Rate of Fall of Single Liquid Droplets

William Licht and G. S. R. Narasimhamurty, University of Cincinnati, Cincinnati, Ohio

A study was made of the terminal velocities of fall of liquid droplets in another phase liquid under stationary conditions. The studies include dimensional analysis, experimental work for collection of fall-velocity data, and photographic studies of the drop behavior. Six systems were studied for organic liquids insoluble in and heavier than water. The experimental conditions and procedure have been standardized.

For any given system, as the drop size was increased the fall velocities of the droplets increased gradually, reached a maximum, and then fell off asymptotically. Two mechanisms have been postulated, (1) for the range where the fall velocities increased with an increase of drop size and (2) for the range where the fall velocities decreased with an increase of drop size, with the maximum velocity region corresponding to the transition from region 1 and 2.

Although there is considerable information available on the fall velocities of rigid spheres in fluids, information on the motion of liquid droplets in liquids is limited, despite the fact that it is an integral part of liquid-liquid extractions in spray towers. The present problem, which has been simplified so as to lead to a basic knowledge of the hydrodynamics of liquid droplets in liquids, is specifically, terminal velocities of fall of single liquid droplets inside a stationary and infinite medium of another liquid without mass transfer between the phases.

Ailor(*1*), Conway(*3*), Farmer (*4*), Katz(*9*), and Smirnov and Ruban(*13*), observed the fall velocities of liquid droplets in liquids. Ailor used a 1-in.-diam. glass column, and Katz used 2- and 3-in. diam. glass columns and reported wall effect in the 2-in. glass column. The data of Conway and Farmer on fall velocities are more qualitative than quantitative and are also complicated by the presence of mass transfer. It was found in the course of the present work that Tygon tubing, which Katz used for connecting the burrette to the drop-forming tip, lowered the interfacial tension value of the liquids. It will later be shown that a lower interfacial tension lowers the fall velocity of the drop. Only a part of Smirnov and Ruban's work is strictly applicable to the present problem, as for most runs they used a continuous medium in motion. Also an observation of apparatus figures of all the aforementioned workers shows that they used some connecting tubes to join the glass tubes, though they did not clearly state of what materials those tubes were composed. In any case none of them reported any special precautions to prevent the lowering of the interfacial tension values due to contamination with foreign materials.

Spilhaus(*14*) proposed certain general correlations which took into account the air-liquid interfacial tension for the case of rain drops and tested his correlations against the data of Laws(*10*). Hughes and Gilliland(*7*) using a quasithermodynamic approach for drops falling in a gas arrived at results similar to those of Spilhaus (*14*) and tested their correlations against the data of Laws and Watson(*15*) for liquid drops in air. They presented their correlations in the form of a terminal velocity group *Tv* vs. a gravity group *Wt* with a surface-tension-size group *Sd* as parameter, where

$$Tv = V\left[\frac{3}{4}\,\frac{\rho_o^2}{g\,\mu_o\,(\rho_i-\rho_o)}\right]^{\frac{1}{3}} \tag{1}$$

$$Wt = D\left[\frac{4\,g\,\rho_o\,(\rho_i-\rho_o)}{3\,\mu_o^2}\right]^{\frac{1}{3}} \tag{2}$$

$$Sd = \frac{g_c\sigma}{\mu_o}\left[\frac{3}{4}\,\frac{\rho_o^2}{g\,\mu_o\,(\rho_i-\rho_o)}\right]^{\frac{1}{3}} \tag{3}$$

The present results were checked against the correlations of Hughes and Gilliland to indicate the difference in behavior between gas-liquid and liquid-liquid systems. Tentative correlations for liquid-liquid systems were developed by the authors.

DIMENSIONAL ANALYSIS

The drag coefficient of a drop is assumed to depend upon

$$C_D = \varphi\,(\sigma, g, D, \rho_i, \rho_o, \mu_i, \mu_o) \tag{4}$$

where the drag coefficient for steady state motion is

$$C_D = \frac{4\,g\,(\rho_i-\rho_o)\,D}{3\,\rho_o\,v^2} \tag{5}$$

By means of dimensional analysis a variety of possible dimen-

TABLE 1.—PROPERTIES OF LIQUIDS USED

		Carbon tetra chloride	Tetra chloro ethane	*O*-Nitro toluene	Tetra bromo ethane	Ethyl chloro acetate	Chloro benzene
Density, g/ml.	(Exper.)	$1.5842^{25.0}$	$1.5813^{25.0}$	$1.1578^{25.0}$	$2.9539^{25.0}$	$1.1451^{25.0}$	$1.1008^{25.0}$
	(Lit.)	$1.5843^{25.0}$	1.5869^{25}	1.1626^{20}	2.9529^{25}	1.159	1.10085^{25}
Viscosity, centipoises	(Exper.)	$0.9296^{25.0}$	$1.502^{25.0}$	$2.093^{25.0}$	$9.464^{25.0}$	$1.101^{25.0}$	$0.7625^{25.0}$
	(Lit.)	$0.912^{24.6}$	1.456^{20}	2.37^{20}	......	......	0.7585^{25}
Surface tension, dynes/cm.	(Exper.)	$26.9^{24.4}$	$35.0^{24.4}$	$40.8^{25.0}$	$49.6^{25.2}$	$31.1^{27.9}$	$32.5^{25.6}$
	(Lit.)	26.76^{20}	36.04^{20}	41.67^{20}	49.67^{20}	......	33.19^{20}
Interfacial tension, dynes/cm.	(Exper.)	$41.6^{24.0}$	$31.3^{24.9}$	$26.6^{24.2}$	$36.2^{23.2}$	$14.6^{25.0}$	$35.4^{25.4}$
	(Lit.)	45.0 ± 1.0^{20}	......	27.19^{20}	38.82^{20}	$15.8^{25.0}$	37.41^{20}
Solubility, % *W*/*W* (lit.)	(Liq. in water)	0.077^{25}	0.288^{25}	0.0652	0.0651^{30}	Insoluble	$0.0488^{.0}$
	(Water in liquid)	0.010^{24}	......	......	......	......	4.4
μ_i/μ_o		1.040	1.681	2.342	10.59	1.232	0.8533
Sd		2,445	1,843	2,408	1,424	1,359	3,710

