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by

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NOMENCLATURE

A = cross sectional area, frontal area, $\frac{\pi D^2}{4}$, cm.^{2}

a = semi-major axis of an ellipsoid of revolution, length units.

B = dimensional constant, $\frac{M}{L\theta^2}$

b = semi-minor axis of an ellipsoid of revolution, length units.

C = Coefficient.

C_D = drag coefficient = $\frac{F}{A\left(\frac{\rho V^2}{2}\right)}$, dimensionless.

C_r = $(C_D) / (C_D^s)$, dimensionless.

c = arbitrary constant, dimensionless.

D = equivalent spherical diameter (called as diameter), cms.

D_n = D = True nominal diameter (Equivalent spherical diameter), i.e. diameter of a sphere of the same volume as the body whether solid or liquid, irrespective of the latter's shape, length units.

d = differential sign.

E = dimensional constant, $\frac{M}{L^2\theta^2}$.

F = resisting force, dynes.

G = dimensional constant, $\frac{M}{L^3\theta^2}$.

g = acceleration due to gravity (cms.) (Sec.)$^{-2}$.

g_c = conversion factor, $\frac{ML}{F\theta^2}$ = 1 in c.g.s. system.

H = arbitrary constant.

h = fineness ratio of an ellipsoid, $\frac{b}{a}$, dimensionless.

I_1, I_2 = constants of integration.

J = arbitrary constant.

j = arbitrary constant, dimensionless.

K, K_1 = arbitrary constants, dimensionless.

k = arbitrary constant, dimensionless.

L = length, cms.

l, l_1 = arbitrary constants, dimensionless.

M = mass, gms.

m = arbitrary constant, dimensionless.

n = arbitrary constant, dimensionless.

O = origin of Cartesian coordinates.

P(y) = functions of y.

p = pressure, $\frac{\text{Force}}{(\text{length})^2}$, $\frac{\text{dynes}}{\text{Cm}^2}$.

Q = constant, dimensionless

$q = \frac{dy}{dz}$.

R = radius of curvature.

R_1, R_2 = principal radii of curvature on the surface of the drop.

Re = Reynolds number = $\frac{DV\rho_o}{\mu_o}$, dimensionless.

r = equivalent spherical radius (called radius) of drop, cms.

S = surface area of a solid (or liquid drop) of any shape, cm^2.

s = surface area of a sphere of the same volume as the body (solid or liquid), cm^2.

Sd = surface tension-size group,

$$= \frac{g_c \sigma}{\mu_o}\left[\frac{3}{4}\ \frac{\rho_o^{2}}{g\mu_o\,|\rho_1 - \rho_o|}\right]^{1/3}, \text{ dimensionless.}$$

Su = surface tension group = $\frac{g_c \sigma D \rho_o}{\mu_o^{2}}$, dimensionless.

T_v = terminal velocity group,

$$= V\left[\frac{3}{4}\ \frac{\rho_o^{2}}{g\,\mu_o\,|\rho_1 - \rho_o|}\right]^{1/3}, \text{ dimensionless}$$

V = velocity, cms./sec.

v = volume, cm^3.

Wt = gravity group = $D\left[\frac{4}{3}\ \frac{g\rho_o\,|\rho_1 - \rho_o|}{\mu_o^2}\right]^{1/3}$

w = weight, gms.

We = Weber number = $\frac{DV^2 \rho_o}{\sigma}$, dimensionless.

X,Y,Z = coordinate axes.

x,y,z = coordinate values.

$y' = \frac{dy}{dz}$

$y'' = \frac{d^2y}{dz^2}$

Greek Letters

$\beta = \frac{(\rho_i - \rho_o)}{4} gD^2$, dimensionless.

$\gamma = \frac{C_D^f}{C_D^s}$, dimensionless.

θ = time, sec.

μ = viscosity, centipoises.

π = 3.1416...

ρ = density, gms./cm^3.

σ = interfacial tension, dynes/cm.

ϕ = function of ..

ψ = degree of true sphericity = $\frac{s}{S}$

Subscripts

D = Drag

n = nominal

r = ratio

1,2 = fixed values

Superscripts

f = flat plate

s = sphere

ABSTRACT

A study of the terminal velocities of fall of liquid droplets in another phase liquid under stationary conditions was made. The present studies of the subject include dimensional analysis, study of shape of the drop from theoretical considerations, experimental work for collection of fall velocity data and photographic studies of the drop behaviour. The experimental work forms the major part of the work. Six systems have been studied using organic liquids insoluble in and heavier than water. Two types of microburets have been developed. Hypodermic needles and glass tips joined to the bases of hypodermic needles were used to produce drops of uniform size. The experimental conditions and procedure have been standardized.

For any given system, as the drop size is increased, the fall velocities of the droplets increase gradually, reach a maximum and then fall off asymptotically. Two mechanisms have been postulated, one for the range where the fall velocities increase with an increase in drop size, and another for the range where the fall velocities decrease in an asymptotic manner with an increase of drop size, with the maximum velocity region corresponding to the transition from one to the other. The photographic evidence also supports this view.

The drop behaviour in the region of increasing fall velocities with an increase of drop size may be represented by:

$$\text{Weber number} = H\,(\text{Reynolds number})^{J}$$

where H and J are constants.

The drop behaviour in the region of decreasing fall velocities with an increase of drop size may be represented by

$$\left(\frac{\text{Drag coefficient of drop}}{\begin{array}{l}\text{Drag coefficient of a rigid}\\ \text{sphere of same volume and}\\ \text{density}\end{array}}\right)_{\begin{array}{l}\text{Reynolds number}\\ \text{constant}\end{array}} = K_2\beta + Q$$

where K_2 and Q are constants and β is a dimensionless group characteristic of the drop properties.

INTRODUCTION

The subject of "Rate of Fall of Liquid Droplets" belongs to the realm of two phase fluid flow. More specifically, but still in general terms, it can be called the case of motion of one phase particles or bodies inside another phase fluid. This distinguishes the present problem from the other case of flow of a fluid inside another phase material like pipes, ducts, channels, etc. The problem can be classified into five special cases (by placing restrictions on the properties of viscosities of the two phases and the interfacial tension) as shown in Table I. It is not practicable to have more than five cases, since an infinite viscosity of the continuous phase will remove it from the case of a fluid, and a negligible or zero interfacial tension will make the two phases mix together and become a single phase.

It is the aim of this thesis to investigate Case IV of motion of one liquid droplets in another liquid. The problem of Case IV can still further be subdivided into:

Case IVa. Motion of one liquid droplets in another liquid without mass transfer between the phases.

Case IVb. Motion of droplets of one liquid in another liquid with mass transfer between the phases present.

The much more specific case of IVa has been chosen for detailed experimental study. During the course of the work, it was found advisable to make use of the data, correlations and information available up to now about the other cases of I, II, III and V. Such information has to be used with due

TABLE I

CLASSIFICATION OF TWO PHASE FLOW PROBLEMS

Case	Viscosity of Continuous Phase	Viscosity of Discontinuous Phase	Interfacial Tension	General Types of Problems with Examples
I	Negligible	Infinite	Infinite	Movement of solids in gases. Ex. Dust collection, aerodynamic problems, etc.
II	Significant	Infinite	Infinite	Movement of solids in liquids. Ex. Sedimentation, submarine motion, etc.
III	Negligible	Significant	Significant	Movement of liquid droplets in gases. Ex. Mist collection, spray absorbers and dryers. Rain drops in air.
IV	Significant	Significant	Significant	Movement of one liquid droplets in another liquid. Ex. Liquid-liquid extraction in spray towers.
V	Significant	Negligible	Significant	Movement of gas bubbles in liquids. Ex. Air jet lift, air mixers, stripping.

caution. It would, in fact, seem that Cases III and V are much closer to the present work than the first two. But there is greater information available in the literature concerning the first two cases. Any general correlation to include all the five or at least Cases III to V is highly desirable. Since Cases III and IV are much closer in their properties on account of the additional property of incompressibility of the liquid drops, attempts have been made to use the information of Case III more freely than any of the others.

In Case IVa the liquid droplet might be either rising or falling. For convenience of experiment, the question of falling drops was chosen.

The drops can fall either with an accelerating velocity or with a constant terminal velocity. Only the case of drops falling with constant terminal velocities was made the subject of the present investigation.

Since a liquid-liquid system was employed, it was considered reasonable to assume that the liquids are incompressible.

It is known that the velocity of a particle falling in a fluid medium is influenced by the ratio of the diameter of the particle to the diameter of the container. It was decided that the experiments should be conducted in such a way that there would be no wall and end effects.

The continuous medium may be either stationary or moving. It was chosen that the continuous medium be stationary. Consequently, the problem might be restated as: Terminal Velocities of Fall of Liquid Droplets inside a Stationary and Infinite Medium of Another Liquid.

The above may make one feel as though the problem has been very much simplified. A further analysis of the problem will show that there are many more variables that have to be taken into consideration.

In the case of the present problem, the factors influencing the rate of fall are densities and viscosities of the two phases, interfacial tension, volume and shape of the drop and acceleration due to gravity. In other words, there are eight variables, or if the acceleration due to gravity is constant throughout, there will still be seven variables left for consideration. A detailed consideration of these will be given later.

In the above, all except the shape of the drop can be easily determined. Blanchard[6], Edgerton and Killian[11], and Henrickson[19] photographed liquid droplets in air while Ailor[3] and Farmer[12] took photographs of liquid droplets in liquids. But since the purpose of any study is to present a correlation that would obviate the trouble of repeating the work, it was considered worthwhile to try to find out the shape of the drop in terms of the other physical variables. The determination of the shape of the drop is further complicated by the fact that the shape itself is dependent on the velocity of fall, which velocity of fall it is intended to discover. The studies concerning the determination of shapes of drops will be presented later.

Since the purpose of the experimental work is finally to be able to get some correlation of the terminal velocity of droplets in terms of the known variables, it was found necessary to produce drops of known and reproducible sizes and physical properties. The preliminary work done to secure reliable data is also indicated later.

Thereafter, the drops of known sizes and properties were introduced into a continuous medium of known properties and the velocities of fall were measured. The details of this work as well as the final results obtained are presented in the main body of the thesis.

The information obtained on this topic of rate of fall of liquid droplets in liquids will be of great use in the study of two phase flow problems as well as form a good basis from which to start the study of the mechanism of liquid-liquid extraction in single drop extraction columns.

Since most of the extraction towers are worked in the turbulent range, the data was collected mostly in the range of about Reynolds number 300 to 1500.

REVIEW OF PREVIOUS WORK

It has been known for a long time that raindrops in the air attain a maximum terminal velocity of about 8 to 9 meters per second as the drop size is increased and that the velocities afterwards are independent of the drop size. Data on the fall velocities of water drops or rain drops in the air were reported by Flower,[13] Gunn and Kinzer,[17] Laws,[23] Lenard,[24] Meinzer[28] and Schmidt.[38] They have noted that the velocities of water drops in air were lower than those predicted by the assumption of the drop as a rigid sphere of equivalent spherical diameter. Liznar[25] and Imai[21] have tried to develop certain correlations to predict the fall velocities of raindrops. Hughes and Gilliland[20] and Spilhaus[41] have also developed certain correlations for the deformation as well as drag coefficient of liquid drops in gases. Liquid drops, mostly water in air, were photographed by Edgerton and Killian,[11] Lenard[24] and Magono.[27] Henrickson[19] observed the deformation of water, nitrobenzene and n-propanol drops in air.

