Fall of Liquid Drops in Water

Terminal Velocities

P. M. KRISHNA[1], D. VENKATESWARLU, and G. S. R. NARASIMHAMURTY[2]
Indian Institute of Technology, Kharagpur, India

Motion of liquid drops in immiscible liquids is important in liquid-liquid extractors, in separators used with distillation columns, and in packed towers when the packing is not wetted by the disperse phase. A knowledge of the factors which influence their motion is essential for the design and evaluation of performance of equipment used in process industries. The terminal velocities of single liquid drops falling under steady-state conditions in a stationary continuous medium of water with no mass transfer are reported.

Twenty-five pure liquids and six mixtures were studied in the following ranges.

Drop diameter, 0.0636 to 4.24 cm.
Drop liquid density, 1.016 to 2.939 grams per cc.
Drop liquid viscosity, 0.653 to 27.06 centipoises
Interfacial tension, 4.13 to 45.67 dynes per cm.
Reynolds number, 2.5 to 4,158

PREVIOUS WORK

The theoretical equations of Hadamard (*4*), Rybzynski (*11*) and Boussinesq (*3*) and the experimental investigations of Bond (*1*), and Bond and Newton (*2*) are limited to the Stokes' law region of Reynolds number less than 1.

The principal investigators on the fall of liquid drops at higher Reynolds numbers are Smirnov and Ruban (*11*), Licht and Narasimhamurty (*10*), and Hu and Kintner (*6*). Smirnov and Ruban's work is limited to Reynolds number less than 1000. Licht and Narasimhamurty studied the terminal velocities of six organic liquids in water covering the drop diameter and Reynolds number ranges of 0.114 to 1.119 cm. and 186 to 1700, respectively. Hu and Kintner determined the terminal velocities of drops of ten organic liquids covering the diameter range of 0.115 to 1.563 cm. and Reynolds range of 10 to 2226. Recently, Klee and Treybal (*9*) reported data on the fall of pentachloroethane drops in water reaching a maximum diameter of 1.2 cm. After completion of this work, "Velocity of Fall of Circulating and Oscillating Drops through Quiscent Liquid Phases" by Johnson and Braida appeared (*8*).

APPARATUS

Test Column. A test column 6 inches square in section and 6 feet in height, shown in Figure 1, is constructed of 1/8-inch transparent plastic sheet (Perspex) with a supporting framework of aluminium angles. The base and the zone of free water surface at the top are protected against attack of organic liquids by the lining of 10-gage copper sheet.

The terminal velocity zone of 100 cm. is marked on opposite sides of the column to note the passage of drops across those marks without parallax error, leaving about 60 cm. from the top for the drops to attain their terminal velocities and about 20 cm. from the bottom to avoid end effects.

Burets. Four special burets with carefully drawn out nozzles and about 35 cm. of graduated scale are constructed for the release of drops. Two of these burets (2 and 3) used for moderate sized drops are provided with horizontal length for the calibrated scale and are similar to those of Licht and Narisimhamurty (*10*). Buret 1, constructed of fine capillary tubes for releasing small drops, and buret 4, of ordinary glass tubing for releasing larger drops, are each provided with a vertical calibrated scale to facilitate manipulation and to avoid meniscus errors, respectively.

