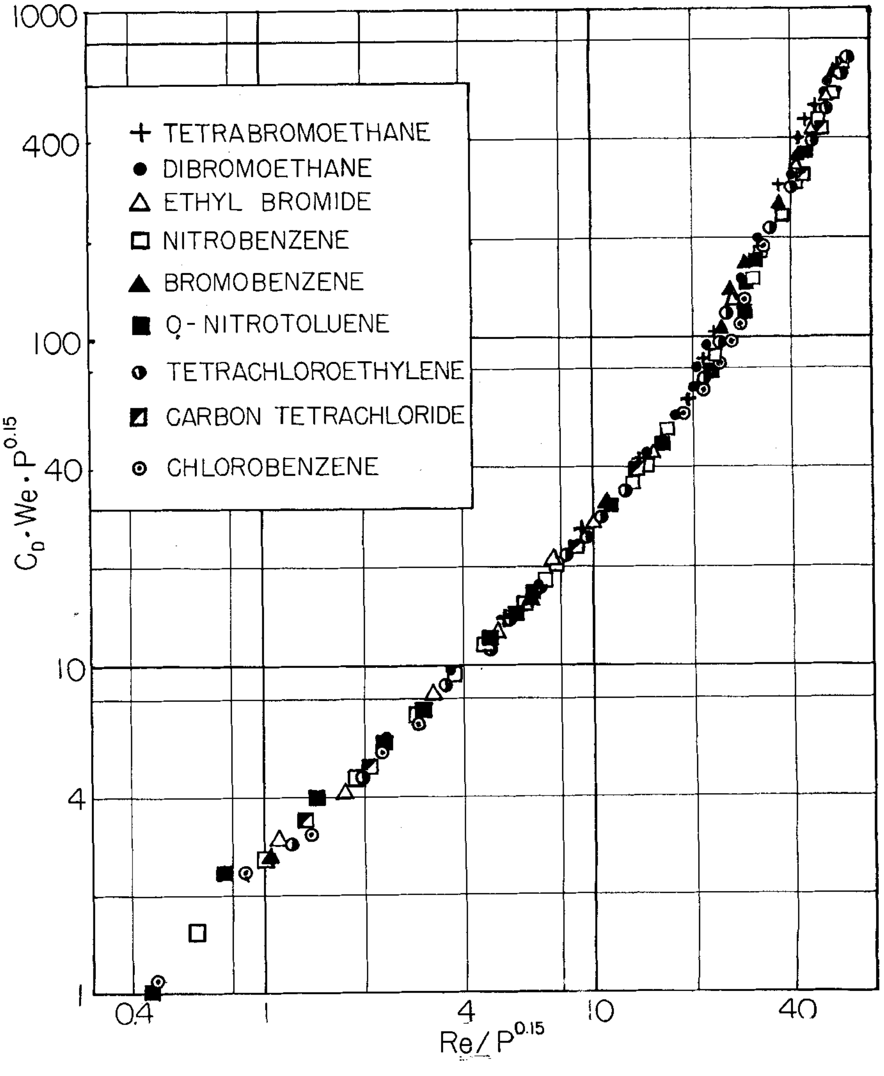

FIG. 3. GENERAL CORRELATION.

TABLE 2.—DATA AT PEAK TERMINAL VELOCITIES

$$\left(\frac{Re}{We}\right)_p = 0.816\ P^{0.238} \qquad (U_o)_p = 1.23\left(\frac{\sigma_i}{\mu}\right)P^{-0.238}$$

$$(We)_p = 3.58 \qquad (d)_p = 3.58\left(\frac{\sigma_i}{\rho}\right)\times\frac{1}{U_o^2}$$