G. S. R. Narasimhamurty is at Andhra University, Waltair, India.

sionless correlations may be obtained among these variables. Two which were tested in this work are

$$We = f_1\,(Re, \mu_r) \qquad (6)$$

and

$$\frac{c_D}{c_D^{\,s}} = f_2\,(\beta, Re, \mu_r) \qquad (7)$$

Here *We* represents the Weber number, *Re* the Reynolds number, and $c_D^{\,s}$ the drag coefficient of a rigid sphere of equivalent volume. The two other dimensionless groups used are defined as

$$\beta = \frac{g\,D^2\,(\rho_i - \rho_o)}{4\sigma} \qquad (8)$$

and

$$\mu_r = \frac{\mu_i}{\mu_o} \qquad (9)$$

SHAPE OF THE DROP

Much interest has been evinced in the shape of the drop in motion by Hughes and Gilliland(*7*), Imai (*8*), McDonald(*11*), Saito(*12*), and Spilhaus(*14*). For drops and bubbles one can write

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{P_i - P_o}{\sigma} \qquad (10)$$

This leads to the fundamental differential equation which, when solved and the boundary conditions substituted, should be expected to give the equation for the shape of the drop.

In order to get a reasonable solution of the foregoing equation, the following simplifying assumptions have been made: The drop (1) can attain an equilibrium shape; (2) has no oscillations or rotations; (3) falls vertically in a straight line; and (4) has axial symmetry about the vertical axis. Photographs taken during the present work indicate the extent to which these assumptions were met. In some instances they are reasonably valid, but there are outstanding exceptions which would seem to defy mathematical treatment of this question.

EXPERIMENTAL WORK

The experimental work consisted of (*a*) the preliminary work done to standardize the experimental conditions with a view to getting reproducible and reliable data as well as ascertaining the accuracy of the results obtained, and (*b*) the main experimental work done to collect data on the terminal velocities of fall of liquid droplets as well as photographic studies of the drop behavior.

Preliminary Work The preliminary work comprised the following:

1. Choice of systems.
2. Determination of the physical properties of the liquids.
3. Production of drops of constant volume.
4. Determination of the initial height of fall of drops required to attain terminal velocities.
5. Elimination of wall effect and end effects.
6. Temperature control.

1. Six organic liquids insoluble in and heavier than water were chosen for the discontinuous phase; whereas laboratory-distilled water was selected for the continuous phase. Only research-grade organic liquids were used. The physical properties of the six organic liquids chosen are given in Table 1. The properties of distilled water were taken from literature and used as $\rho = 0.99707$ g./ml. at 25.0°C. and $\mu = 0.8937$ centipoise at 25.0°C. The properties of the organic liquids were determined experimentally, distilled water being used as a reference liquid. The solubility data were taken from literature.

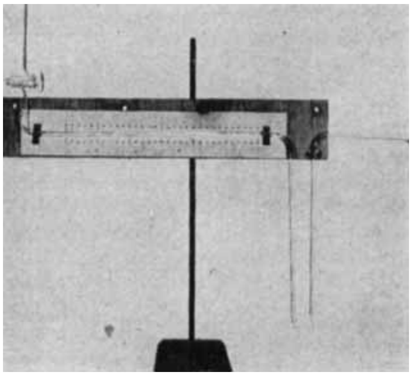

Fig. 1. Capillary *U*-tube microburette.

Carbon tetrachloride was selected first in order that comparisons might be made with previous work on this material. It has a viscosity near that of water. Tetrachloroethane was selected as its density is very near that of carbon tetrachloride, but its interfacial tension with water is different. *O*-Nitrotoluene was selected as it has almost the same *Sd* value as carbon tetrachloride. This was done in order to facilitate comparison with the correlations of Hughes and Gilliland. *S*-Tetrabromoethane was selected because it has a high viscosity and high density, ethyl chloroacetate because it has a very low interfacial tension with water, and chlorobenzene because it has a low viscosity and density.

2. The densities of the organic liquids were determined with a pycnometer at 25.0° ± 0.1°C., with distilled water as a reference liquid. Viscosities were measured at 25.0° ± 0.1°C., with an Ostwald viscometer, distilled water being used as a reference liquid. A Du Noüy tensiometer, by means of the ring method, was used for the measurement of surface tension of the liquids in air and the interfacial tension of the liquids to water. The surface and interfacial tension values read with the instrument were corrected with a factor obtained from the graph of Zuidema and Waters(*16*), and the corrected values were taken as true values.

3. The production of drops of constant volume consisted of developing (*a*) nozzles and (*b*) burettes which would obviate the necessity of using any materials that may dissolve in and change the properties of the organic liquids used.

Nozzles were used to form the liquid droplets at their tips. To start with, hypodermic needles were used. The oblique cut on the end of the needle was removed and the tip rounded off with a file to give a sharp-edged circular tip. Later the range of drop sizes was extended to smaller values by joining glass tips to the metal bases of hypodermic needles with Corroseal solder. By this procedure all the nozzles produced had the same standard Luer grinding of the hypodermic needles.

Burettes were used for measuring the volume of the droplets. Apart from the question of accuracy of measurement, the other important considerations were (*a*) to avoid any lubricants coming in contact with the organic solvents at any time, (*b*) to regulate the rate of flow of liquids to extremely small flow rates, maintaining, if possible, a steady flow rate for any particular setting, and (*c*) to have a tip to which the nozzles could be attached directly without any other connecting tubes. The following two burettes were developed for the purpose.

One burette was a 5- by 1/100-ml. microburette. It was provided with an adjustable-flow stopcock, precision ground so that it might be worked without grease. A standard Luer grinding was provided at the tip of the burette to take the nozzles at the bottom. In order to minimize the effect of liquid-level variation on the drop formation time, experiments were run with the burette filled to zero mark before each run.

The second burette consisted of a 4-ft. Pyrex-glass capillary tube bent to the shape shown in Figure 1. At the upper end of the tube was fused an adjustable-flow stopcock and at the lower end a 1-cc. tuberculin syringe. The stopcock was open to the air, and hence a small amount of grease might be used, thus obviating precision grinding. The horizontal length of the capillary was 40cm., giving 30 cm. for the graduated section. In this burette the 1-mm. length of the capillary tube corresponded to an average volume of 7.23×10^{-4} ml. Because of the horizontal portion of the capillary, this burette gave more uniform drop-formation times than the previous one.