The fall velocities of drops after different heights of fall have been experimentally studied by Flower[13] for water and methyl salicylate drops in air and by Laws[23] for water drops. Their aim was to find the initial height of fall required for the drops to reach the terminal velocities.

McDonald[26] made a detailed theoretical study of the shape and aerodynamics of large raindrops. He also used the photograph of water drops in air taken by Magono[27] and calculated the pressure distribution around a water drop of 6 mm. diameter falling with a velocity of 8.8 meters per second.

Several workers have observed that air bubbles rising in liquids are deformed and that their terminal velocities differ from those of rigid spheres of the same equivalent spherical radii. The work on air bubbles in liquids of various workers has been summarized by Allawala[4] and Peebles and Garber[33] who themselves have studied the subject in detail.

As an extensive study of liquid drops in gases or of gas bubbles in liquids is outside the scope of the present work, no attempt has been made to present a comprehensive summary of the work done on these two types of problems. The results and correlations of individual workers in these two fields will be referred to from time to time and used for purposes of comparison with the present work. Suffice it to say that drop and bubble deformations were observed and that their terminal velocities were often times found to deviate from those of equivalent spheres.

Hadamard[18] and Rybczynski[36] theoretically extended Stokes' equation for the resistance to the motion of a rigid sphere to the case when the sphere is composed of a fluid immiscible with the medium. Bond[7], Bond and Newton[9] experimentally studied the behaviour of bubbles and drops in liquids and compared it with Stokes' law. They also derived a modified form of Stokes' law for bubbles and drops. Since their work is concerned with the Stokes' law region, it is not of much help in the present work which is primarily in the turbulent zone.

Conway[10] observed that liquid droplets did oscillate in their shape and were periodically deflected from the vertical path while falling through another liquid. He also compared the fall velocities of the drops with those of rigid spheres and found that the liquid droplets fell with slower velocities. He felt that (1) mass transfer, (2) deviation from spherical shape and (3) the deviation from the vertical motion of the drops might be responsible for this difference. He also observed that smaller drops fell with greater velocities than larger drops. This anomolous result was also observed by Lenard[24] for water drops in air and was later observed by Katz[22] for liquid droplets in liquids without mass transfer between the phases. It will, however, be taken up in detail in the main part of the work. Katz[22] collected data on the fall velocities of different liquid droplets in water under varying conditions using 2-inch and 3-inch diameter glass columns. He observed the wall effect in the 2 inch column and was not sure about the same in the 3-inch column. As such he could not develop any quantitative relationships based on his work.

Ailor[3] used a 1-inch glass column and observed the velocities of fall of liquid droplets in liquids in all the three regions of viscous, transition and turbulent flow. He found that the drag coefficient for drops in the viscous region was twice that predicted by Stokes' law for rigid spheres. He also observed that the drag coefficients of drops followed a curve similar to that of rigid spheres when the drag coefficients of drops were plotted versus Reynolds number in the transition range. In the turbulent region he found that the drag coefficients of the drops were dependent to a great extent on the drop properties. He did not, however, develop any quantitative relationships in the transition and turbulent regions. It is also possible that his data could be in error due to wall effect in his 1-inch column.

Farmer[12] found that the hydrodynamics of falling drops inside another liquid agreed very well with the laws of hydrodynamics for rigid spheres in the transition and viscous flow regions. He found that the laws assuming the drops to be rigid spheres no longer applied in the turbulent region. He made some photographic studies on the shape of the drops and found that the drops tended to deform with increasing diameter and had the shape of an ellipsoid of revolution. He concluded that, as a result of drop deformation, the velocity of the drop reached a maximum around a Reynolds number of 600 and no further increase in velocity occurred with increase in drop size. He used a 2-inch column.

Smirnov and Ruban[39] measured the relative velocities of liquid droplets in liquids and also measured the velocities of air bubbles in water. They varied the drop sizes and also the velocities of the continuous medium. They used dimensional analysis and developed certain correlations. They did not consider the interfacial tension at the surface of the drop.

At this stage it seems appropriate to summarize the treatment of the subject by the different workers in the field. All the workers have accepted the importance of drop size, density of drop material as well as the density and viscosity of the continuous medium in determining the drag coefficient of drops. It may be noted that all these factors are also of importance in the case of rigid spheres falling in a fluid. The next question is to find out what additional drop properties should be considered. The two drop properties to be considered are viscosity of drop material and interfacial tension. Hughes and Gilliland[20], Emai[21] and Spilhaus[41] have considered interfacial tension to be important and ignored the viscosity of the liquid drop. Bond[7], Bond and Newton[8], Saito[37], Smirnov and Ruban[39] took the drop viscosity into consideration and either relegated interfacial tension to lesser importance or completely ignored it. The correlations developed by Flower[13] and Liznar[25] are based on experimental data for fall of liquid droplets in air and both their correlations contain empirical constants calculated for each of the systems from the available data.

Katz[22] undertook to study the influence of both drop viscosity and interfacial tension, but he could not arrive at any quantitative correlations. Ailor[3], who also had a similar idea, did assume many of the properties of his materials used to be the same as literature values. He did not make use of the drop viscosity and interfacial tension values to any considerable extent.

It seems, therefore, appropriate to show conclusively whether both or only one of the two drop properties of viscosity and interfacial tension are of importance in influencing the fall velocity of the drop, and if so, to what extent. Hughes and Gilliland[20] and Spilhaus[41] have proposed certain general quantitative correlations which take into account interfacial tension, but have tested them only against the experimental data of Laws[23] on water drops in air. It is to be seen whether their correlations are applicable to the fall of liquid droplets in liquids. The correlations of Smirnov and Ruban[39] for relative motion of liquid droplets in liquids are not strictly applicable to the present work as they used a continuous medium in motion. A detailed study of the subject with a view to find out quantitative relationships seems to be a necessity. Even a good set of reliable and reproducible data over a wide range of conditions should be of great help, as there is not even available in the literature such data.

THEORY

(a) Basic Principles and Previous Work

The resisting force to the fall of a body immersed in a fluid was first considered by Newton[29] according to whom we can write

$$F = C_D A \left(\frac{\rho_o V^2}{2}\right) \qquad (1)$$

where F = resisting force to the motion of the body.

C_D = a numerical constant which Newton believed to vary with the geometrical form of the body and its orientation with respect to the direction of motion. It is also called "drag coefficient."

A = cross-sectional area of the body, projected on a plane perpendicular to the direction of motion.

ρ_o = the density of the resisting medium.

V = velocity of the body.

Newton's basic equation is dimensionally correct, but his reasoning as to the magnitude of C_D is physically unsound. This factor depends not only on the shape and orientation of the body, but varies considerably with the viscous characteristics of the motion. Thus, for solid bodies the factor C_D is found to vary with Reynolds number. It is also known that the influence of viscosity decreases with an increase of the Reynolds number.

The extent to which the variation in the geometrical form of the body will influence the resistance depends largely upon the magnitude of Reynolds number. At low Reynolds numbers shape is of secondary importance; but as Reynolds number increases, the body tends to leave in its course a turbulent wake, the size and intensity of which is governed largely by the form of the longitudinal profile. A stream-lined body leaves a wake of minimum size, whereas a very angular profile will produce eddies of greatest magnitude.

Stokes[44] developed an expression for the resistance of rigid spheres at very small Reynolds numbers up to 0.5, on the assumption that in this range the mass acceleration of the fluid caused by the motion of the sphere is negligible in comparison with the influence of viscosity. Stokes' equation may be written as:

$$F = \frac{24}{Re} A \frac{\rho_o V^2}{2} \qquad (2)$$

in which $C_D = \frac{24}{Re}$ and

Re = Reynolds number

Stokes' analysis was extended analytically to higher values of Re by Oseen[30] who obtained the approximate relationship,

$$C_D = \frac{24}{Re} \left(1 + \frac{3}{16} Re\right) \qquad (3)$$

Goldstein[15] gave an exact solution of Oseen's equation with the result

$$C_D = \frac{24}{Re} \left(1 + \frac{3}{16} Re - \frac{9}{1280} Re^2 + \frac{71}{20480} Re^3 - \dots\right) \qquad (4)$$

As Re becomes small, the additional terms drop out and both Oseen's and Goldstein's equations approach that of Stokes. Stokes' equation yields good results up to Re = 0.5, while Oseen's expression is good somewhat beyond that of Stokes, and Goldstein's is good up to about Re = 2.

Beyond this there are no analytical methods giving dependable results. Hence the correlations are purely empirical. But enough data has been collected for rigid non-spinning spheres and C_D is found to be a single-valued function of Re. Curves correlating C_D with Re are available in the literature and are given by Brown[9], Gaudin[14], Perry[31] and Rouse.[35] Spronck[42] and Rouse[35] have also given nomograms while Zahm[46] has given a formula correlating C_D with Reynolds number for rigid spheres.

In the case of a particle falling at the terminal velocity, the resistance opposing its fall is equal to the weight of the body less the bouyancy effect. Hence.

$$F = (\rho_1 - \rho_o)\, g\, v \qquad (5)$$

where

ρ_1 = density of the particle

v = volume of the particle and

g = acceleration due to gravity.

If the particle were a sphere, we have

$$v = \frac{\pi D^3}{6} \text{ and } A = \frac{\pi D^2}{4}, \text{ so that}$$

$$C_D^s = \frac{4\,(\rho_1 - \rho_o)gD}{3\,\rho_o V^2} \qquad (6)$$

where

C_D^s = drag coefficient of the sphere

D = diameter of the sphere, and

V represents the terminal velocity of the sphere.

Treatment of variation of C_D with shape is by no means simple, for a complete definition of shape requires more than one parameter. The additional problem of orientation with respect to the direction of motion involves further difficulties. Squires and Squires[43], however, feel that though the initial orientation is an important factor in the viscous range, it is not so in the turbulent range for discs, with which they have experimented. Goldstein[15] shows clearly that the drag coefficient of a rotating sphere decreases first to a minimum value and then increases as the ratio of equatorial speed to wind speed is increased. Hence the rotation or spinning of a body is an important factor influencing the drag coefficient. Its importance on shapes other than spheres is not known. Prandtl[34] referring to the experiments on discs at Gottingen states that data on discs was collected on either side of the range of Re = 80 to 3000. He states "the region between Re = 3000 and Re = 80 has to be left out, since the discs oscillate vigorously to and fro and hence give values of resistance up to 50 per cent greater than if they fell steadily." This behaviour of oscillations is important, for us, since the present work is concerned with this particular range of Reynolds numbers.

For shapes other than spherical it is very difficult to estimate D, especially in view of the fact that many bodies do change their orientation erratically during fall. So the true nominal diameter D_n has been suggested by Wadell.[45] The true nominal diameter, also referred to as equivalent or mean spherical diameter, has been defined as the diameter of a sphere of the same volume as the solid irrespective of the latter's shape. In the present work the symbol D will be used to designate the nominal diameter. Since the nominal diameter of a sphere is its actual diameter, there can be no confusion caused by the use of the symbol D. The nominal diameter idea is extended to the case of liquid droplets in the present work.

Wadell[45] also introduced the idea of the degree of true sphericity, $\psi = \frac{s}{S}$, where s = the surface area of a sphere and S = the surface area of a solid of any shape having the same volume as the sphere. Wadell has drawn curves for C_D versus Re with ψ as parameter. This chart is reproduced by Brown[9] . It would, however, seem that further experimentation is necessary before these curves could be accepted with much confidence. The data on spheres may be considered much more reliable having been obtained by many workers under widely varying conditions. It appears that, if the degree of sphericity can somehow be estimated, it may be possible to estimate the drag coefficient using the charts of Wadell, if there are no other complicating factors like spinning, vibrations and orientation.