The scale portions of the burets are calibrated with pure re-

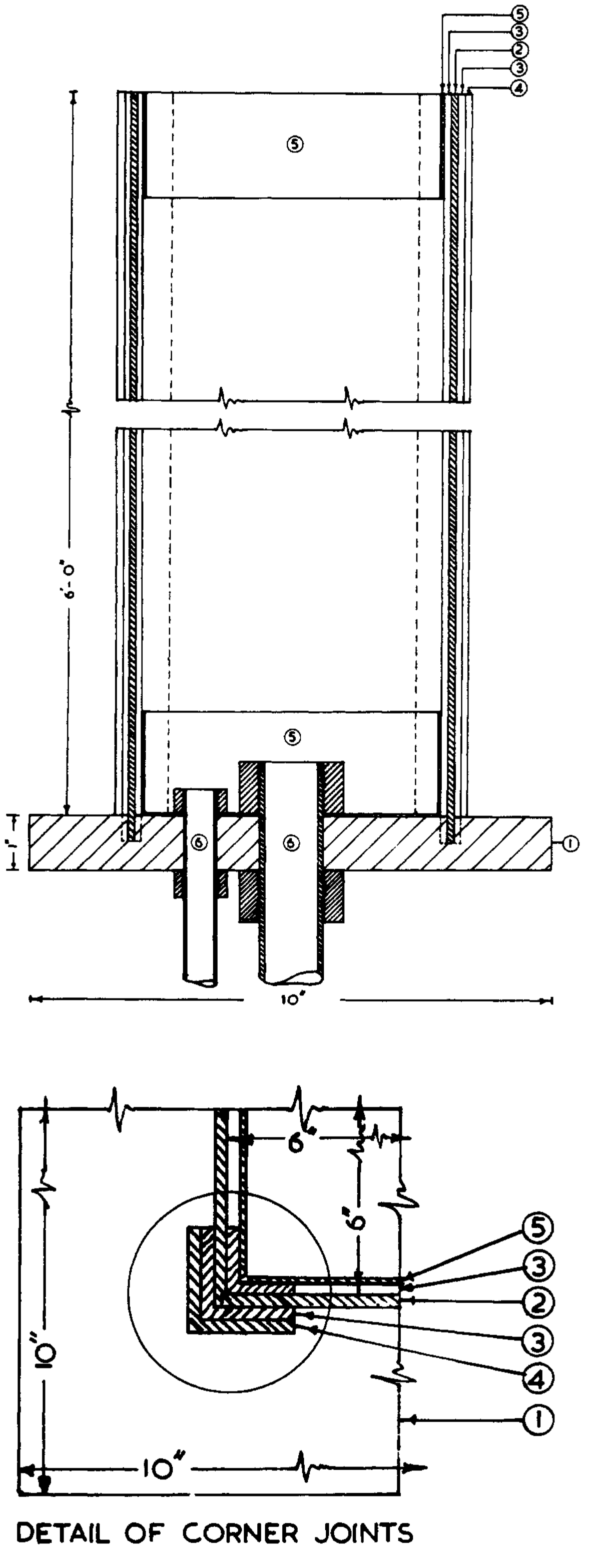

Figure 1. Test column

1. Plastic square plate
2. Plastic plate, 1/8 inch
3. Plastic angles
4. Aluminium angles
5. Copper lining
6. Brass nozzles

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[1] Present address, University of Minnesota, Minneapolis, Minn.
[2] Present address, Indian Institute of Technology, Bombay, India.

distilled mercury. As the maximum deviation for the entire scale of the burets was ±1% of the average values, the following average volumes in milliliters per centimeter length of graduated scale are taken for the different burets: 1, 0.001347; 2, 0.006205; 3, 0.02503; and 4, 0.1033.

LIQUIDS AND THEIR PROPERTIES

Choice of Liquids. Twenty-five organic liquids of laboratory reagent grade and six liquids whose interfacial tensions are lowered to different values by adding a surface active agent (Empilan) are used so that the significance of all the variables involved may be determined. The continuous medium used in all experiments is filtered tap water.

Physical Properties of Liquids. The physical properties of the drop liquids and the continuous medium (Table I) are determined at the same temperature as the terminal velocities: densities of liquids by a 25-cc. density bottle with thermometer pocket and side neck, viscosities with an Ostwald viscometer, interfacial tensions by the drop weight method (*5*). The values of the *Sd*-group, characterizing the liquid systems (*7*) are included in the table.

PROCEDURE

Release of Drops. The burets are filled with the drop liquid by suction applied at the stopcock end avoiding the suction of liquid into or above the stopcock, then taken to the top of the column with the nozzle immersed in the liquid to avoid liquid draining off and hooked in position with the nozzle immersed about 3 cm. below the water surface. Drops of the required size are generated at the nozzle tip by manipulation of the vacuum and released with the help of a platinum wire fused in a glass rod.