4. Preliminary work showed that drops attained terminal velocities of

fall after an initial height of fall of 30 cm. in water. However, some data were collected on drops after they fell initially through a height of 60 cm., to serve as check points.

5. Data were collected on carbon tetrachloride drops in water in a 3-in.-diam. glass column up to $D = 0.448$ cm. and compared with the data on the same system in a 6-in.-diam. Lucite column, as shown in Figure 3. It can be seen that there is no noticeable difference in the fall-velocity data in the 3- and 6-in.-diam. columns. However for additional confidence and convenience, the 6-in.-diam. column was used for later work.

To avoid end effects, the lowest mark on the column for collecting the fall-velocity data was kept higher than 15 cm. from the bottom of the column.

6. In the early stages of the work the temperatures in the column were maintained at 25.0° ± 0.5°C., but they were later maintained at 25.0° ± 0.1°C. by a thermostatic water jacket in order to give added confidence to the results obtained.

Main Experimental Work

Apparatus. An oblique view of the apparatus is shown in Figure 2. It consisted of a 6-in.-diam. by 6-ft.-long Lucite column with the top end open and the bottom closed with ½-in.-thick Lucite sheet. One end of a ¼-in. brass pipe was screwed into the Lucite sheet and the other end connected to a petcock to serve as a drain.

One mark was placed on the column 3 cm. from the top, which corresponded to the position of the drop-forming tip. Three more marks were placed on the column at 30, 60, and 160 cm. below the drop-formation point. The last mark was 19.9 cm., from the base of the column. In the photograph these marks are indicated by the upper edge of the white bands around the outside.

Mirrors were placed at the 160-cm. mark so that it might be viewed at the 30-cm. level. This not only served as a convenience but also helped in avoiding any vibrations in the column arising from the operator's moving his position to take the readings. The 160-cm. mark at the bottom was generally illuminated by an electric light placed about 1 meter away.

As Lucite was affected by the organic solvents used in the present work, an 800-ml. beaker of glass was placed at the bottom of the column to collect the organic liquid droplets as they fell. At the end of the day's work the beaker could be removed with the help of copper wires tied to it. The fall-velocity data were checked with and without the beaker and the copper wires, but no difference could be found.

The column proper was surrounded by a square jacket (made of sheets of Lucite) in which water circulated for temperature control. The rest of the apparatus consisted of standard equipment for this control by which the temperature could be maintained at 25.0° ± 0.1°C.

Procedure. The following procedure was adopted as the one giving the most consistent and reliable results.

As will become evident in a later part of the present work, interfacial tension is a very important factor controlling the rate of fall of liquid droplets. Hence absolute cleanliness is of great importance in the work.

After all the apparatus had been cleaned, an 800-ml. beaker was lowered into the 6-in.-diam. Lucite column with the help of copper wires tied to it. The drain cock at the bottom of the column was closed and the column was then filled with distilled water of temperature 25.0° ± 0.1°C. The thermoregulator was turned on and the temperature of water circulating in the jacket outside the 6-in. column was adjusted to 25.0° ± 0.1°C.

The cleaned hypodermic or glass nozzles were attached to tips of the burettes provided with the standard Luer grinding. The capillary U-tube burette was used invariably for the small-sized drops, and the 5 × 1/100 ml. burette was used for the larger sized drops. As a check a certain number of medium-sized drops were obtained with both the burettes. The appropriate burette, filled with the organic liquid, was mounted on top of the column with the tip of the nozzle submerged in water at the 3-cm. mark. Care was taken to center the tip in the column. Also, the column was maintained in the vertical position by being checked occasionally with a plumb bob and the necessary adjustments made.

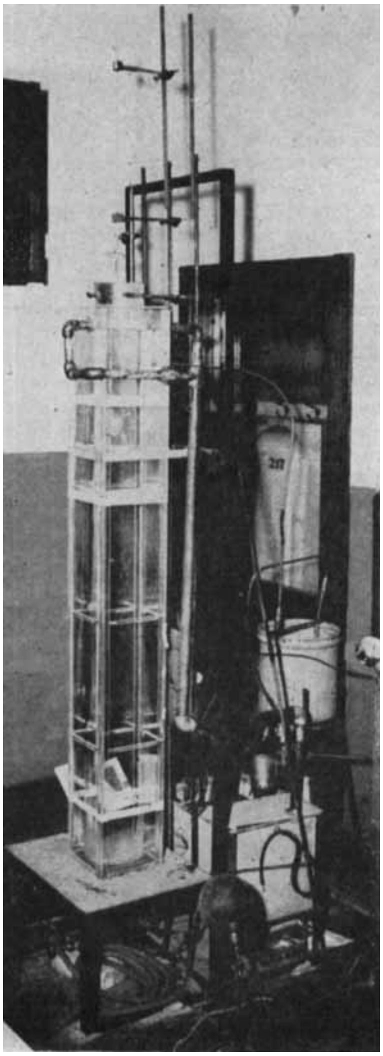

Fig. 2. Apparatus for study of fall of liquid droplets.

The top and bottom temperatures of the water in the column were taken. Generally about 1 hr. after the column was first filled with water, or ½ hr. after the column reached the equilibrium temperature of 25.0° ± 0.1°C., the data were collected.

The burette stopcock was opened slightly and the rate of flow of the liquid adjusted to give approximately the desired drop-formation time. Drop-formation times were chosen to be greater than 30 sec. with greater times for the larger sized drops. The frequency of the drops used in the present work was such that enough time was given for the turbulence in the wake of the previous drop to die down before the next drop fell, so that distortion of the motion of the next drop was avoided. Also, the rate of flow from the burette should be slow enough so that the readings of the liquid levels in the burette might be taken with negligible error. After the rate of flow was adjusted, the volume of liquid run down for a known number of drops was noted. Simultaneously the times of fall of as many drops as possible from 30- to 160-cm. marks were noted. From these values the average drop volume and the average time of fall of the drops were obtained and the fall velocity calculated. During each run the time of formation of every fifth drop was generally noted. In the tables of data* only the lowest and highest drop-formation times were noted. For each run the initial and final temperatures of water at top of the column were recorded.