	System	$(d)_{p\ obs}$, cm.	$(U_o)_{p\ obs}$ cm./sec.	$(Re)_p$	$(We)_p$	$\left(\frac{Re}{We}\right)_p$	P	$(d)_{p\ calc}$, cm.	Dev. d, %	$(U_o)_{p\ calc}$ cm./sec.	Dev. U_o, %
1	Aniline	0.726	6.2	474	10.3	46	$1.41\text{x}10^8$	0.600	—17.4	4.1	—33.9
2	TBE	0.200	25.9	576	3.73	155	$3.73\text{x}10^9$	0.190	— 5.0	26.0	+ 0.4
3	DBE	0.230	22.1	565	3.51	161	$4.39\text{x}10^9$	0.232	+ 0.9	22.2	+ 0.5
4	EB	0.345	17.4	679	3.47	195	$1.01\text{x}10^{10}$	3.356	+ 3.2	17.4	0
5	NB	0.485	13.5	741	3.66	202	$1.18\text{x}10^{10}$	0.475	— 2.1	13.5	0
6	BB	0.395	19.2	823	3.74	220	$1.75\text{x}10^{10}$	0.377	— 4.6	19.0	— 1.0
7	NT	0.630	13.7	957	4.45	215	$1.80\text{x}10^{10}$	0.554	—10.6	13.1	— 4.4
8	TCE	0.350	20.6	804	3.33	241	$2.25\text{x}10^{10}$	0.362	+ 3.4	21.0	+ 1.9
9	CT	0.350	20.2	903	3.50	258	$3.14\text{x}10^{10}$	0.354	+ 1.1	20.3	+ 0.5
10	CB	0.790	13.1	1142	3:68	310	$7.30\text{x}10^{10}$	0.792	+ 0.3	12.9	— 1.5

Aniline = aniline
TBE = tetrabromoethane
DBE = dibromoethane
EB = ethyl bromide
NB = nitrobenzene
BB = bromobenzene
NT = *o*-nitrotoluene
TCE = tetrachloroethylene
CT = carbon tetrachloride
CB = chlorobenzene

TABLE 3.—Re AND C_D AT PEAK TERMINAL VELOCITIES

$$(Re)_p = 2.92\ P^{0.238}$$

$$(C_D)_p = 1.66\ P^{-0.038}$$

	System	Re*, calc.	Re*, obs.	Dev., %	C_D, calc.	C_D, obs.	Dev., %
1	Tetrabromoethane.....	550	580	— 5	0.719	0.762	—5.6
2	Dibromoethane........	580	570	+ 2	0.714	0.715	—0.1
3	Ethyl bromide.........	700	680	+ 3	0.692	0.672	+3.0
4	Nitrobenzene..........	730	740	— 1	0.688	0.689	—0.1
5	Bromobenzene.........	800	820	— 3	0.678	0.690	—1.7
6	*o*-Nitrotoluene.........	810	960	—16	0.677	0.706	—4.1
7	Tetrachloroethylene ...	850	800	+ 6	0.671	0.667	+0.6
8	Carbon tetrachloride...	920	900	+ 2	0.663	0.655	+1.2
9	Chlorobenzene.........	1125	1140	— 1	0.642	0.619	+3.7

*Values rounded off.

drop size gradually increases, the terminal velocity increases linearly with it. This part of the U_o vs. d curve is a straight line and the corresponding C_D values fall along the log C_D vs. log Re curve for solid spheres. Further increase in drop size brings about a zig-zag motion of the drops, and the apparent U_o measured is actually lower than the vectorial velocity value. This phenomenon may be the reason that the C_D stays practically constant and always higher than the C_D for solid spheres up to the abrupt rise. Correspondingly, the U_o vs. d plot starts to show a slight curvature as the peak is approached. When U_o vs. d is plotted on log-log paper for the curved part before the peak, it has been found that U_o varies as the 2/5 to 3/4 power of d, corresponding to the third region in Allen's analysis in which U_o varies as $d^{0.5}$. In other words, d/U_o^2 is approximately constant and thus C_D, being equal to $K(d/U_o^2)$, is almost independent of Re in this region.