Drops larger than those produced by burets are released with the help of clean porcelain hemispherical vessels using a technique similar to that of Hu and Kintner (*6*). The required volume of liquid is dropped into the vessel from a buret or a 10-ml. graduated pipet, under the water surface in the vessel to avoid spreading of liquid on the free water surface in the column. The vessel is immersed to a depth of about 3 cm. under the water surface in the column and tilted carefully releasing the liquid as a single drop.

Determination of Terminal Velocities. The terminal velocities of drops are determined by noting their time of fall through the terminal velocity zone using a stopwatch with an accuracy to ±0.025 second. Fall under steady-state conditions through the timing zone is ensured by noting the times of fall for the first 50 cm. and next 50 cm. separately, of a few selected drops of each system in its entire range. Visual observations on the shape of the drop, path of travel (straight or zigzag), oscillation, and rotation are also recorded.

OBSERVATIONS AND RESULTS

Figure 2 is the terminal velocity plot of chlorobenzene drops. The Broken line gives the terminal velocities of rigid spheres. Figures 3 to 6 give the terminal velocities of some of the liquids, grouped together to determine the effect of physical properties. The terminal velocities of the other liquids are given in Figures 7 and 8. The visual observations and the pertinent features of the terminal velocity data are given in Table II.

The visual observations lead to these conclusions:

1. There is a characteristic limiting size of drop that may be generated from a liquid.
2. For each liquid, there are characteristic ranges of diameters in which the drop is spherical, oblate spheroidal, or indefinable in shape.
3. Drops of some liquids follow a zigzag path in their fall, particularly in some diameter ranges.
4. Drops exhibit oscillations and vibrations about their equilibrium shape, in certain characteristic diameter ranges.
5. Rotation about the minor axis occurs in drops of some liquids.

Table I. Physical Properties of Liquids

Drop Liquid	Temp., °C.	Density of Drop Liquids, ρ_0 (G./Cc.)	Viscosity of Drop Liquid $\mu_0 \times 10^{-2}$ Cps.	Density of Water, ρ(G./Cc.)	Viscosity of Water, $\mu \times 10^{-2}$ Cps.	Interfacial Tension, σ Dynes/Cm.	Density Difference, $\Delta\rho$ (G./Cc.)	Ratio of Viscosities, μ_0/μ	*Sd* Value