All timing was done with stop watches manually operated and calibrated against standard time signals to an agreement within less than 1/10 sec. in 10 min. The error in timing would at most amount to ± 1/10 sec. in 5. sec., but in most runs was much less.

The two phases used were not saturated with one another before the runs. As the mutual solubilities were in all cases very low and as no difference in results were noted either with variation in drop-formation time or with the use of the same body of water for an entire day it was assumed that no appreciable error was introduced.

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*For tables of complete velocity vs. diameter data order document 4655 from the Photo duplication Service, American Documentation Institute, Library of Congress, Washington 25, D.C., remitting $1.75 for microfilm or $2.50 for photoprints.

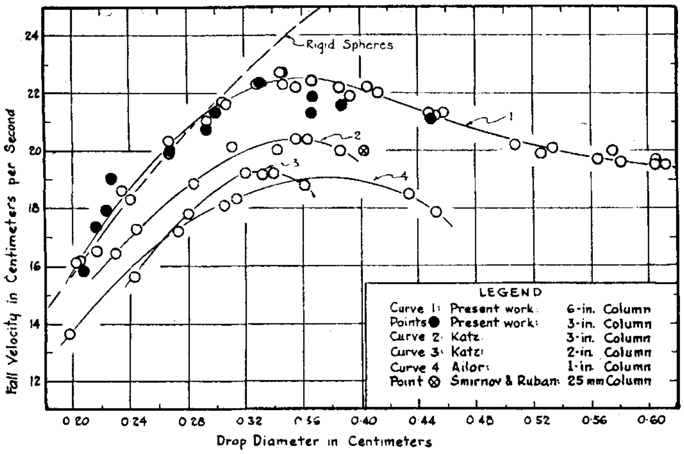


Fig. 3 Fall velocities of carbon tetrachloride drops in water.

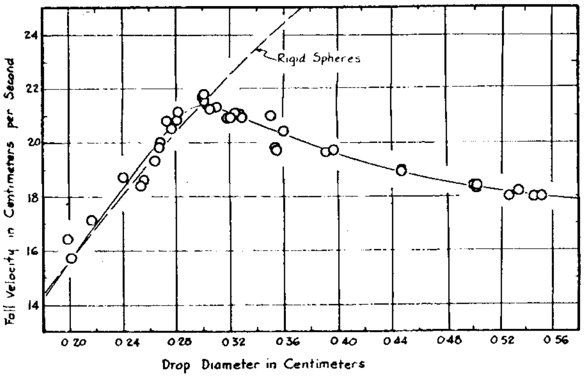


Fig. 4. Fall velocities of tetrachloroethane drops in water.

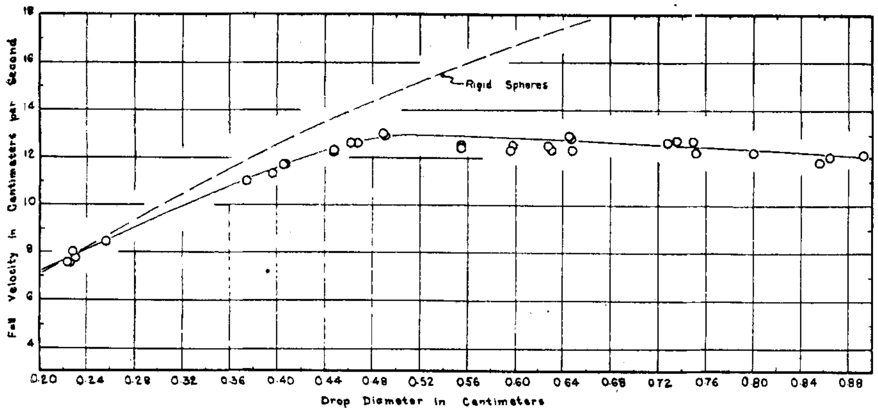


Fig. 5. Fall velocities of *O*-nitrotoluene drops in water.

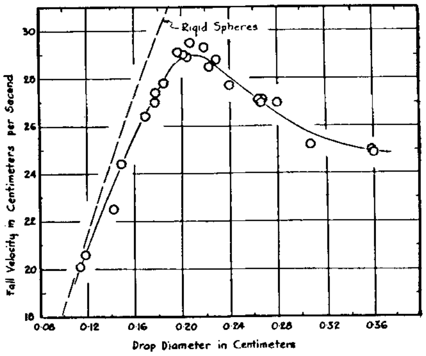


Fig. 6. Fall velocities of *s*-tetrabromoethane drops in water.

RESULTS

The data obtained on the terminal velocities of fall of the different liquid droplets in water are shown on Figures 3 to 8 as experimental points with solid smoothed curves drawn through them. In addition calculated fall-velocity curves for rigid spheres of the same density and volume are shown as dashed lines. Figures 3, for carbon tetrachloride, shows data obtained in both the 3- and 6-in. columns, the smooth curve being only for the 6-in. column. The data of Ailor(*1*), Katz(*9*), and Smirnov and Ruban(*13*) for carbon tetrachloride are also shown for comparison.

The range of drop sizes used for the different liquids was limited by the following considerations. (1) The nozzles used for producing very small drops were very fragile. Therefore the lower limit of drop sizes was fixed when the drops fell with almost the same velocity as rigid spheres. In the case of chlorobenzene drops, data could not be collected on the smaller size drops produced, as they drifted greatly and bumped as many as three to

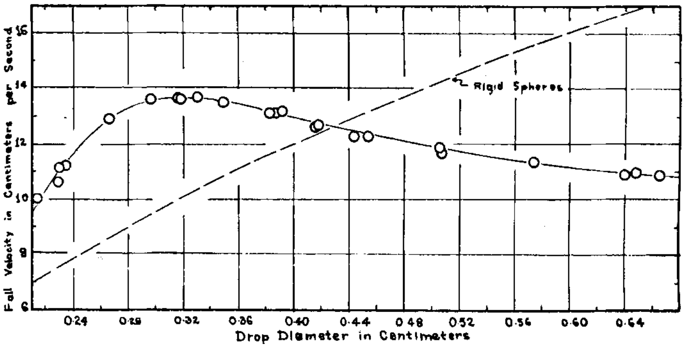


Fig. 7. Fall velocities of ethyl chloroacetate drops in water.