For deformable spheres, Saito[37] has worked out theoretically the "shape of the nearly spherical drop which falls through viscous fluid," and predicted whether a drop will be an oblate or prolate spheroid depending upon the properties of the system. His method is applicable only at very low velocities and does not give any method of estimating the actual amount of deformation produced in a liquid drop.

Imai[21] studied the velocity of falling raindrops. He also assumed the deformation of the raindrops to be small. In order to arrive at a solution he assumed the potential flow of air around a spherical drop and arrived at a method of predicting the velocities of water drops in air. However, he neglected the hydrostatic head of water inside the drop. He also assumed that the drag coefficient of the deformed drop is the same as for a sphere and he allowed for the increased cross-sectional area of the deformed drop. By this method he could estimate the fall velocities of rain drops up to D = 0.2 cm., where his calculated values compared very well with the experimental data of Gunn and Kinzer.

Imai's final results and procedure may be summarized as follows:

$$r = \left(\frac{3 \mu_o^2 \; C_D \; Re^2}{32 \rho_o \; g} \right)^{1/3} \qquad (7)$$

$$V = \frac{\mu_o \; Re}{2 \; \rho_o \; r} \left(1 - \frac{3}{128} \; \frac{\mu_o Re^2}{r \sigma \rho_o} \right) \qquad (8)$$

where μ_0 = Viscosity of air

r = radius of an undeformed drop

ρ_o = density of air, and

σ = surface tension of water in air.

Imai assumed values of Re and read off the C_D values for rigid spheres. Then he calculated r and V. From these he drew the curve of V versus r.

Spilhaus[41] considered rain drop size, shape and falling speed. He considered large size drops and assumed that the drop deforms to an oblate spheroid. He considered that the extreme case of such a deformation is a flat plate. If

$$\gamma = \frac{C_D^f}{C_D^s}$$

where C_D^f = drag coefficient of a flat plate

then he assumes that the drag coefficient of the drop is

$$C_D = C_D^s \left[\gamma - h\,(\gamma - 1)\right]$$

where

h = fineness ratio of the spheroid = $\frac{a}{b}$

a = semi-major axis of the spheroid

b = semi-minor axis of the spheroid.

He neglected the hydrostatic head of the water inside the drop. His assumption that the excess of internal pressure of the drop over the external pressure is equal to $\frac{2\sigma}{a}$ is questionable. He obtained

$$\frac{V^2}{r} = \frac{4\,\rho_i\,g}{3\,\rho_o C_D^s}\left[\gamma - h\,(\gamma - 1)\right]^{-1} h^{2/3} \qquad (9)$$

Spilhaus considered the drag coefficients of spheres and flat plates to be constant in the range of Re = 750 to 2,000. For smaller drops of radii less than 1.3 mm., he considered the deformation to be negligible. He checked his prediction of fall velocities with the data of Laws[23] for water drops in air.

Hughes and Gilliland[20] studied the mechanics of drops and obtained the following dimensionless groups.

$$\text{Reynolds number} = \text{Re} = \frac{DV\rho_o}{\mu_o} \qquad (10)$$

$$\text{Surface tension group} = \text{Su} = \frac{g_c\,\sigma D\,\rho_o}{\mu_o} \qquad (11)$$

$$\text{Gravity group} = \text{Wt} = D\left[\frac{4}{3}\;\frac{g\rho_o\left|\rho_i - \rho_o\right|}{\mu_o^2}\right]^{1/3} \qquad (12)$$

$$\text{Acceleration group} = \text{Ac} = \frac{D}{V^2}\,\frac{dV}{d\theta} \qquad (13)$$

Since the present work is concerned primarily with the terminal velocities, the acceleration group may be omitted for the present purposes. The surface tension group and gravity group may be rewritten in a different form thus:

$$\text{Su} = \text{Re}\left(\frac{\sigma}{\mu_o V}\right) \qquad (14)$$

$$\text{Wt} = (C_D\,\text{Re}^2)^{1/3} \qquad (15)$$

So $\left(\frac{\sigma}{\mu_o V}\right)$ is a new group which they have introduced to account for the drop property of interfacial tension. They have also presented a plot of C_D vs. Re with Su group as a parameter. It is very difficult to justify their surface tension group

$$Su = \frac{g_c \sigma D \rho_o}{\mu_o^2}$$

since it can easily be seen that σ and D act in opposite directions during the deformation of a drop. While an increase in interfacial tension decreases the deformation of the drop and makes it behave more like a rigid sphere, an increase in the diameter of the drop tends to increase the deformation of the drop. Since σ and D act in opposite directions, a group with both of them multiplying each other does not seem to be reasonable. So dimensionless groups that overcome this difficulty have to be developed and used.

Hughes and Gilliland[20] using a quasi-thermodynamic treatment, arrived at the same results as Spilhaus.[41] They also tried to estimate the drop deformation and then interpolate the drag coefficient of the drop from the values for spheres and flat plates. They also assumed the drop as a spheroid and that the increase in the surface energy of the drop is equal to the frictional work done upon the drop during its fall. Their theoretical correlations also make use of the data of Laws[23] for water drops in air. They used the following three dimensionless groups:

$$\text{Gravity group} = Wt = \left[C_D \; Re^2\right]^{1/3} = D\left[\frac{4}{3} \; \frac{g\,\rho_o\,|\rho_i - \rho_o|}{\mu_o^2}\right]^{1/3} \quad (16)$$

$$\text{Terminal velocity group} = \frac{Re}{Wt} = \left(\frac{C_D}{Re}\right)^{-1/3} = V\left[\frac{3}{4} \; \frac{\rho_o^2}{g\,\mu_o\,|\rho_i - \rho_o|}\right]^{1/3} \quad (17)$$

$$\text{Surface tension size group} = \; Sd = \frac{Su}{Wt} = T_v\left(\frac{\sigma}{\mu_o V}\right)$$

$$= \frac{g_c\,\sigma}{\mu_o}\left[\frac{3}{4} \; \frac{\rho_o^2}{g\,\mu_o\,|\rho_i - \rho_o|}\right]^{1/3} \quad (18)$$

They have plotted T_v vs. Wt with Sd group as parameter and tested it against the data of Laws[23] and Watson.[49]

With this background of the theoretical developments, it seems appropriate to proceed next to the theoretical work which includes dimensional analysis and a study of the shape of the drop.

(b) Dimensional Analysis

For the planning of any experimental work as well as for the analysis of the data so obtained, the method of dimensional analysis is helpful. So, it has been taken up first.

The variables influencing the rate of fall of a liquid droplet at terminal velocity in another liquid may be summarized as follows:

TABLE II

VARIABLES INFLUENCING THE RATE OF FALL OF A LIQUID DROPLET

Variable	Symbol	Dimensions
Drag coefficient	C_D	none
Interfacial tension	σ	$\frac{M}{\theta^2}$
Acceleration due to gravity	g	$\frac{L}{\theta^2}$
Drop volume	v	L^3
Velocity of fall	V	$\frac{L}{\theta}$
Density of continuous phase	ρ_o	$\frac{M}{L^3}$
Density of discontinuous phase	ρ_i	$\frac{M}{L^3}$
Viscosity of continuous phase	μ_o	$\frac{M}{L\theta}$
Viscosity of discontinuous phase	μ_i	$\frac{M}{L\theta}$

In the above list of variables it may be observed that there is no factor included for the shape of the drop. Since the shape of the drop will be a dimensionless quantity and since it is not possible to determine it directly, it could not be included in the list. It is felt that the other variables giving the physical properties of the drop might take care of the shape factor, since deformation of the drop is very likely to depend on the physical properties of the drop and the flow pattern.

Further, since the diameter of a particle is generally used in dimensionless groups, it is felt convenient to use the equivalent spherical diameter D (or called diameter for the sake of simplicity) of the drop instead of volume V, in the dimensional analysis.

Since the deformation of a drop is itself a variable and the actual frontal area is changed by the deformation, it is not practicable to estimate the true drag coefficient of the drop. So a hypothetical drag coefficient has been defined using the equivalent spherical diameter of the drop. This hypothetical drag coefficient of the drop is simply referred to as the drag coefficient of the drop.

$$C_D = \frac{F}{\frac{\pi D^2}{4} \frac{\rho_o V^2}{2}} = \frac{8F}{\pi D^2 \rho_o V^2} \qquad (19)$$

In the case of a drop falling at its terminal velocity

$$C_D = \frac{4g\,(\rho_i - \rho_o)D}{3\,\rho_o V^2} \qquad (6)$$

For dimensional analysis it can be written that

$$C_D = \phi(\sigma, g, D, V, \rho_i, \rho_o, \mu_i, \mu_o) \qquad (20)$$

where

ϕ = function of ...

There are eight variables and only three dimensions. Hence, in general, we may expect five dimensionless groups including C_D. It is advisable to see if some simplification can be done before solving the above equation. Also variables like ρ_i and ρ_o as well as μ_i and μ_o have the same dimensions and dimensional analysis will not be of much help with such repeated quantities. Also, if ρ_i is to be introduced to indicate the drop property, it appears that the expression $|\rho_i - \rho_o|$ will be much more appropriate, since it is only the difference in densities that actually causes the motion of the drop. However, since dimensional analysis cannot take care of repeated quantities, only one variable ρ will be used without any subscript. Whether to use $|\rho_i - \rho_o|$ or ρ_o should be decided by inspection of the group.

Similarly μ_i and μ_o are repeated quantities. One can by inspection write $\left(\frac{\mu_i}{\mu_o}\right)$ as one dimensionless group. Since viscosities may enter other groups also, the variable μ is used without subscript. Here also the choice of μ_i or μ_o will have to be a matter of judgment.

Then

$$C_D = K\, \sigma^c\, g^k\, D^f\, V^l\, \rho^m\, \mu^n \left(\frac{\mu_i}{\mu_o}\right)^j \qquad (21)$$

where

K,c,k,f,l,m,n,j are all arbitrary constants.

Substituting the dimensions, one obtains:

$$I = \left(\frac{M}{\theta^2}\right)^c \left(\frac{L}{\theta^2}\right)^k L^f \left(\frac{L}{\theta}\right)^l \left(\frac{M}{L^3}\right)^m \left(\frac{M}{L\theta}\right)^n \qquad (22)$$

$$M: \quad 0 = c + m + n \qquad (23)$$

$$\theta: \quad 0 = -2c-2k-l-n \qquad (24)$$

$$L: \quad 0 = k + f + l - 3m - n \qquad (25)$$

Since there are six unknowns and only three equations, it is not possible to solve the above completely. But they can be solved to the extent of reducing the total number of unknowns from six to three.

Solving the above equations, one can obtain

$$f = c + 3k + l \qquad (26)$$

$$m = c + 2k + l \qquad (27)$$

$$n = -2c-2k-l \qquad (28)$$

Substituting the above values in equation (21), we get

$$C_D = K\, \sigma^c\, g^k\, D^{c+3k+l}\, V^l\, \rho^{c+2k+l}\, \mu^{-2c-2k-l} \left(\frac{\mu_i}{\mu_o}\right)^j$$

$$= K \left(\frac{\sigma D \rho}{\mu^2}\right)^c \left(\frac{g D^3 \rho^2}{\mu^2}\right)^k \left(\frac{D V \rho}{\mu}\right)^l \left(\frac{\mu_i}{\mu_o}\right)^j \qquad (29)$$

In the above we can easily recognize the group $\frac{DV\rho}{\mu}$ as Reynolds number. So the ρ may be assigned the value ρ_o and μ called μ_o to suit the general definition of Reynolds number. Also for convenience let us designate $\frac{\mu_i}{\mu_o} = \mu_r$ and $Re = \frac{DV\rho_o}{\mu_o}$.