n-Amyl phthalate	28	1.016	18.49	0.9981	0.8280	20.20	0.0179	22.33	4214
Aniline	28	1.016	2.835	0.9981	0.8280	6.545	0.0179	3.42	1366
Bromoform	24	2.850	2.127	0.9989	0.9156	40.60	1.8541	2.32	1578
n-Butyl phthalate	23	1.044	15.38	0.9981	0.9499	23.61	0.0449	16.19	3021
Carbon disulfide	23	1.260	0.6531	0.9991	0.9499	45.67	0.2609	0.69	3250
Carbon tetrachloride	24	1.584	1.048	0.9989	0.9156	44.66	0.5851	1.14	2462
Chlorobenzene	28	1.096	0.7861	0.9981	0.8280	36.02	0.0979	0.95	4261
1-Chloronaphthalene	32	1.200	2.289	0.9953	0.7660	41.90	0.2047	2.99	4296
m-Cresol	28	1.028	7.732	0.9981	0.8280	4.134	0.0299	9.34	726.9
Epichlorohydrin	29	1.169	0.9116	0.9975	0.8085	10.98	0.1715	1.128	1111
Ethyl chloroacetate	30	1.134	0.9642	0.9961	0.7848	15.46	0.1379	1.228	1752
Ethyl cinnamate	28	1.042	4.811	0.9981	0.8280	21.68	0.0439	5.81	4122
Ethyl phthalate	21	1.128	10.86	0.9995	0.9759	14.40	0.1285	11.13	1252
1,2-Dibromoethylene	24	2.17	1.752	0.9989	0.9156	36.58	1.1711	1.91	1658
Eugenol	28	1.058	5.43	0.9981	0.8280	12.34	0.0599	6.56	1721
Isoeugenol	23	1.083	27.06	0.9991	0.9499	9.38	0.0839	28.49	974.3
Methyl phthalate	30	1.180	9.383	0.9961	0.7848	12.26	0.1839	11.96	1262
Nitrobenzene	29	1.195	1.512	0.9975	0.8085	24.81	0.1975	1.87	2396
m-Nitrotoluene	22	1.156	2.044	0.9993	0.9594	28.38	0.1567	2.13	2370
o-Nitrotoluene	30	1.153	1.666	0.9961	0.7848	26.03	0.1569	2.12	2826
Diphenyl ether	22	1.067	2.633	0.9961	0.7848	40.80	0.0709	3.35	5773
1,2-Dichloropropene	30	1.146	0.7966	0.9958	0.7850	31.11	0.0502	1.01	3424
1,1,2,2-Tetrabromoethane	29.5	2.939	5.495	0.996	0.7809	33.35	1.943	7.04	1575
1,1,2,2-Tetrachloroethane	28	1.581	1.452	0.9981	0.8280	30.09	0.5829	1.75	1964
Tetrachloroethylene	30	1.609	0.9497	0.9961	0.7848	43.38	0.6129	1.21	2997
n-Amyl phthalate (interfacial tension reduced)	28	1.016	16.38	0.9981	0.8280	7.071	0.0179	19.78	1475
Chlorobenzene (Interfacial tension reduced to 24.54)	28	1.088	0.7877	0.9980	0.8280	24.54	0.090	0.95	2988
Interfacial tension reduced to 19.56	28	1.072	0.7637	0.9980	0.8280	19.56	0.074	0.92	2542
Interfacial tension reduced to 14.07	28	1.072	0.7716	0.9980	0.8280	14.07	0.074	0.932	1829
Interfacial tension reduced to 9.143	28	1.073	0.7843	0.9980	0.8280	9.143	0.075	0.0947	1182
Nitrobenzene (interfacial tension reduced to 15.84)	28	1.157	1.838	0.9981	0.8280	15.84	0.1589	2.22	1596