The peak terminal velocity is a point of special interest in view of its direct bearing on practical liquid-liquid contacting operations. Data for the peak indicate that the following relationship exists:

$$(Re/We)_p = 0.816\ P^{0.238} \qquad (1)$$

On simplification,

$$(U_o)_p = 1.23\ \left(\frac{\sigma_i}{\mu}\right) P^{-0.238} \qquad (2)$$

Equation (2) shows that the peak terminal velocity of a system is dependent only upon its physical properties. For a rain drop falling in air at 20°C. Equation (2) gives $(U_o)_p = 800$ cm./ sec., which checks closely with reported values (*16*). It is however impossible to find the corresponding drop size by means of Equation (2) alone, because all the d terms are cancelled.

It is interesting to note that the peak of the U_o vs. d curve corresponds to the nose of the log C_D vs. log Re plot, at which the abrupt rise starts. A straight line joining all the nose points indicates very little scattering of the data except for the aniline system, which seems to be out of line with the other nine systems. The equation of this line, shown by the broken line in Figure 1, is found to be

From this equation

$$(C_D)_p = 1.973\ (R_e)_p^{-0.161} \qquad (3)$$

$$(d)_p = \left(\frac{1.51\times10^{-3}}{\Delta\rho}\right)^{0.861}$$

$$\rho^{0.723}\ \mu^{0.139}\, U_o^{1.584} \qquad (4)$$

Equation (4), though capable of giving good results, is rather inconvenient to use because of the fractional powers involved. A much simpler method has been found by plotting log We vs. log Re. Parallel straight lines with a constant slope of about 1.4 were obtained; however, these straight lines started to level off within a small We range of 3.33 to 3.68. It was found that these We values also corresponded to the respective peaks and an average of 3.58 seemed to be a good representation for all the systems. Thus

$$(We)_p = 3.58 \qquad (5)$$

which agrees quite well with the value of 3.65 reported by Peebles and Garber (*23a*) for gas bubbles in liquids.

When $(d)_p$ is solved for,

$$(d)_p = 3.58\ \left(\frac{\sigma_i}{\rho}\right)/\left(U_o^2\right)_p \qquad (6)$$

Values of $(U_o)_p$ and $(d)_p$ for all the systems, calculated by Equa-

TABLE 4.—CRITICAL DIAMETERS

$$a_c = \left[1.452 \times 10^{-2} \left(\frac{\sigma_i}{\Delta\rho}\right)\right]^{1/2}$$

	System	$\Delta\rho$	σ_i	$(d_c)_{calc}$, cm.	$(d_c)_{obs}$, cm.	Dev., %
1	Aniline	0.0197	2.8	1.436		
2	Tetrabromoethane	1.9501	35.9	0.517	0.511	+ 1.2
3	Dibromoethane	1.1575	31.9	0.633	0.674	— 6.1
4	Ethyl bromide	0.4501	30.0	0.984	0.914	+ 7.7
5	Nitrobenzene	0.1975	24.1	1.331	1.537	—13.4
6	Bromobenzene	0.4910	37.9	1.059	1.132	— 6.4
7	*o*-Nitrotoluene	0.1606	26.5	1.548		
8	Tetrachloroethylene	0.6173	44.4	1.022	1.040	— 1.7
9	Carbon tetrachloride	0.5813	40.6	1.007	1.040	— 3.6
10	Chlorobenzene	0.1026	37.4	2.301		
11*	Dirty water	0.980	48.0	0.70	0.84	+20.0
12*	Distilled water	0.980	70.6	1.00	1.02	+ 0.2
13*	Carbon tetrachloride	1.568	24.5	0.36	0.48	+33.3
14*	Methyl salicylate	1.303	34.3	0.65	0.62	— 4.6
15*	Methyl salicylate (thickened)	1.313	37.2	0.85	0.64	—24.7
16*	Glycerine+2% Water	1.186	63.7	0.90	0.88	— 2.2

*Data by Merrington and Richardson on liquid drops in air.

tions (2) and (6), are found in Table 2. However a more direct method of estimating the nose C_D and *Re* is by substituting Equation (5) into Equation (1) and eliminating *Re* from Equation (3). Thus,

$$(Re)_p = 2.92\ P^{0.238} \qquad (7)$$

$$(C_D)_p = 1.661\ P^{-0.038} \qquad (8)$$

Values of $(Re)_p$ and $(C_D)_p$ are found in Table 3.