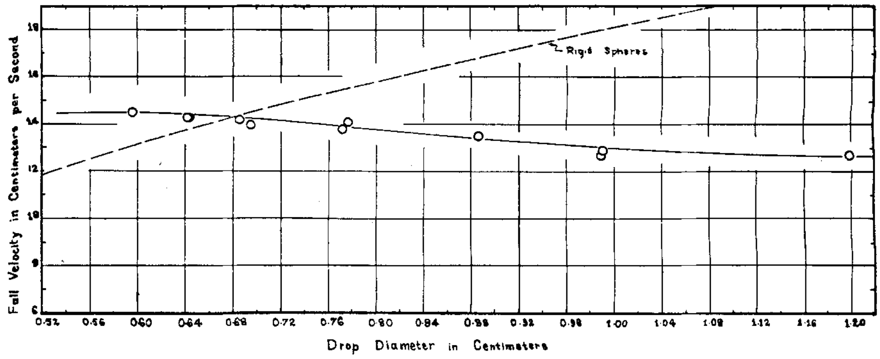


Fig. 8. Fall velocities of chlorobenzene drops in water.

four times on the walls of the 6-in. column in a travel of about 1 meter.* (2) There is a maximum limit to the size of a stable drop. Whether that upper limit was reached in the present work may be questioned, but the maximum drop sizes obtained in the present work can be claimed to be near the upper size limits for stable drops.

The range of variables studied in this work are

$(\rho_i - \rho_o) = 0.1037$ to 1.9568 g./ml.
$D = 0.114$ to 1.119 cm.
$\mu_i/\mu_o = 0.8533$ to 10.59
$\sigma = 14.6$ to 41.6 dynes/cm.

PHOTOGRAPHIC STUDIES

During the course of the work it was felt advisable to record photographically certain observations made visually with a view to explaining the nature of the fall-velocity curves.

Since the drop had to be photographed in motion, a very short exposure time was necessary. As there was also the question of drop vibrations, it was felt that multiple images of the same drop should be taken at small intervals of time. A Strobolux working in conjunction with a Strobotac, producing high-intensity flashes of a duration of 15 to 50 μsec. at frequencies adjustable from 1 to 6,000/min. was used for the work. A 4- by 5-in. Crown graphic camera with a 135-mm. lens was used for taking the picture. A suitable emulsion was found to be Eastman Kodak Super XX.

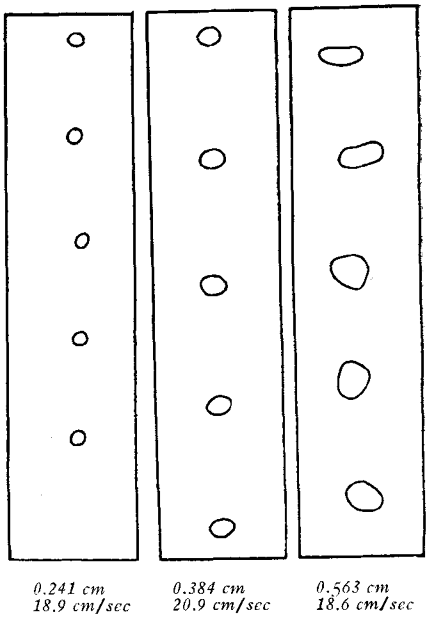
In order to take the pictures of drops, the same procedure as for the determination of fall velocities was followed, and the drops falling in the center of the column were photographed. A Strobolux frequency of 10 flashes/sec. was found to give satisfactory results with a camera lens opening of $f/11$. A ½-sec. exposure resulting in four or five images of the same drop on the plate was usually found to give good results. Dichlorofluorescein was added to some of the liquids used for producing the drops in order to increase the reflectivity of the drops. Since this addition changes the properties of the drop, the photographic data should be considered as qualitative rather than quantitative. Drop sizes and velocities were generally noted for each of the drops photographed. Finally the pictures of the drops thus obtained were enlarged and are shown in Figures 9 through 13.

The poor contrast in some of these photographs makes it difficult to see the drop behavior. Accordingly tracings were made from the prints of Figures 9, 11, and 12 and are reproduced here.

DISCUSSION OF RESULTS

Figure 3 shows that the fall-velocity data obtained in the present work on carbon tetrachloride drops in water are definitely higher than those reported by the previous workers. This is no doubt due partially to wall effects present in the data obtained with 1- and 2-in. columns. But the lower values reported by Katz (*9*) for a 3-in. column cannot be so explained as the technique was generally the same as in the present work. The disagreement is attributed to contamination of the carbon tetrachloride by stopcock lubricants which lowered the interfacial tension in Katz's experiment. In the authors' work special care was taken to avoid this, as explained above.

Comparison of Figures 3 and 4 shows the fall velocities of tetrachloroethane drops to be lower than those of carbon tetrachloride drops, especially in the larger drop sizes, though their densities are very nearly the same, being 1.5813 and 1.5842 g./ml., respectively, at 25.0°C. The difference in $(\rho_i - \rho_o)$ is only 0.5% and rigid-sphere fall-velocity curves are virtually the same in both figures, and so the difference in fall-velocity curves cannot be thereby explained. The viscosity ratio $(\mu_i/\mu_o) = 1.681$ for tetrachloroethane and 1.040 for carbon tetrachloride. The influence of viscosity, if any, should be to make the drop behave more like a rigid sphere with an increase of viscosity of the drop. Hence the viscosity increase cannot explain the lowering of the fall velocities of tetrachcloroethane drops. The interfacial tension of tetrachloroethane is only 31.3 dynes/cm., compared with 41.6 dynes/cm. for carbon tetrachloride drops. It seems appropriate to assign the responsibility for the lower fall velocities on the lower interfacial tension. It is also consistent, since the rigid spheres with infinite interfacial tension fall faster than the drops in the same region.