Table II. Summary of Visual Observations and Terminal Velocity Data

Drop, Liquid	Diameters of Drops, Cm. Maximum	Maximum for spherical shape	Range for oblate spheroidal shape	Range for undefinable shape	Oscillation commences	Rotation commences	Departure from rigid spheres	At peak velocity	Peak Velocity Cm./Sec.	Path of Fall
n-Amyl phthalate	4.24	1.25	1.25–3.0	3.0 –4.24	2.25	. . .	0.80	1.40–1.53	7.70	Straight[a]
Aniline[b]	2.80	0.60	0.60–1.25	1.50–2.80	1.25	2.0	. . .[c]	0.75	6.28	Straight
Bromoform	0.557	0.30	0.30–0.557	. . .	All, >0.35, marked	. . .	0.08	0.19 0.424	24.20 22.91	Drops <0.2 cm. tend to move sideways
n-Butyl phthalate	4.24	0.80	0.80–2.0	2.0 –4.24	Drops near maximum size oscillate slightly and rotate		0.10	1.0	9.80	Straight except near maximum size
Carbon disulfide	1.67	Very small	Up to 1.0	1.0 –1.67	1.25	. . .	0.27	0.58	16.9	Straight
Carbon tetrachloride	1.18	0.60	0.60–1.0	1.0 –1.18	All	. . .	. . .	0.335	19.95	Straight
1-Chloronaphthalene	1.97	0.36	0.36–1.97	. . .	Small drops	. . .	0.20	0.63	15.8	Straight
Chlorobenzene[b]	3.16	0.50	0.50–1.25	1.25–3.16	. . .	. . .	0.20	0.8	13.1	Nearly straight path
m-Cresol	1.79	0.70	0.70–1.25	1.25–1.79	. . .	1.42	>1.25,[d] Continuous deformation	0.62–0.72	5.6	Nearly straight
Epichlorohydrin	1.24	0.40	0.40–0.85	0.85–1.24	0.60	0.85	. . .[e]	0.28	12.3	Straight
Ethyl chloroacetate[f]	1.37	0.30	0.30–0.65	0.65–1.37	>0.45, Considerable	. . .	. . .[g]	0.31	13.56	Zig-zag 0.30–0.35 cm. diam.
Ethyl cinnamate	2.71	1.0	1.0 –2.0	2.0 –2.71	1.4	2.25	0.80	0.86	10.1	Straight
1,2-Dibromoethylene	0.83	0.45	0.45–0.65	0.65–0.83	All, except very small	. . .	0.10	0.24	22.3	Straight
Ethyl phthalate	1.68	0.4	0.4 –1.2	1.2 –1.68	0.65	1.15	0.37	0.57	11.9	Straight, except 0.20–0.30 cm. diam.
Eugenol	1.73	0.40	0.40–1.24	1.24–1.73	0.62	1.60	0.54[h]	0.60	9.3	Straight
Isoeugenol	1.68	0.45	0.45–1.20	1.20–1.68	1.0	1.4	0.23	0.64	9.3[i]	Straight
Methyl phthalate	1.18	0.36	0.36–0.83	0.83–1.18	0.60	0.80	. . .[j]	0.425	12.3	Straight
Nitrobenzene[b]	1.79	0.60	0.60–1.25	1.25–1.79	0.60	1.40	0.30	0.45	13.4	Straight
m-Nitrotoluene	2.12	0.43	0.43–1.25	1.25–2.12	0.66	1.25	0.25[k]	0.485	15.2[i]	Straight
o-Nitrotoluene[b]	2.12	0.46	0.46–1.25	1.25–2.12	0.45	. . .	0.20	0.59	13.3[i]	Straight
Diphenyl ether	3.25	0.90	0.90–2.25	2.25–3.25 >2.0, Continuous deformation	1.40	2.0	0.20	0.92–0.97	12.5	Straight
1,2-Dichloropropylene	2.35	0.60	0.60–1.60	1.60–2.35 >1.8, Continuous distortion	0.75	. . .	0.20	0.59	13.4	Straight
1,1,2,2-Tetrabromoethane[b]	0.55	. . .	. . .	. . .	. . .	. . .	. . .	0.196	25.63	Straight
1,1,2,2-Tetrachloroethane[l]	1.10	0.25	0.25–0.50	0.50–1.10	0.40	. . .	. . .	0.306 0.60	18.35[i] second lower peak	Straight
Tetrachloroethylene[b]	1.27	0.45	0.45–0.80	0.80–1.27	0.90	. . .	0.17	0.35	21.0	Nearly straight

[a] Drops above 3.0 cm. wobble considerably.

[b] Good agreement with earlier published data.

[c] Plot cuts rigid sphere line at 0.15 and 0.84 cm. in diam.

[d] Plot shows considerable departure from rigid spheres line even for small drops.

[e] Plot cuts rigid spheres line at 0.16 and 0.342 cm. in diam.

[f] All drops are colorless at start, but become opaque during fall.

[g] Plot cuts rigid spheres line at 0.17 and 0.42 cm. in diam.

[h] Plot cuts rigid spheres line at 0.35 cm. in diam.

[i] Plot shows tendency to rise towards end of diameter range investigated.

[j] Plot cuts rigid spheres line at 0.35 cm. in diam.

[k] Plot cuts rigid spheres line at 0.52 cm. in diam.

[l] Data do not agree well with earlier published data.