The abrupt rise in the log C_D vs. log *Re* curve corresponds to the dip after the peak in the U_o vs. *d* plot. Vigorous oscillation has been observed in the falling drops, and the deformation in drop shape may or may not be noticeable, depending on the relative magnitudes of the interfacial tension and the density difference of the system concerned. This indicates that the oscillation induced by the peak velocity is the main reason for the rapid increase in C_D and the corresponding decrease in U_o within this short *Re* range. The effect of deformation seems to be only of secondary importance.

After the abrupt rise in the log C_D vs. log *Re* curve, deformation of the drops becomes more and more severe and the oscillation progressively erratic. This part of the curve has a fairly constant slope which later increases slightly owing to the excessive deformation of the drops; however, the family of curves for different systems exhibits a great extent of intercrossing with one another in this region, even though the same general pattern is retained. Different methods have been tried to disentangle the data and the best solution found after many trials is a log $C_D \cdot We\ P^{0.15}$ vs. log $(Re/P^{0.15})$ plot, which surprisingly enough is capable of bringing all the data into one single curve for the entire *Re* range covered in this study, the aniline system being again excluded. This generalized curve, shown in Figure 3, has a slight curvature in the low *Re* region and shows a distinct break at an *Re* region approximately corresponding to that where the peak terminal velocity occurs. When the pertinent physical properties of a system are known, this curve can be used directly to obtain the terminal velocity for any given drop size and hence the corresponding C_D, *We*, and *Re* are readily calculable, involving no trial-and-error solution at all.

That prediction of drop motion is not possible in the absence of such a generalized curve is obvious. In this event, the following two equations representing the generalized curve may be used to advantage:

$$Y = (4/3)\ X^{1.275} \text{ for } 2 < Y \leq 70 \quad (9)$$

$$Y = (0.045)\ X^{2.37} \text{ for } \quad Y \leq 70 \quad (10)$$

where

$$Y = C_D\ We\ P^{0.15}$$

and

$$X = (Re/P^{0.15}) + 0.75$$

These two equations give errors usually well below 10%. Furthermore, at the break point of $Y = 70$ and $X = 22.25$, calculated values of drop size, Reynolds number, and terminal velocity are found to check with the observed peak quantities within 10% with the exception of four occasions. The break point can be therefore safely used to calculate the peak quantities in addition to methods outlined above.

It is interesting to note that the chlorobenzene system, with a low density difference coupled with a comparatively high interfacial tension, behaves almost ideally. Even large drops are only slightly deformed into a symmetrical, ellipsoidal shape and maintain a smooth motion with very little oscillation. On the other hand, the interfacial tension of the aniline system is so low that, despite its very low density difference, even a medium-sized drop will be badly deformed into a rather irregular shape being continually deformed into various queer shapes by the wake turbulence. Furthermore, the system itself is not stable against oxidation and discoloration. In view of these facts, it is not surprising to find the data for this system fall out of line with the rest.

It should be pointed out that the viscosity of the drop liquid seems to have no discernible effects on drop motion. The most that can be said is that, according to the analyses of previous workers (*13, 29*), this viscosity might exert a damping action on the oscillation of the drops. But this effect is difficult to isolate from the much greater effects of deformation and oscillation in gross-motion measurements.

The velocity measurements were discontinued when either the oscillation became too erratic or the deformation too severe, thus rendering the data meaningless. However, for certain systems the experiment had to be terminated at an early stage, long before any severe deformation or oscillation had taken place, because further increase in size would cause splitting of the drops. This phenomenon indicates that, for each system, there is a definite maximum drop size, or critical size, beyond which the falling drops cannot be preserved without being ruptured.