Then

$$C_D = K \left(\frac{\sigma D \rho}{\mu^2}\right)^c \left(\frac{g D^3 \rho^2}{\mu^2}\right)^k Re^l\, \mu_r^j \qquad (30)$$

In the above, if c = -k, the two factors may be combined:

$$\frac{gD^3 \rho^2}{\mu^2} \times \frac{\mu^2}{gD^3 \rho^2} = \frac{gD^2 \rho}{\sigma}$$

Since D^2 suggests the area of the drop and $g\rho$ suggests the weight, it appears as though the ρ term may be considered as composed of $|\rho_i - \rho_o|$. Then the above can be written as:

$$\frac{gD^2 \, |\rho_i - \rho_o|}{\sigma}$$

For convenience at a later stage of work, this term has been used as:

$$\beta = \frac{gD^2 \, (\rho_i - \rho_o)}{4\,\sigma} \qquad (31)$$

The factor 4 in the denominator suggests that the equivalent spherical radius is used instead of the equivalent diameter. The use of $(\rho_i - \rho_o)$ instead of $|\rho_i - \rho_o|$ can be justified, since the present studies are concerned with falling drops and $\rho_i > \rho_o$.

Then one possible way of writing equation (30) will be:

$$C_D = K \beta^k \, Re^l \, \mu_r^j \times 4 \qquad (32)$$

In the above, the β group has the advantage that it is free from the velocity term and contains only the diameter of the drop as a variable for any given system.

Since c and k are arbitrary constants, equation (30) may be rearranged and written in a still different way.

First, the group $\frac{\sigma D\rho}{\mu^2}$ may be expanded:

$$\frac{\sigma D\rho}{\mu^2} = \left(\frac{DV^2\rho}{\sigma}\right)\left(\frac{\sigma^2}{\mu^2V^2}\right)$$

Then again, if we put c = -k

$$\left(\frac{\sigma D\rho}{\mu^2}\right)^c \left(\frac{gD^3\rho^2}{\mu^2}\right)^k = \left(\frac{DV^2\rho}{\sigma}\right)^c\left(\frac{\sigma^2}{\mu^2 V^2}\right)^c \left(\frac{\mu^2}{gD^3\rho^2}\right)^c$$

$$= \left(\frac{DV^2\rho}{\sigma}\right)^c \left(\frac{\sigma^2}{gD^3\rho^2V^2}\right)^c$$

In the above $\frac{DV^2\rho}{\sigma}$ can be recognized as Weber number. Then we can assign specific values of ρ_o and μ_o to the ρ and μ terms and write

$$We = \frac{DV^2\rho_o}{2\sigma} \qquad (33)$$

Then equation (30) becomes:

$$C_D = K(We)^c \left(\frac{2\sigma^2}{gD^3\rho^2V^2}\right)^c Re^1 \quad \mu_r^j$$

In the group

$$\frac{2\sigma^2}{gD^3\rho^2V^2}$$

the term D^3 suggests the volume of the drop and ρg suggests the weight term. So, at least one of the two ρ's can be replaced by $(\rho_i - \rho_o)$. Then it may be written as :

$$\frac{2\sigma^2}{gD^3(\rho_i-\rho_o)V^2}$$

The term $\frac{\rho V^2}{2}$ suggests the combination $\frac{\rho_o V^2}{2}$ in the kinetic energy term and the ρ may be replaced by ρ_o.

If c = -1

$$\frac{C_D}{\frac{gD^3(\rho_i - \rho_o)}{2}\ \frac{\rho_o V^2}{2}} = \frac{4g(\rho_i - \rho_o)D\ 2\sigma^2}{3\rho_o V^2 gD^3\ (\rho_i - \rho_o)\ \rho_o V^2}$$

$$= \frac{2}{3}\left(\frac{2\sigma}{DV^2\rho_o}\right)^2 = \frac{2}{3}\ (We)^{-2}$$

Now the equation (30) can be written as:

$$\frac{2}{3}(We)^{-2} = K\ We^{-1}\ Re^{1}\ \mu_r^{j}$$

or
$$We = \frac{2}{3K}\ Re^{-1}\ \mu_r^{-j} \qquad (34)$$

At this stage it seems appropriate to point out that equations (32) and (34) are special cases of the general equation (30), based upon the restrictions that $c = -k$ in one case and $c = -k = 1$ in the other case. It is entirely possible to get a different set of groups either by imposing different conditions on c and k, or by solving the three equations (23), (24), (25) for a different set of three constants instead of c, k, and l chosen in the present solution. Thus for example, if k, m, and n are chosen, one will obtain:

$$C_D = K\left(\frac{DV^2\rho}{\sigma}\right)^m \left(\frac{V\mu}{\sigma}\right)^n \left(\frac{gD}{V^2}\right)^k \left(\frac{\mu_i}{\mu_o}\right)^j$$

and one can write $\frac{DV^2\rho_o}{2\sigma} = We$. The above can be rearranged and written as:

$$C_D = K\left(\frac{DV^2\rho_o}{2\sigma}\right)^m \left(\frac{DV\rho_o}{\mu_o}\right)^n \left(\frac{\mu^2}{D\rho\sigma}\right)^n \left(\frac{gD}{V^2}\right)^k \left(\frac{\mu_i}{\mu_o}\right)^j$$

$$= K\ (We)^m\ (Re)^n \left(\frac{\mu^2}{D\rho\sigma}\right)^n \left(\frac{gD}{V^2}\right)^k \mu_r^j \qquad (35)$$

Hence the real value of the dimensionless groups can only be assessed by testing on actual data. As such, the dimensional analysis will have to be left at this stage and taken up again in the discussion of the experimental results.

(c) Shape of the drop

In this section an attempt has been made to determine the shape of a liquid drop falling through another liquid. The difficulties as well as limitations in a purely theoretical method of approach to this problem are also indicated. It is a well known equation of physics that, for a liquid drop or bubble

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{pi - po}{\sigma} \qquad (36)$$

where

R_1 and R_2 are the two principal radii at any point on the surface of a drop, and

pi and p_o are the internal and external pressures at the surface of the drop.

By virtue of the surface tension property, the drop or bubble tends to assume the smallest surface area so that the surface energy is at a minimum. Because of this property of the drop or bubble to shrink to a minimum surface, there is an excess of internal pressure over the external pressure as indicated by equation (36). A derivation of the above formula is given by Adams[1] and it can be found in most textbooks of physics and physical chemistry.

Bashforth and Adams[5] have calculated the shape of sessile drops, and presented their data in the form of elaborate tables. Adams[2] gives a brief summary of the principles underlying the calculation of Bashforth and Adams.

In their work the drop is attached to a surface and is stationary.

The present problem deals with a drop in motion. In order that it may be possible to solve equation (36) and use it for the determination of the drop shape, it is necessary to make certain simplifying assumptions. They are:

1. The drop has axial symmetry about the vertical axis.
2. The drop can attain an equilibrium shape.
3. The drop has no oscillations or rotations.
4. The drop falls vertically in a straight line.

In addition to the above, we can also consider that

1. The volume of the drop is known,
2. The physical properties of the system are known, and,
3. The drop is falling with the terminal velocity, i.e. there is no acceleration.

Let the equivalent spherical radius of drop = r

Volume of the drop = $\frac{4}{3}\pi r^3 = v$

For the sake of convenience let us choose the Cartesian coordinates X, Y, Z with the origin O at the center of the drop. Also let the drop be considered stationary and the external liquid moving with the terminal velocity V.

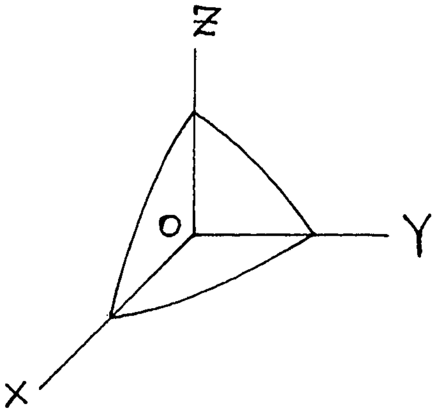


Fig. 1

Coordinate System for the Droplet.

Since there is axial symmetry, we can as well solve the problem as a two dimensional one, using Y and Z coordinates. Any curve obtained can be rotated about the Z axis to get the three dimensional shape of the drop.

In equation (36)

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{p_i - p_o}{\sigma}$$

the two principal radii at any point on the surface of revolution can be expressed by:

$$R_1 = - \frac{\frac{d^2y}{dz^2}}{\left[1 + \left(\frac{dy}{dz}\right)^2\right]^{3/2}} \text{, and}$$

$$R_2 = \frac{1}{y\left[1 + \left(\frac{dy}{dz}\right)^2\right]^{1/2}}$$

where y and z are the coordinates of the point along the Y and Z axes, and d indicates the differential sign. For the sake of simplicity let us write

$$y' = \frac{dy}{dz} \text{ and } y'' = \frac{d^2y}{dz^2} .$$

Then equation (36) can be written as:

$$- \frac{y''}{\left[1 + (y')^2\right]^{3/2}} + \frac{1}{y\left[1 + (y')^2\right]^{1/2}} = \frac{p_i - p_o}{\sigma} \qquad (37)$$

This is the fundamental differential equation to be solved. In the above the interfacial tension value, σ can be considered a constant for the range of drop sizes dealt with in the present work. As such it is possible to absorb σ in any arbitrary constant, if necessary. It is next necessary for us to express $(p_i - p_o)$ as a function of y so that the differential equation can be solved. We can use the following boundary conditions for any particular drop:

$$\text{At } z = 0,\ y' = 0 \qquad (38)$$

$$y = 0,\ y' = \infty \qquad (39)$$

$$\text{Total volume of the drop} = 2\int_0^{y_1} \pi y^2 dz$$

$$= v_1 = \text{a known constant} \qquad (40)$$

The subscript 1 is used to indicate definite values.

$(p_i - p_o)$ = constant, only when the drop is stationary and the hydrostatic head can be neglected within the drop from its top to bottom. For a general case let $\frac{(p_i - p_o)}{\sigma} = P(y)$, where P(y) indicates the excess pressure as function of y. Let it also be possible to expand P(y) in terms of y. One could immediately be faced with the question that P(y) is not the same both above and below the equator of the drop. So we have to deal with it as two separate cases of $z \geqq 0$ and $z \leqq 0$.