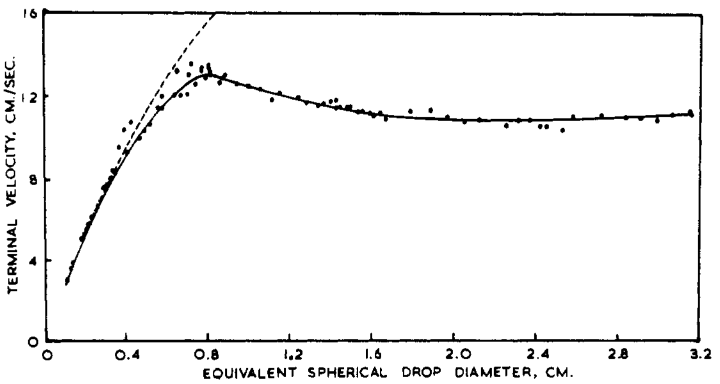

Figure 2. Terminal velocities of chlorobenzene drops in water

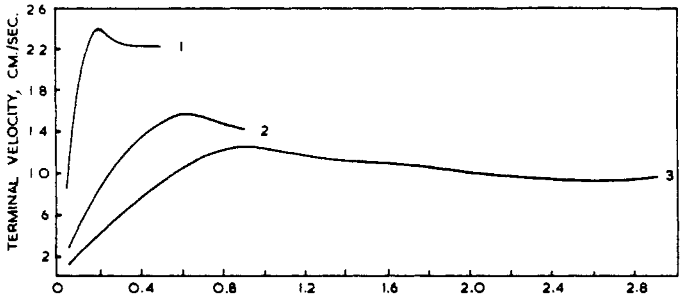

Figure 3. Effect of liquid density on terminal velocity of drops

1. Bromoform. 2. 1-Chloronaphthalene. 3. Diphenyl ether.

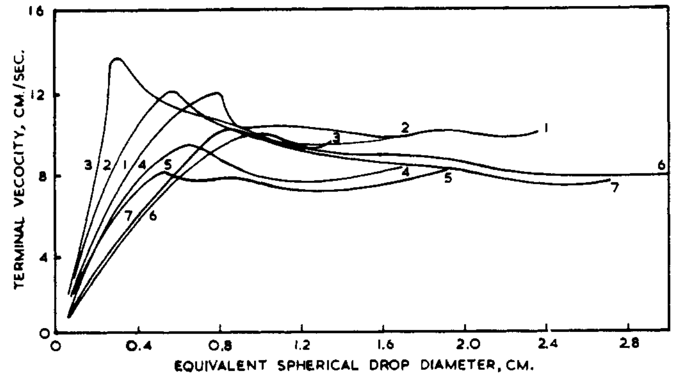

Figure 4. Effect of liquid viscosity on terminal velocity of drops

1. Chlorobenzene, σ = 24.54
2. Ethyl phthalate
3. Ethyl chloroacetate
4. Isoeugenol
5. Chlorobenzene, σ = 9.14
6. *n*-Butyl phthalate
7. Ethyl cinnamate

Study of the terminal velocity data and plots leads to the following conclusions:

1. For most liquids, the terminal velocities of the smaller drops are about the same as those of rigid spheres, but for a few liquids, the curves for rigid spheres and liquid drops are a little apart even for the smaller drops. For some other liquids, the liquid drops curve cuts the rigid spheres curve.

2. The terminal velocities of drops of all liquids increase with increasing drop diameter up to a characteristic maximum or peak velocity. A second lower peak seems to appear in a few cases. For some liquids, the curve has a tendency to go upward at the end of the diameter range. Thus all liquids depart considerably from rigid spheres, except in lower drops sizes.

EFFECT OF PHYSICAL PROPERTIES

Drop Liquid Density. Figure 3 gives the terminal velocity plots of bromoform, 1-chloronaphthalene, and diphenyl ether, which have approximately same viscosities and interfacial tension but differ in density. It may be concluded from this figure and Table II that increased drop liquid density leads to higher terminal velocities, higher peak velocities, lower peak velocity diameters, and lower maximum drop diameters.

Drop Liquid Viscosity. Figure 4 shows the terminal velocity plots of seven liquids which may be grouped such that the liquids have nearly the same density and interfacial tension but widely vary in viscosity. The drop liquid viscosity does not seem to have a significant effect on terminal velocity.

Interfacial Tension. Comparison of the terminal velocity plots of chlorobenzene and chlorobenzene-empilan mixtures of different interfacial tension shows that as interfacial tension increases, the terminal velocity and peak velocity increase, and the peak velocity diameter and maximum drop size decrease.

Physical Property Group, *Sd*. Comparison of terminal velocity plots of liquids, grouped according to *Sd* value and plotted in Figure 6, shows that *Sd* group by itself may not completely characterize the system.