0.241 cm
18.9 cm/sec

0.384 cm
20.9 cm/sec

0.563 cm
18.6 cm/sec

Fig. 9. Carbon tetrachloride and dichlorofluorescein drops in water. →

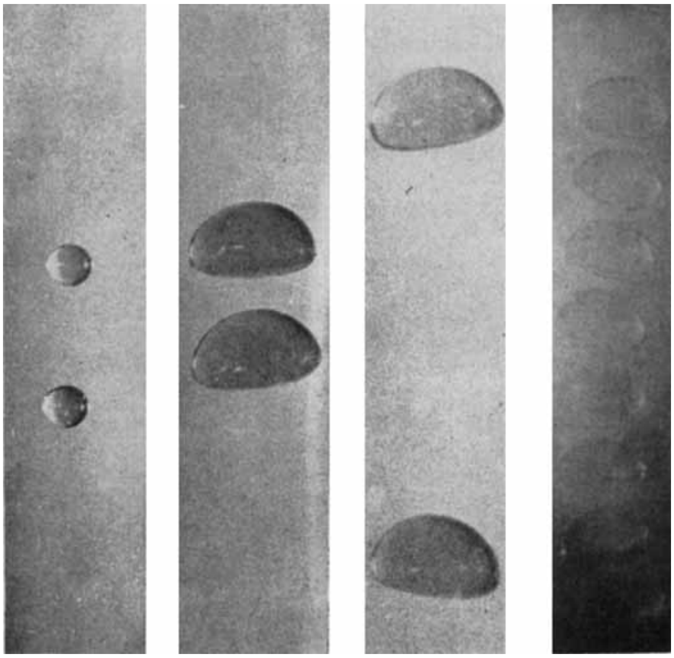


Fig. 10. *O*-Nitrotoluene drops in water. ↓

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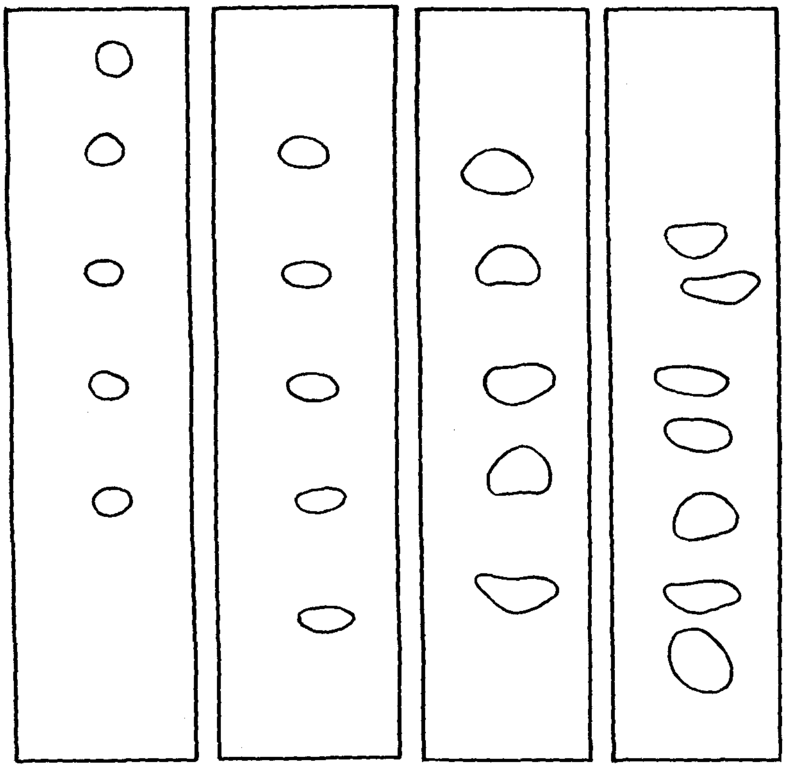
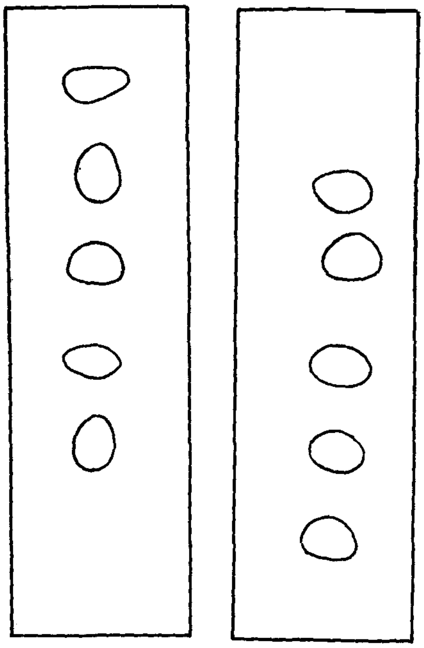
*This may have been due to the effect of convection currents, as the density difference $(\rho_i - \rho_o)$—is very small for this system.



0.399 cm 12.3 cm/sec. 0.399 cm 12.3 cm/sec. 0.633 cm 11.1 cm/sec. 0.633 cm 11.1 cm/sec.

Fig. 11. Ethyl chloroacetate drops in water.



0.638 cm 0.638 cm

Fig. 12. Ethyl chloroacetate and dichlorofluorescein drops in water.

Figures 3 to 8 show that the fall velocities of liquid drops increase first, reach a maximum, and then fall off asymptotically as the drop size is continuously increased. This behavior of liquid drops contradicts that of rigid spheres. Hence it will be discussed in detail in the light of both the experimental and photographic results.

In the smaller range of drop sizes, the fall velocities of some liquid droplets are greater than those of rigid spheres over a certain range of drop sizes. This criss crossing of the fall-velocity curves of drops and rigid spheres is significant in the case of ethyl chloroacetate and chlorobenzene, which are both characterized by a combination of low values for $(\rho_i - \rho_o)$ and (μ_i/μ_o), but not in the case of the other systems. It has been proposed by various workers that internal circulation with the drop will cause the fall velocity to become greater than for rigid spheres.

In the larger sizes all drops fell with velocities lower than those of rigid spheres. Since drops are deformable, they deviate from the shape of a rigid sphere, and the total surface area of the drop increases and naturally gives rise to a higher drag coefficient. The different shapes of drops given in Figures 9 to 13 will bring out this point clearly.