For the present, let us concentrate our attention on the solution of the differential equation. Later on the solution can be modified and dealt with as two special problems. Let

$$P(y) = B + Ey + Gy^2 + \dots \qquad (41)$$

where B,E,G are constants. It should, however, be noted clearly that B,E,G, etc. are dimensional constants. Thus B has the dimensions of pressure, $\frac{M}{L\theta^2}$, E has the dimensions $\frac{M}{L^2\theta^2}$, and G has the dimensions $\frac{M}{L^3\theta^2}$. For the sake of simplicity, let us first use only the first power of y and neglect higher powers of y, and solve the differential equation. If necessary the higher powers of y can always be introduced and the differential equation solved by similar methods. The differential equation (37) can be written as

$$-\frac{y''}{\left[1+(y')^2\right]^{3/2}}+\frac{1}{y\left[1+(y')^2\right]^{1/2}}=B+Ey \qquad (42)$$

Put $y' = q$

Then $y'' = q\,\frac{dq}{dy}$

$$-\frac{q\,\frac{dq}{dy}}{(1+q^2)^{3/2}}+\frac{1}{y\,(1+q^2)^{1/2}}=B+Ey$$

Multiplying throughout by ydy, we get

$$-\frac{yq\,dq}{(1+q^2)^{3/2}}+\frac{dy}{(1+q^2)^{1/2}}=Bydy+Ey^2dy$$

Integrating both sides, we get

$$\frac{y}{(1+q^2)^{1/2}}=\frac{By^2}{2}+\frac{Ey^3}{3}=I_1 \qquad (43)$$

where I_1 = constant of integration

At z = 0, y = y, and q = 0

$$y_1=\frac{By_1^2}{2}+\frac{Ey_1^3}{3}+I_1$$

or
$$I_1=y_1-\frac{By_1^2}{2}-\frac{Ey_1^3}{3} \qquad (44)$$

At $y = 0$, $z = z_\ell$ and $q = \infty$

$$0 = I_1$$

Hence

$$y_1 = \frac{By_1^2}{2} + \frac{Ey_1^3}{3}$$

or
$$\frac{By_1}{2} + \frac{Ey_1^2}{3} = 1 \tag{45}$$

Since it is a quadratic, we can solve for y_1

$$y_1 = \frac{3}{4E}\left(-B \pm \sqrt{B^2 + \frac{16E}{3}}\right) \tag{46}$$

If B and E are known, y_1 can be calculated. Assuming that y_1 can be obtained, we shall proceed to the next stage of the solution.

$$\frac{y}{(1+q^2)^{1/2}} = \frac{By^2}{2} + \frac{Ey^3}{3}$$

Squaring both sides and rearranging

$$\frac{y^2}{\left(\frac{By^2}{2} + \frac{Ey^3}{3}\right)^2} - 1 = q^2 = \left(\frac{dy}{dz}\right)^2$$

$$z = \int dz = \int \frac{\frac{By}{2} + \frac{Ey^2}{3}}{\sqrt{1-\left(\frac{By}{2} + \frac{Ey^2}{3}\right)^2}}\, dy + I_2 \tag{47}$$

where I_2 = integration constant. The sign before the right hand side integrand will be - or + whether the integration is in 1st or 4th quadrant. In the above, B and E are constants. As far as the author is aware, there is no simple integration formula for the above. One way of integrating equation (47) is as follows:

$$\Delta z = \frac{\frac{By}{2} + \frac{Ey^2}{3}}{\sqrt{1 - \left(\frac{By}{2} + \frac{Ey^2}{3}\right)^2}} \quad \Delta y \qquad (48)$$

The best place to start is $y = 0$. Then at $y = 0$, $z = z_1$. So choose a certain value of $z = z_1$. Then, if it is in the 1st quadrant,

$$\Delta z = - \frac{\frac{By}{2} + \frac{Ey^2}{3}}{\sqrt{1-\left(\frac{By}{2} + \frac{Ey^2}{3}\right)^2}} \quad \Delta y \qquad (49)$$

Next choose small values of Δy. For the first step, after $y = 0$, we can use

$$\Delta z \approx - \frac{\frac{B(\Delta y)^2}{4}}{\sqrt{1 - \left(\frac{B\Delta y}{4}\right)^2}} \approx - \frac{B(\Delta y)^2}{4}$$

Using these values of Δz and Δy, the next step can be calculated and thus a step by step numerical integration carried out until $y = y_1$, or $\left(\frac{By}{2} + \frac{Ey^2}{3}\right) = 1$ when Δz will approach ∞. This is a difficulty in this method of integration. Some of the calculation work may be simplified if the denominator term in equation (49) is expanded by way of binomial expansion and omitting the last terms of the expansion.

At this stage one is forced to use specific values for B and E, if the work is to be carried further. In order to get the values B and E one must get the nature of $P(y)$ in which p_o is a function indicating pressure distribution outside the drop. In order to get the outside pressure distribution, one should have an idea of the shape of the body. It was felt that the shape of the body may be assumed as a first approximation, either as an ellipsoid or even as a sphere. The available data on pressure distributions around solid bodies is all at very high Reynolds numbers and hence could not be used for the present purposes.

At this stage it looks advisable to consider the difficulties in the present method of solution.

1. There is no data on pressure distribution at the Reynolds numbers of interest to the present work.

2. Even if pressure distribution data at the Reynolds numbers and about the shapes of interest for the present work is available, the numerical step by step integration is too laborious to be of any practical use.

3. Even if the shape of the drop is obtained, there is no certainty that the drag coefficiens given for different shaped solid bodies (which data itself is not comprehensive) can be used for liquid droplets.

It is also proper at this stage to discuss the validity or otherwise of the first four assumptions made at the beginning of this section.

Assumption 1: The drop has axial symmetry about the vertical axis. This assumption is not strictly valid. It is nearly true only in smaller drop sizes. Some of the photographs of drops taken in the present work have clearly shown that the larger drops do not have axial symmetry. The photographic evidence is presented later.

Assumptions 2 and 3: The drop can attain an equilibrium shape and the drop has no oscillations or rotations. The photographic evidence shows that only small drops have a reasonably stable equilibrium shape. The bigger drops have been found to change their shape very violently. In that process the drops oscillate in their shape. The oscillations in shape were also observed by Blanchard[6] for water drops in air. He also mentions the rotation of the water drops. The exact question of rotation could not be decided in the present work though nutation of drops could be observed clearly. There is a strong suspicion that the smaller drops were actually rotating, though it could not be shown clearly in the photographs. The drops do not only vibrate or oscillate in their shape, but also oscillate as a whole and follow a zig-zag path during fall.

Assumption 4: The drop falls vertically in a straight line. As has already been mentioned above, the drops fall in a zig-zag path. Actually very small drops seem to fall in a reasonably vertical path, and as drop size is increased, their drifting increases to a maximum and then decreases, but it does not die down. This maximum drifting of drops makes one think that the vibrations introduced in the drop by the flow patterns are coinciding with the natural frequency of the drop. This behaviour has been observed by Gunn[16] who found that 1/2 mg water droplets drifted 2-3 ft. in 50 ft. of fall in still air.

In view of the fact that all the basic assumptions of the theoretical work are violated, and the theoretical work does not seem to offer much hope, even when the simplifying assumptions were made, it was felt that the actual solution for the present problem should be sought only by experimental methods.

EXPERIMENTAL WORK

The experimental work consisted of (a) the preliminary work done to standardize the experimental conditions with a view to get reproducible and reliable data as well as to ascertain the accuracy of the results thus obtained, (b) the main experimental work done to collect data on the terminal velocities of liquid droplets of different sizes and properties falling through water, and (c) the photographic studies of the falling drop behaviour.

(a) Preliminary Work

The preliminary work comprised of the following:

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

1. Choice of the Systems

Organic liquids heavier than water and insoluble in it were chosen for the discontinuous phase while laboratory distilled water was selected for the continuous phase. The following six organic liquids were chosen in view of the considerations noted against each of them. Only research grade organic liquids were used.

I. Carbon tetrachloride. It is cheap and large quantities of pure material can easily be procured. It has a density of 1.5842 $\frac{\text{gms}}{\text{ml}}$ at 25.0°C., an interfacial tension with water of 41.6 $\frac{\text{dynes}}{\text{cm.}}$ at 24.6°C., and a viscosity very near that of water with $\left(\frac{\mu_i}{\mu_o}\right)$ = 1.040 at 25.0°C. It has an Sd value with water of 2,445.

II. Tetrachloroethane. It has almost the same density as carbon tetrachloride with a value of 1.5813 $\frac{\text{gms}}{\text{ml}}$ at 25.0°C., but has an interfacial tension of 31.3 $\frac{\text{dynes}}{\text{cm.}}$ at 24.9°C. which is much lower than that of carbon tetrachloride. So it was selected with a view to find out the importance of interfacial tension on the drop velocities of liquid droplets. It has a $\frac{\mu_i}{\mu_o}$ = 1.681 at 25.0°C., and an Sd value with water of 1,843.

III. O-Nitrotoluene. It has an Sd value of 2,408 which is very near to that of carbon tetrachloride. However, it has a density of 1.1578 $\frac{\text{gms}}{\text{ml}}$ at 25.0°C., an interfacial tension of 26.6 $\frac{\text{dynes}}{\text{cm.}}$ at 24.2°C., and $\frac{\mu_i}{\mu_o}$ = 2.342 at 25.0°C. It was selected with a view to use the data on O-Nitrotoluene and carbon tetrachloride drops to test the correlations of Hughes and Gilliland.

IV. S-Tetrabromoethane. It is selected for its high density of 2 .9539 $\frac{\text{gms}}{\text{ml}}$ at 25.0°C. It has Sd = 1,424; σ = 36.2 $\frac{\text{dynes}}{\text{cms.}}$ at 23.2°C., and $\frac{\mu_i}{\mu_o}$ = 10.5 at 25.0°C.

V. Ethyl Chloroacetate. It is selected since it has an Sd = 1,359 which is very near to that of s-tetrabromoethane, and the data on both these should serve as a second check on Hughes and Gilliland's correlation. It has $\rho_i = 1.1451 \ \frac{\text{gms}}{\text{ml}}$, $\sigma = 14.6 \ \frac{\text{dynes}}{\text{cm.}}$ and $\frac{\mu_i}{\mu_o} = 1.232$ all at 25.0°C. This is also of special interest because of the very low interfacial tension.

VI. Chlorobenzene. It is of special interest because of a high Sd with water of 3,710, and also of a low $\frac{\mu_i}{\mu_o}$ of 0.8533 at 25.0°C. It has $\rho_i = 1.1008 \ \frac{\text{gms.}}{\text{ml.}}$, and $\sigma = 35.4 \ \frac{\text{dynes}}{\text{cm.}}$ 25.4°C.

The physical properties of the above six liquids are given in Table A-8. The properties of distilled water are taken from literature and used as $\rho_o = 0.99707 \ \frac{\text{gms.}}{\text{ml.}}$ at 25.0°C. and $\mu_o = 0.8937$ centipoises at 25.0°C.

2. <u>Determination of physical properties of the liquids</u>.

The densities of the liquids were determined with a pyknometer at 25.0 ± 0.1°C., using distilled water as a reference liquid.

Viscosities were measured at 25.0 ± 0.1°C. with an Ostwald viscometer, using distilled water as a reference liquid.

Du Noüy tensiometer, catalog number 70545 of Central Scientific Co., Chicago, Illinois, using ring method, was used for the measurement of surface tension of the liquid in air and the interfacial tension of the liquid to water.

The tensiometer was first adjusted so that the reading on the scale corresponds to $\frac{\text{dynes}}{\text{cm.}}$. Zuidema and Waters[48] suggest that the reading obtained by the ring method must be multiplied by a correction factor. A curve giving their correction factor is also reproduced in the pamphlet accompanying the instrument. Zuidema and Pilz[47] have shown that the same correction factors as given by Zuidema and Waters are applicable whether the ring is pulled or pushed through the interface. While taking the interfacial tension in the present work, it was generally found to be convenient to push the ring through the interface from the water to the heavy organic liquid phase. The values of surface and interfacial tension readings corrected by using the factor given by Zuidema and Waters[48] are taken as true values.

3. Production of drops of constant volume.

This work consisted of developing the burets and tips that would obviate the necessity of using any materials that may be soluble in the organic liquids used herein. As the liquids are very good solvents, and as even small quantities of foreign materials in solution will affect the interfacial tension, all tubes like tygon, greases, etc. have been avoided. Actual experience has shown that all of them including teflon should be avoided. The apparatus used for the purpose of producing constant volume drops can be divided into (a) nozzles and (b) burets.