NOMENCLATURE

g = Local acceleration due to gravity, 978 grams per second

g_c = Conversion factor, 980.6 (grams mass) (cm.) per (gram force) (sec.^{2})

Sd = Physical property group,

$$\frac{g_c\sigma}{\mu}\left[\frac{3}{4g}\cdot\frac{\rho^2}{\mu(\rho_0-\rho)}\right]^{1/3}, \text{ dimensionless}$$

ρ = Density of water grams per cc.

ρ_0 = Density of drop liquid, grams per cc.

μ = Viscosity of water, grams per cm. sec.

μ_0 = Viscosity of drop liquid, grams per cm. sec.

$\Delta\rho$ = Difference in densities, $(\rho_0 - \rho)$, grams per cc.

σ = Interfacial tension between drop liquid and water, dynes per cm.

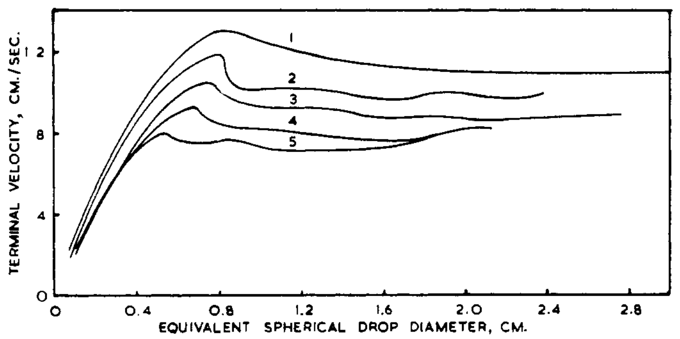

Figure 5. Effect of interfacial tension on terminal velocity of chlorobenzene drops

σ

1. 36.02
2. 24.54
3. 19.56
4. 14.07
5. 9.14

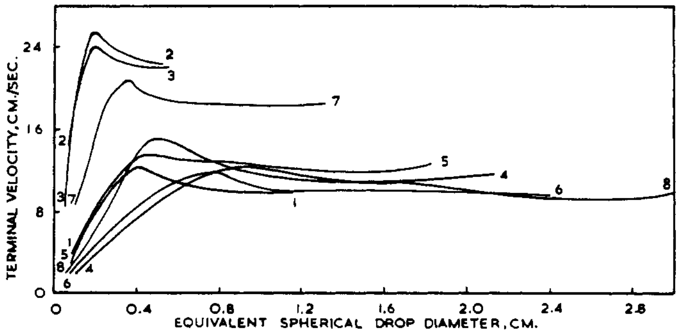

Figure 6. Effect of *Sd* on terminal velocity of drops

	Sd Value		*Sd* Value
1. Methyl phthalate	1262	6. Chlorobenzene (σ = 24.54)	2988
2. 1,1,2,2-Tetrabromethane	1575	7. Tetrachloroethylene	2997
3. Bromoform	1578	8. Diphenyl ether	5773
4. *m*-Nitrotoluene	2370		
5. Nitrobenzene	2396		