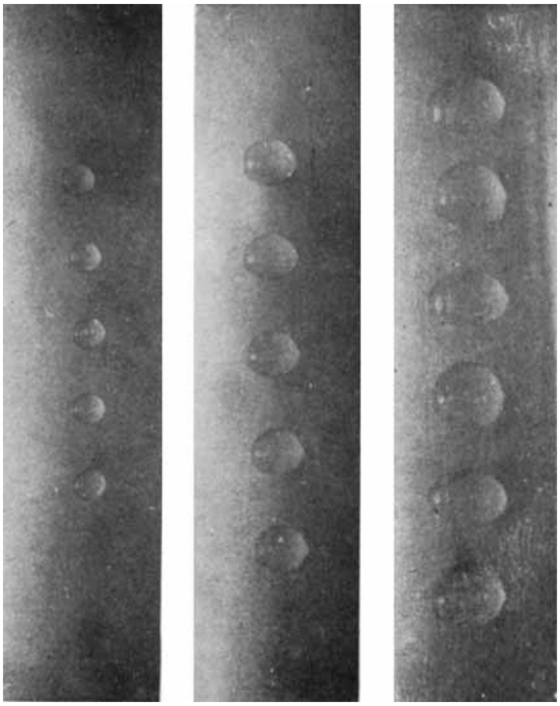


Fig. 13. Chlorobenzene and dichlorofluorescein drops in water.

From a study of the photographs the following observations can be made:

1. The smaller size drops are less deformed than the larger ones and are more nearly spherical.
2. Some of the deformed drops in the larger sizes show vibrations in shape, which were also observed by Blanchard (*2*) in the case of water drops in air.
3. The drops with an equilibrium shape fall with a uniform velocity even when the time intervals taken for the determination of the fall velocity are small.
4. The drops with vibrations show unequal velocities of fall when the time intervals are reduced, as in the

present work, to say 1/10 sec. This can clearly be seen from the unequal distances between consecutive images in the photographs where the drops showed vibrations in shape.

5. Several drops fall in a path which is neither strictly vertical nor even a straight line. In fact, visual observation shows them to fall in a zig-zag path. It was visually observed that as the drop sizes were gradually increased the smallest drops fell nearly vertically, and for the larger sizes the deviation from the vertical path increased to a maximum (when it was even impossible to collect data as the drops fell on the walls of the container), and then the deviation decreased with further increase of drop sizes, though the deviation did not die down completely. The maximum drifting of the drops as noted above may correspond to the case where the natural frequency of the drop coincides with the frequency of the eddies formed in the wake, as referred to by Gunn(*6*) in the case of water drops in air.

6. In the case of several drops there is no axial symmetry in the shape of the drop.

With the experimental and photographic results combined, the fall-velocity curve for each of the systems studied can be divided into two regions: (*a*) for drop sizes smaller than those corresponding to the maximum fall-velocity and (*b*) for the larger drop sizes. Region (*a*) is characterized by (1) an increase of fall velocity with an increase of drop size and (2) the existence of an equilibrium shape of the drop, though the drop may be vibrating as a whole. Region (*b*) is characterized by (1) a decrease of fall velocity with an increase of drop size and (2) the nonexistence of an equilibrium shape. The range of maximum fall velocities seems to correspond to the transition from region (*a*) to (*b*).

The fall velocity values taken from the smooth curves for drops in Figures 3 to 8, and the experimentally determined properties of liquids as given in Table 1, are used for further calculations.

In Figure 14 the drag coefficients of liquid drops were plotted against Reynolds number. For purposes of comparison, the drag coefficients of rigid spheres were shown in the figure with a broken line. It can be seen that the drag coefficients of liquid drops reach a minimum and then climb up steeply with increasing Reynolds number. An attempt was made to see whether the C_D vs. R_e plot could not be redrawn with Weber number as parameter, but it did not prove to be successful.

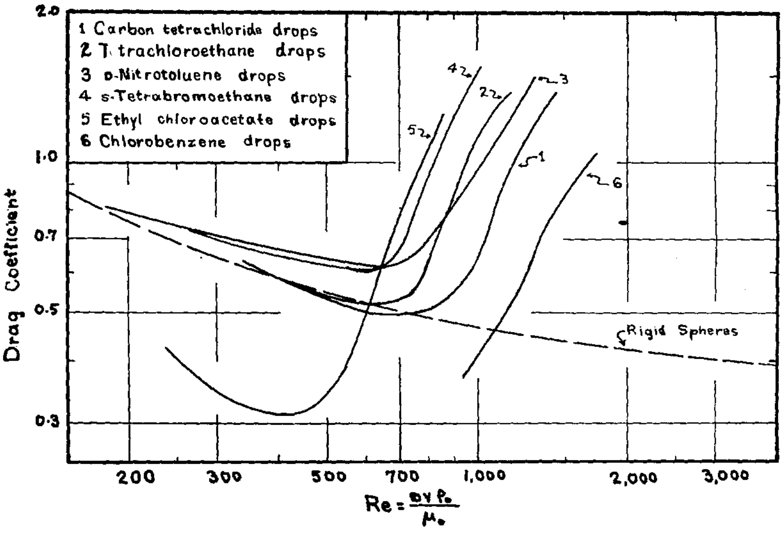


Fig. 14. Drag coefficients of liquid droplets.