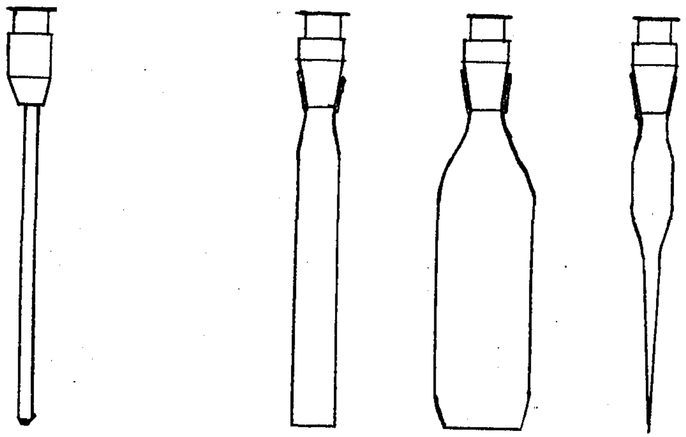
(a) Nozzles: These are used to form the liquid droplets at their tips. To start with, hypodermic needles were used. The oblique cuts on their ends were removed and the tips were rounded off carefully with a file to give a circular tip. The range of drop sizes was extended by joining glass tips to the metal bases of the hypodermic needles. To make a good glass tip with metal base, the regular hypodermic needles were taken, the needle pulled off from its base and the outside of the metal base ground to have a tapering. An ordinary (Kester acid filled) solder was first used on the tip. A glass tube drawn roughly to fit the metal base taper is taken and then joined to the metal base using cerroseal solder supplied by Cerroseal solder corporation, New York City. The drop producing ends of the glass tips were then ground to remove all irregularities in the tip, after drawing them to the approximate size desired. By this procedure all the nozzles produced had the same standard luer grinding of the hypodermic needles. Some of the typical shapes of the nozzles used for the purpose are shown in figure 2.

(b) Burets: These are used for measuring the volume of the droplets. Apart from the question of accuracy of measurement, the other important considerations are: (1) to avoid any grease at any time coming in contact with the organic solvents; (2) 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 (3) to have a tip to which the nozzles can be attached directly without any other connecting tubes.

The following two types of burets were developed for the purpose.

One buret was a 5 x $\frac{1}{100}$ ml. microburet. It was provided with an adjustable flow stopcock of the type shown in figure 3. Though adjustable stopcocks were being made, their use to burets does not seem to be in practice. To avoid any grease, the stopcock had to be precision ground so that it could be worked without grease. A standard luer grinding was provided at the tip to take the nozzles at the bottom. This buret was made to special order by Ace Glass Inc. of Vineland, New Jersey.

The second buret consisted of a 4ft. pyrex glass capillary tube bent to the shape shown in figure 4. The horizontal length of the tube was 40 cms., giving 30 cms. for the graduated scale. The two legs of the U were 3 cms. apart. The distance of the horizontal tube between U and the next vertical bend was 15 cms. At the end of the capillary was fused a tuberculin syringe of 1 cc. capacity. The end of the syringe had a standard luer grinding and could take on the usual hypodermic needles. This ground end was 11 cms. long. At the higher end of the tube was fused an adjustable flow stopcock of the same type as shown in figure 3. The center of this stopcock was 8 cms. above the horizontal portion of the capillary. This provided the variable flow control.



Hypodermic needle Glass tips with bases of hypodermic needles

FIG.2 GLASS AND HYPODERMIC NEEDLES

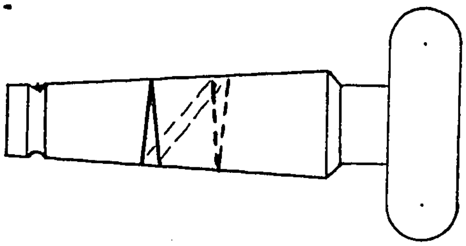


FIG. 3 ADJUSTABLE FLOW STOPCOCK

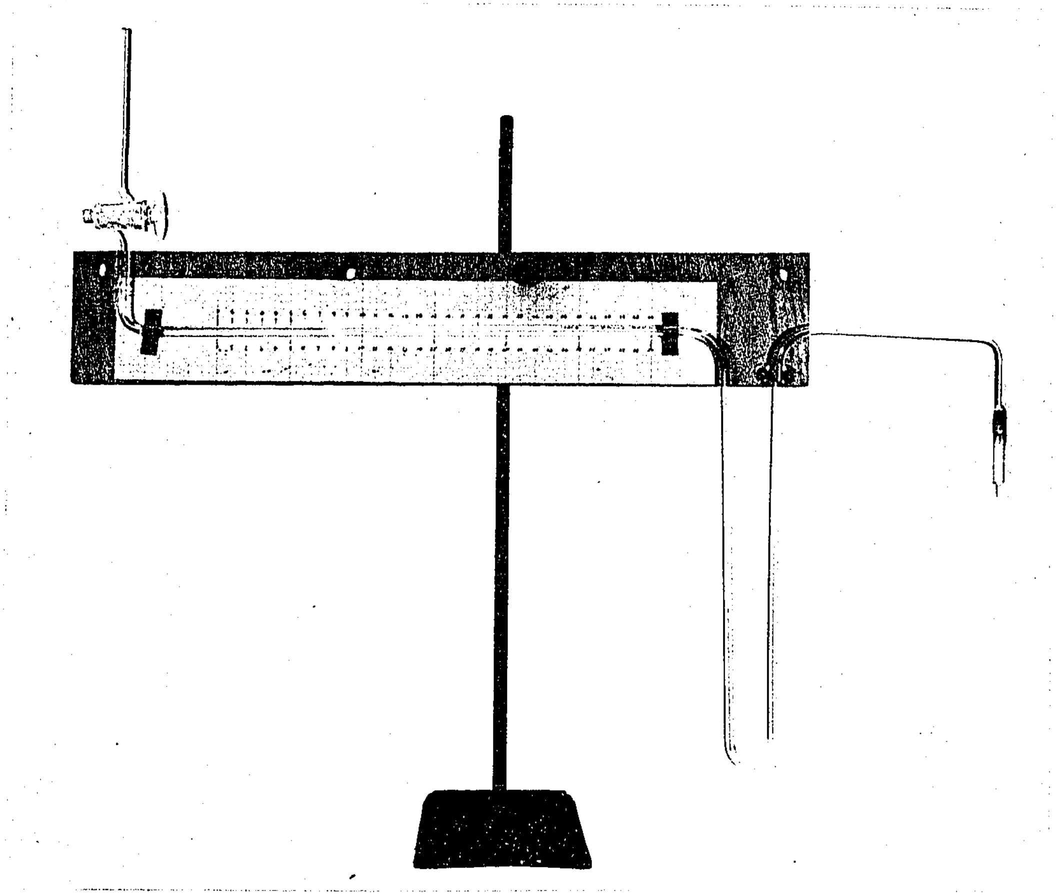


Fig. 4. Capillary U-tube Microburet

Since it was in contact only with air and not the solvents, a slight amount of grease could be used and hence it did not require precision grinding as in the previous case. The horizontal portion of the capillary was mounted on a wooden board on which a graph paper was pasted. This horizontal portion was calibrated by sucking a small pellet of mercury, measuring its length at various positions in the buret and weighing it later. Knowing the density of mercury, the capacity of the microburet for each position was calculated, and a calibration curve for the buret was obtained. Just to give an idea, it may be mentioned that 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 buret gave more uniform drop formation times than the other one.

4. Determination of the initial height of fall required for the drops to attain terminal velocities.

Since the present work is concerned with the terminal velocities of liquid droplets, it was necessary to know how far the liquid droplets should be allowed to fall initially before they can be considered to have attained the terminal velocity. For this purpose a 6 in. diameter by 6 ft. long lucite column was filled with distilled water. In the beginning, carbon tetrachloride was chosen and its drops allowed to form about 3-5 cms. under the water surface.

Distances of 30 cms., 60 cms. and 160 cms. from the drop formation point were marked on the column. Then the drops were allowed to fall, and the time of fall of droplets from 30 to 160 cm. marks, as well as for 60 to 160 cm. marks, were noted simultaneously on several drops, using two stop-watches. The average of each of them was taken. If the ratio of the time of fall from 30-160 cms. to the time of fall from 60-160 cms. was within 1% of the value 1.30, the drops were considered to have attained the terminal velocity. It was found that in most cases of the work the first 30 cms. of fall was enough for the drops to attain the terminal velocities. Now and then the distance 60-160 cms. was used in other systems to serve as a check on this assumption, and there was no appreciable difference observed.

5. Elimination of wall effect and end effects.

For this purpose fall velocity data were collected for carbon tetrachloride drops in water using a 3 inch diameter glass column and a 6 inch diameter lucite column. The data are presented in Tables A-1 and A-2, and plotted in Figure 8 . The data in the three inch column were obtained up to D = 0.448 cms. of the drop. It can be seen that there is no noticeable difference between the fall velocity data in the 3-inch and 6-inch diameter columns. However, for additional safety, and convenience, the 6-inch column was used for later work. Also, if there is no wall effect in the 3-inch column up to

D = 0.448 cms., there should be no wall effect in the 6-inch column at least up to D = 0.896 cms. This gives a wide range of drop sizes to work with.

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

6. Temperature control.

In the early stages of the work, the temperature of the water was adjusted to 25.0±0.1°C. and it was then poured into the 6-inch column. Because of the large capacity of this quantity of water in 6 inch dia. 6 ft. long column, the water retained its temperature to within 25.0 ± 0.5°C. for several hours. Later when the weather became hot, it was found necessary to have temperature regulation. The 6 inch dia. column was surrounded by an 8 inch square column of 5' - 10" height. Water adjusted to a temperature of 25.0 ± 0.1°C. was pumped continuously into this square column and the overflow allowed to flow back into the constant temperature tank. The temperature regulating equipment is shown in Figure 5.

7. Estimation of errors.

The detailed estimation of the errors is given in Appendix . They are summarized in Table III. The total of all errors of 8.20% is only indicative of the maximum possible errors involved. In actual practice much closer agreement was obtained. But a ± 8.20% variation should be borne in mind when comparing entirely different systems with a view to find a general correlation.

TABLE III SUMMARY OF ERRORS

Measurement	Error
(a) Physical properties	
Density	0.004%
Viscosity	0.20 %
Interfacial tension	0.40 %
(b) Drop volume	3.0 %
(c) Times of fall of the drops	2.17 %
(d) Distance of fall of the drops	0.078%
(e) Variation in physical properties for a variation in temperature of 0.5°C.	1.35 %
(f) Errors due to initial turbulence, and turbulence produced by drops themselves	negligible
(g) Errors due to solubility effects	1.0 %
(h) Errors due to zig-zag path followed by drops)	Considered to be inherent in drop behaviour
(i) Errors due to oscillations)	
Total of all errors	8.20 %

(b) Main Experimental Work

Apparatus

The general arrangement of the apparatus is shown in Fig. 5 and two photographic views of it are given in Figs. 6a and 6b. The main portion of the apparatus consists of a column filled with water and a buret with a nozzle attached to it to produce the liquid droplets. All the rest of the apparatus was built around these things. The burets have been already discussed under the preliminary work. The details of the columns used are given in Table IV. As was already mentioned, 3 inch and 6 inch diameter columns were first used for checking the question of wall effect. Later only the 6 inch diameter column was used for all the experimental work. The positions of the different marks used on the columns for timing the fall of the liquid droplets are also given in the table. Lucite was selected for the 6 inch column on account of its transparency and cheapness. It has also the additional advantage over glass of being less fragile, light in weight and ease of working. Lucite tubes can be joined together with a cementing liquid (a liquid solvent supplied by the makers of lucite). The 6 inch lucite column was attached at the base to a 1/2 inch thick lucite sheet.