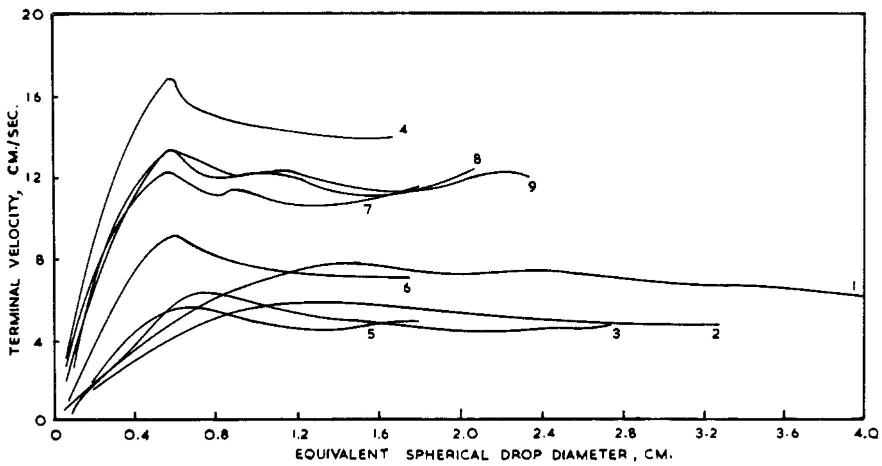

Figure 7. Terminal velocities of liquid drops

1. *n*-Amyl phthalate
2. *n*-Amyl phthalate, $\sigma = 7.07$
3. Aniline
4. Carbon disulfide
5. *m*-Cresol
6. Eugenol
7. Nitrobenzene, $\sigma = 15.84$
8. o-Nitrotoluene
9. 1,2-Dichloropropene

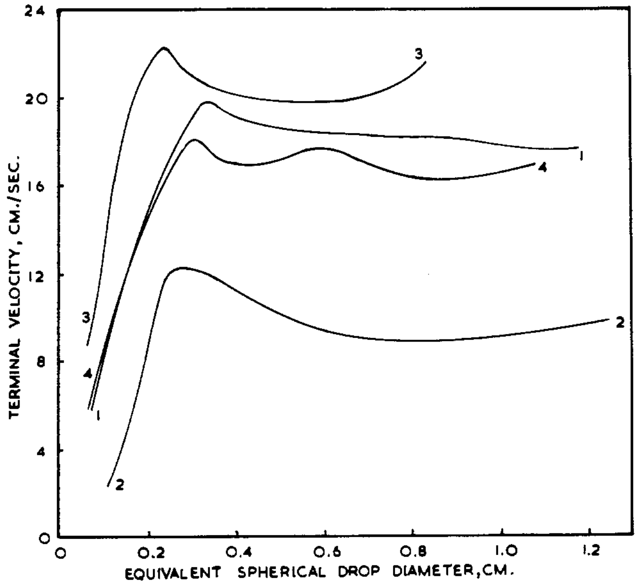

Figure 8. Terminal velocities of liquid drops

1. Carbon tetrachloride	3. 1,2-Dibromoethane
2. Epichlorohydrin	4. 1,1,2,2-Tetrachloroethane

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Fall of Liquid Drops in Water

Drag Coefficients, Peak Velocities, and Maximum Drop Sizes

P. M. KRISHNA[1], D. VENKATESWARLU, and G. S. R. NARASIMHAMURTY[2]
Indian Institute of Technology, Kharagpur, India

The equations for the drag coefficients of rigid spheres given by Stokes (*9*), Oseen (*6*), and Goldstein (*1*) are valid up to Reynolds numbers of about 4. The drag coefficients of rigid spheres and a few other regular shapes at higher Reynolds numbers have been experimentally determined by many investigators (*8*).

Licht and Narasimhamurty (*5*) correlated their data on the fall of liquid drops of diameter greater than the peak velocity diameter by an equation involving C_R and B groups. Hu and Kintner (*2*) correlated their experimental data by an equation involving Re, C, We, and P groups. They correlated the peak velocity and maximum drop size data also. Hughes and Gilliland (*3*) used Wt, Tv, and Sd groups in their study of the mechanics of drops. Peebles and Garber (*7*) used a plot of C *vs.* G in correlating their data on gas bubbles. In their work, Sd group is used as Sd values of liquids changing from 727 to 5773 seem to give a physical sense and to characterise a system. Also, the Sd group is simple to calculate.

Dimensional Analysis. The terminal velocity of a liquid drop in water may be assumed to depend upon a number of variables and is given by the equation,

$$U = f(D, d, \rho_0, \rho, \mu_0, \mu, \sigma, g) \qquad (1)$$

Of these, d may be omitted if wall effect is considered negligible. μ_0 may also be dropped, because terminal velocity studies have shown its effect not to be significant (*4*). A variety of dimensionless groups may be obtained from the other variables, and a suitable combination of these variables gives the equation,

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[1] Present address, University of Minnesota, Minneapolis, Minn.
[2] Present address, Indian Institute of Technology, Bombay, India.