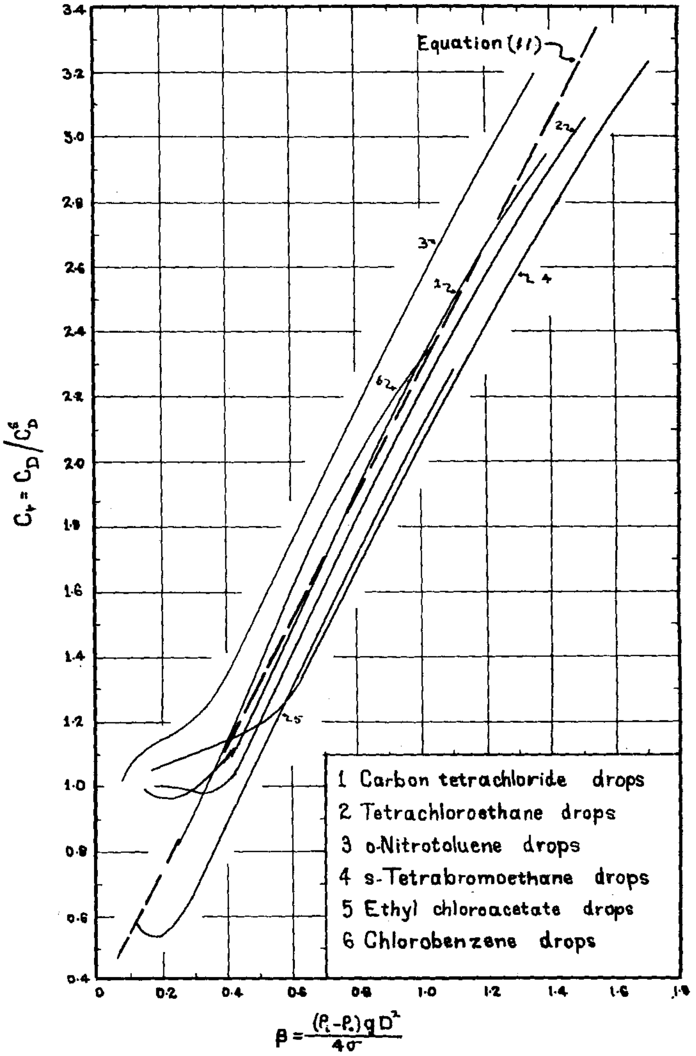


Fig. 15. Correlation of C_r with β group.

The data were also checked against the correlations of Hughes and Gilliland(*7*) in terms of the groups *Tv*, *Wt*, and *Sd* as given by Equations (1), (2), and (3). Poor agreement was obtained, which ought not to have been unexpected. In developing their correlations Hughes and Gilliland had to assume that the density ratio and viscosity ratio were unimportant. This is reasonably true for liquid drops falling through gases but not through liquids.

The Weber numbers of falling drops were plotted against Reynolds number on logarithmic coordinates in an effort to check the correlation suggested by Equation (6). A family of parallel straight lines was obtained below Reynolds number of 700, but they did not correlate with the viscosity ratio. This type of plot is unreliable in any case because both the major variables, diameter and velocity, appear in both groups.

In Figure 15 the ratio of the drag coefficient of a liquid drop to that of a rigid sphere of the same volume and density properties and the same fall velocity as the droplet is plotted vs. the dimensionless group β. In the larger drop sizes, as represented by higher values of β, the curves are nearly linear on a rectangular coordinate plot. The dotted line represents the mean of all the six curves, thus giving a relationship of

$$C_r = \left(\frac{C_D}{C_D^s}\right)_{Re} = 1.87\,\beta + 0.425 \tag{11}$$

Equation (11) may be used with reasonable confidence for the case of liquid drops in water for the range of $\beta \geqq 0.4$.

In this correlation the influence of μ_r seems to be negligible. It may need further refinement and accuracy as well as a more extensive range of experimental conditions to determine exactly the role played by the viscosity of the discontinuous phase.

CONCLUSIONS

1. In addition to the four properties of diameter and density of the discontinuous phase as well as the density and viscosity of the continuous medium, which are important in the case of motion of rigid spheres in fluids, the liquid drops falling in liquids are also influenced by the interfacial tension property. A lower interfacial tension lowers the fall velocities of the drops.
2. For any given system the liquid drops falling in another liquid are nearly spherical in smaller sizes, and their deformation increases with an increase of drop size.
3. Large-sized drops show vibrations in their shape and do not have any equilibrium shape.
4. Drops showing an equilibrium shape fall with a uniform velocity irrespective of the time interval chosen for the measurement of terminal velocity. Drops exhibiting vibrations in their shape do not fall with a uniform velocity if the time interval chosen for the measurement of velocity is small, but they fall with a uniform average terminal velocity if the time interval chosen is large enough.
5. For any given system as the drop size is increased the fall velocity of the drop first increases to a maximum and then falls off asymptotically.
6. The foregoing behavior of drops has been divided into two regions, (*a*) for drop sizes smaller than those corresponding to the maximum fall velocity and (*b*) for drop sizes larger than those corresponding to the maximum fall velocity, with the maximum-fall-velocity region corresponding to the transition from region (*a*) to (*b*).

In region (*b*), above $\beta = 0.4$ the behavior of all liquid droplets insoluble in and falling through water can be represented by $(C_D/C_D^s)_{Re} = 1.87\,\beta + 0.425$.

OTHER WORK

As this manuscript was being prepared, a similar piece of work by Hu and Kintner was reported (*17*). Four of the liquids used were the same in both studies: carbon tetrachloride. *O*-nitrotoluene, tetrabromoethane, and chlorobenzene. A comparison of data indicates that in the present work higher fall velocities were found in each of these four cases. The difference amounts to roughly 10% and may be due to a difference either in the methods of timing employed or in properties of the materials used.

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NOTATION

C_D = drag coefficient, Equation (5), dimensionless
$C_D{}^s$ = drag coefficient of a rigid sphere, dimensionless
C_r = $(C_D/C_D{}^s)$, dimensionless
D = equivalent spherical diameter = $(6v/\pi)^{1/3}$, cm.
g = acceleration due to gravity, cm./(sec.)^2
g_c = conversion factor, —980 dynes/g.
P = pressure, force/area, dyne/cm.^{2}
R_1, R_2 = principal radii, length units cm.
Re = Reynolds number, dimensionless
Sd = surface-tension size group, Equation (3), dimensionless
Tv = terminal-velocity group, Equation (1), dimensionless
v = volume cc.
V = velocity, cm./sec.
We = Weber number, dimensionless
Wt = gravity group, Equation (2), dimensionless
β = drop property group, Equation (8), dimensionless
μ = viscosity, poises
μ_r = μ_i/μ_o, dimensionless
ρ = density, g./cc.
σ = interfacial tension, dynes/cm.
ϕ = function of

Subscripts

i = discontinuous-phase property
o = continuous-phase property

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