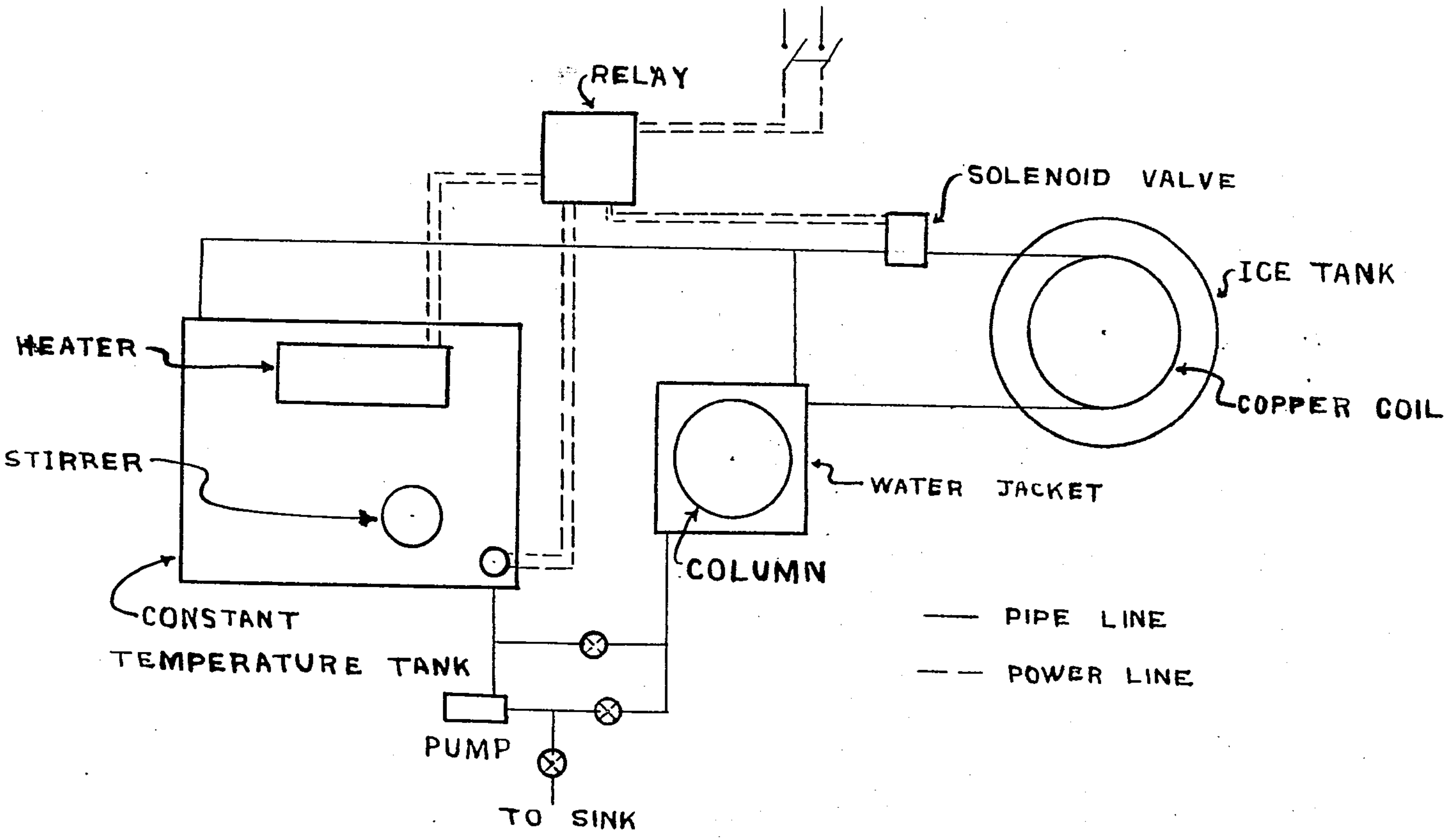


FIG.5 APPARATUS FOR STUDY OF RATE OF FALL OF LIQUID DROPLETS (Flow Diagram)

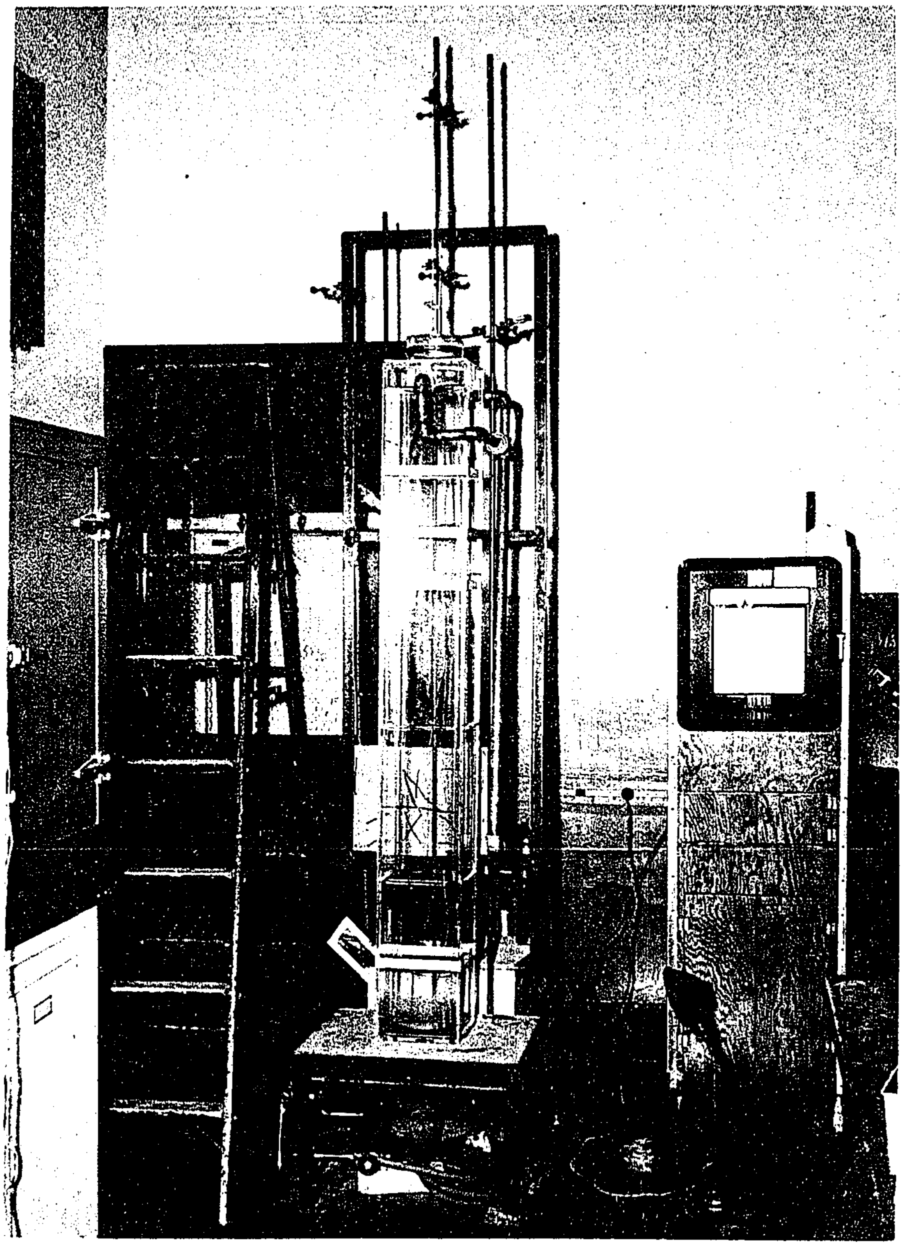


Fig. 6a. Apparatus for Study of Rate of Fall of Liquid Droplets. (front view)

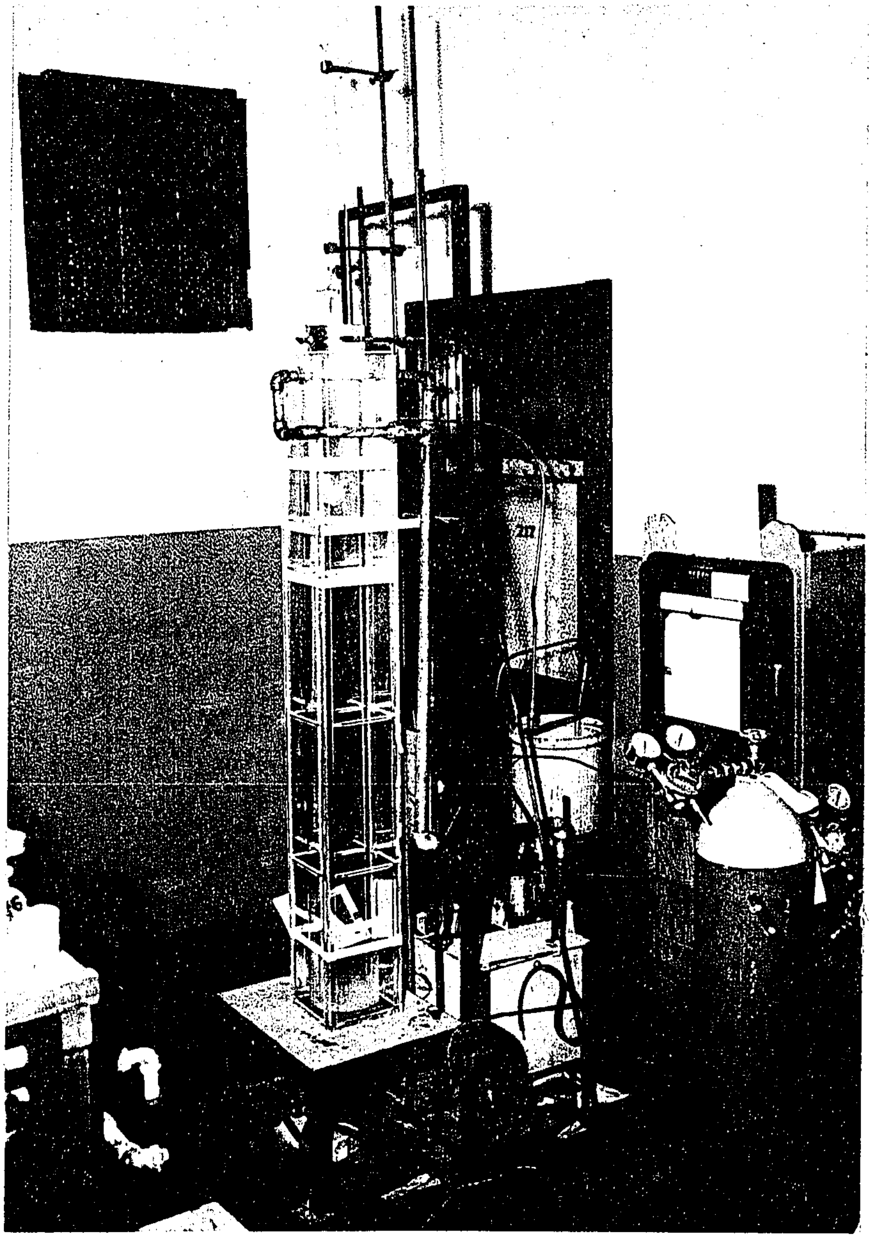


Fig. 6b. Apparatus for Study of Rate of Fall of Liquid Droplets. (oblique view)

TABLE IV

DETAILS OF THE COLUMNS USED

	Column I	Column II
Nominal size	6 in.	3 in.
Inside diameter	5 3/4" or 14.6 cms.	7.0 cms.
Wall thickness	1/8" or 0.32 cms.	0.24 cms.
Outside diameter	6" or 15.24 cms.	7.5 cms.
Total length above base	6' or 182.9 cms.	5'-8 1/2" or 174.0 cms.
Material of the column	Lucite	Pyrex glass
Position of end of drop forming tip from the top of column	3.0 cms.	3.0 cms.
Position of 1st mark from the tip of the nozzle	30.0 cms.	30.0 cms.
Position of 2nd mark from the tip of the nozzle	60 cms.	60.0 cms.
Position of 3rd mark from tip of nozzle	160 cms.	160.0 cms.
Position of 3rd (last) mark from bottom of the column	19.9 cms.	11.0 cms.
Closure for the bottom	1/2" thick lucite sheet	Rubber stopper

Mirrors have been used to bring the bottom mark just a little below the 30 cm. mark. This served not only as a convenience, but also helped in avoiding any vibrations being introduced in the column due to climbing up and down the ladder by the side of the column in order to take the readings. The bottom 160 cm. mark was generally illuminated with an electric light placed about one meter away.