Similar phenomena have been observed in the past for the case of liquid drops falling in air. The analysis made by Hinze (*12*) on the data of Merrington and Richardson (*20*) on drops of six liquids falling in a tower over 125 ft. high suggests a critical *We*, based on the density of the liquids, as the criterion for the critical drop size. Although the *We* values so calculated vary from 7.7 to 14.9 for the six liquids, Hinze concludes that an average of 10 can be used for the characterization.

Applying the previous argument to the systems in which splitting has been observed in the present study, the critical We are found to vary from 10 to 22. Evidently We is not the only dimensionless group that characterizes the critical drop size. From both dimensional analysis and force balances, it has been found that *(12a)*

$$(WeC_D)_c = \text{constant}$$

From the data

$$d_c = \left[1.452 \times 10^{-2} \left(\frac{\sigma_i}{\Delta\rho}\right) \right]^{\frac{1}{2}} \quad (11)$$

Table 4 gives the calculated values of d_c for all the ten systems used in the present work as well as for some air-liquid systems reported in the literature *(20)*.

SUMMARY

Terminal velocity measurements have been made on single drops of ten organic liquids falling in a stationary water field in a glass-walled tank 1 ft. square and 4 ft. high. The drop size ranged from 1.6×10^{-4} to 2.000 mm. in diameter, covering a range of Reynolds numbers from 10 to 2,200.

The aniline-water system has a very low interfacial tension and is very easily oxidized and discolored by contact with air. Results of the other nine systems, however, have been successfully correlated by means of a dimensionless parameter, P, composed of the pertinent physical properties of a system in the following manner:

$$P = \frac{\rho\sigma_i^{3}}{g\mu^{4}} \frac{\rho}{\Delta\rho} = \frac{3}{4} \frac{(Re)^4}{C_D\,(We)^3}$$

which is a constant for each system at constant temperature. It has been found that for the data on all the nine systems (excluding aniline-water) a plot of log $C_D We P^{0.15}$ against log $(Re\,P^{0.15})$ resulted in a single unique curve for the entire range of Reynolds numbers covered. Equations have been obtained for this generalized curve and either the equations or the curve can be used directly to obtain the terminal velocity, drag coefficient, and Reynolds and Weber numbers for any given drop size. The errors involved are generally less than 10%. The curve contains a break point which can be used to predict the peak terminal velocity and its related quantities. Alternate methods for predicting the peak quantities have also been presented.

The maximum, or critical, drop size of the systems is found to be dependent on the criterion

$$(C_D We)_c = \text{a constant}$$

which leads to an equation for estimating the critical drop size from the density difference and interfacial tension of a system.

Work on systems of very low interfacial tension and on systems using field liquids other than water is in progress.

NOTATION

a = constant
c = constant
C_D = drag coefficient
d = diameter of a solid sphere or equivalent diameter of a drop
d_c = critical equivalent drop diameter for maximum drop size
d_p = equivalent drop diameter corresponding to peak terminal velocity
D = diameter of cylindrical container
F = resistance force to motion
Fr = Froude number, $\frac{U_o^2}{gd}$
g = gravitational acceleration, 980.3 cm./sec.^{2} for Chicago
i = constant
K_1 = correction factor for fluidity of moving body
K_w = wall effect correction factors
m, n = constants
M = M group, $\frac{g\mu^4}{\rho\sigma_i^3}$
P = physical property group, $\frac{\rho\sigma_i^3}{g\mu^4} \frac{\rho}{\Delta\rho}$
Re = Reynolds number, $\frac{dU_o\rho}{\mu}$
S = distance of fall required for reaching terminal velocity when starting from rest
t = time
T = temperature
u = velocity
U_o = terminal velocity with no wall effect
We = Weber number, $\frac{U_o^2 d\rho}{\sigma_i}$
μ_o, μ = viscosities of organic and water phase respectively
ν = kinematic viscosity
π = 3.1416
ρ_o, ρ = densities of organic and water phase respectively
$\Delta\rho$ = density difference of a system, $\rho_o - \rho$
σ_i = interfacial tension

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