As lucite is affected by the organic solvents used in the present work, an 800 ml. beaker was placed at the bottom of the 6-inch column to collect the organic liquid droplets at the bottom. At the end of the day's experiment, the beaker could be removed with the help of the copper wires tied to it. There is a drain with a stopcock at the bottom of the column.

All the rest of the apparatus consists of the equipment for temperature control. The 6 inch diameter column is surrounded by an 8 inch square column of clear plexiglass, 5 ft. 10 inches in height. Water could be pumped continuously from the constant temperature tank into this 8 inch square column by means of a pump. The water from the column can overflow back to the tank. The tank was 18" long, 14" wide and 14" deep. A small side stream from the 8" square column was taken through a copper coil in an ice tank and then connected to the main overflow line through a solenoid valve. This valve was actuated by a relay so that it could open if the temperature of water in the tank went high. Then cold water would flow in and cool the water in the tank. There is also an electric heater by which the water could be heated if the temperature went below 25.0°C. This heater also was connected to the relay. The relay was controlled by a thermoregulator. By this arrangement a temperature of 25.0 ± 0.1°C. was maintained in the column.

It should be mentioned that the pump, the constant temperature tank and the solenoid valve were connected to the 8" square column with small rubber tubing in the lines to prevent any vibrations from travelling to the column. Also the columns were set vertical by using a plumb bob.

Procedure

Though several modifications of procedure were tried in the early stages of the work, the following procedure was finally adopted as the one giving the most consistent and reliable results.

Before starting this work, one should remember that interfacial tension property is very sensitive to small impurities. Since the liquids used in this work were very good solvents, special care should be exercised to prevent them from picking up impurities. As will become evident in the later part of this work, interfacial tension is a very important factor controlling the rate of fall of liquid droplets.

The procedure for achieving the cleanliness required for this work is, of course, very simple. After a day's work the burets were cleaned with a cleaning mixture of concentrated sulphuric acid containing potassium dichromate, and then washed at least six times with distilled water. They were then dried at the laboratory temperature. The capillary U-tube buret had to be dried by aspirating air through it, while the 5 x 1/100 ml microburet could be allowed to dry by itself in the laboratory. Similarly, the hypodermic and glass needles required cleaning after they were used. They could all be cleaned conveniently with repeated treatment with chemically pure grade carbon tetrachloride for at least six times. Also it was necessary to clean all the beakers, etc., used in this connection,every day with the cleaning mixture, washed thoroughly with distilled water and dried. The beakers, etc., can, of course, be dried in an oven, though pipettes should be dried only at the laboratory temperature. The 3-inch and 6-inch columns were first throughly washed with distilled water. Afterwards the water in them was drained after a day's work. They were covered at the top with a cardboard to prevent dust and dirt getting in when not in use. The next day they were ready for use, but in general the columns were rinsed with a small quantity of distilled water, before filling them with water. It was also felt necessary to use research grade organic liquids for the discontinuous phase and laboratory distilled water for the continuous phase.

The procedure herein described is for the 6-inch diameter column on which all the data was collected. The procedure for the 3-inch column was much simpler as it was filled with distilled water whose temperature was adjusted to 25.0 ± 0.1°C., and the data collected without any outside temperature control. Also there was no such beaker as was used in the 6-inch diameter column.

The 800 ml beaker was first lowered into the 6-inch column with the help of the copper wires tied to it. The drain cock at the bottom of the column was closed. Next the column was filled with distilled water. If the distilled water temperature was much lower than 25.0°C., the water was first collected in clean buckets and its temperature adjusted to 25.0 ± 0.1°C. by the addition of distilled water heated in a beaker over a bunsen flame. Then the water was poured into the column. When the distilled water temperature was higher than 25.0°C., it was found convenient to fill the column with the distilled water by connecting a rubber hose to the distilled water tap. After the column was nearly full, ice cubes (prepared from distilled water) were introduced into the column and the temperature adjusted to 25.0 ± 0.1°C.

The constant temperature tank was filled with ordinary laboratory tap water. The ice tank was filled with ice. The stirrer in the tank was started and the water temperature was adjusted first by addition of ice with hand, to about 25.0°C. The pump was started and as soon as the 8-inch square column was full, the thermoregulator was turned on. Then it controlled the temperature of the water in the tank to ± 0.1°C. of 25.0°C.

The beakers and burets were rinsed first with the organic liquid to be used. The cleaned and dried hypodermic or glass nozzle was attached to the bottom of the burets provided with the standard luer grinding. The capillary U-tube buret was invariably used for the small size drops while the 5 x 1/100 ml microburet was used for the large size drops. As a check certain medium sized drops were obtained with both burets. When the capillary U-tube buret was used, the organic liquid was sucked through the nozzle, and also let flow through the nozzle. After the capillary buret was filled with the liquid, it was mounted on the stand (supporting the main column) with the tip of the nozzle centered at the top of the column. The 5 x 1/100 ml microburet could be filled with the liquid from the top and could be drained from the bottom. In the case of large sized drops, it was found more convenient to suck the liquid through the glass, nozzle at the bottom, by dipping the end in a weighing bottle containing the liquid. Next the buret and weighing bottle were taken to the stand with the bottom of the nozzle submerged under the liquid. The buret was mounted above the buret in an approximately centered position held by two clamps. Next the buret and weighing bottle were slowly lowered till the bottom end of the nozzle was at the position desired. This position was 3.0 cms. from top of column or about 2.5 cms. below the top water level in the column. The weighing bottle was then carefully removed from under the nozzle and out of the water. This special precaution was necessary in the case of large size drops as any small vibrations were enough to make the nozzles drain themselves and not remain full. With the medium sized nozzles there was no such trouble, and the buret with the nozzle attached to it could be mounted on the column and centered.

All the above work after filling the column generally took more than 1/2 hour and that time was considered enough for the water in the 6-inch column to settle down. A check on the temperatures at the top and bottom of the columns was taken. Generally, about 1 hr. after the column was first filled or 1/2 hr. after the column reached the equilibrium temperature of 25.0 ± 0.1°C., the data were collected.

The buret stopcock was slightly opened 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 size drops. Also the rate of flow should be slow enough so that readings of the liquid levels in the buret could be taken without much error. After the rate of flow was adjusted, the liquid level in the buret was noted as soon as a drop detached itself from the tip of the nozzle. This drop was considered as zero drop. The next drop was counted as the first drop. Then as each drop passed the first mark (30 cms. below the point of drop formation), a stopwatch was started and the stopwatch was stopped when the drop passed the mark 160 cm. below the point of drop formation. The time taken for the drop to travel 130 cms. was read off from the stopwatch. For each run the times of fall of as many drops as possible were noted. The time of fall of the last drop of the run could not be counted, as the liquid level had to be taken as soon as the last drop detached itself from the tip. The total number of drops was noted. The volume of the liquid run down was also noted. From these two values, the average volume of the drops was calculated. During each run the time of formation of each fifth drop was generally noted. In the tables of data given, only the lowest and highest values of drop formation times were noted.

After each run, the nozzle was removed and another clean and dry one fixed to the buret. The buret was filled, the tip of the nozzle centered and submerged in water in the column to the same position as before, and the next run made in a similar way as before. It may be mentioned that each nozzle was cleaned throughly before it could be used a second time.

The initial liquid levels in the burets were generally as near the zero mark as possible. The volume of the liquid run down for each run was about 0.5 ml with the 5 x 1/100 ml buret and 0.15 ml (which represented about 20 cm. length of capillary) on the capillary U-tube buret. These values were only general and variations from them were also adopted several times for the sake of convenience. So also, though the fall time of drops was noted during their fall from the 30 to 160 cm. marks in general, yet some data were collected for their fall from 60 to 160 cm. marks, to serve as a check. For each run the initial and final temperatures of water at the top of the column and of laboratory near the center of the column were taken. As it was found that the laboratory temperature was not of any consequence as long as the water temperature was contolled, only the water temperatures were given as part of the results.

(c) Photographic studies

The theoretical study of the shape of the drop has increased the interest to find out what the actual shape of the drops will be. Blanchard's[6] reference to the oscillations and rotations of water drops in air is a matter of special interest because it violates the assumption of equilibrium shape for the drop while working out the problem from theoretical considerations. Also it was felt necessary to record photographically certain observations made on the drop behaviour during the course of the experimental work.

Since the drop in motion had to be photographed, a very short exposure time was necessary. Preliminary attempts using the ordinary flash bulb and 1/400 sec. exposure showed blurring in the image of the drop due to its motion. So it was felt that a high intensity flash of very short duration would be necessary. Since there was the question of drop vibrations it was felt that multiple images of the same drop be taken at small intervals of time. The Strobolux working in conjunction with a Strobotoc (manufactured by General Radio Co., Cambridge, Massachusetts) seemed to meet these requirements very well. It produces high intensity flashes of a duration of 15 to 50 microseconds at frequencies adjustable from 1 to 6,000 per minute. A 4" x 5" Crown Graphic camera with 135 mm. lens, made by Graflex, Inc., Rochester, New York, was used for taking the pictures. A suitable emulsion was found to be Eastman Kodak Super XX.

The arrangement of the apparatus set up for taking the pictures is shown in Fig. 7.

In order to take the pictures of drops, the same procedure as for the determination of drop velocities was followed. This time when a drop fell in the center of the column, it was photographed. A strobolux frequency of ten flashes per second was found to give satisfactory results. A white paper was used on the back and side of the 8 inch column to reflect the light from the strobolux back on to the drop. The strobolux had to be placed along a diagonal line of the 8 inch column in order that the camera can be placed right in front of the column and both can be on almost the same level. A camera lens opening of $\frac{f}{11}$ was generally used. A 1/2 second exposure, resulting in 4 or 5 drop images was found to give good results. Dichlorofluorescein was added to some of the liquids used for producing drops in order to increase the reflectivity of the drops. Since the addition of dichlorofluorescein changes the properties of the drop, the photographic data should be considered more as qualitative and less as quantitative. Drop sizes and velocities were generally noted for each of the drops photographed.

The wires used for introducing the 800 ml beaker into the 6-inch column were used for focussing the camera. The picture of the drop was taken when it fell in a line between the two wires. This way the drop could be obtained into a reasonably good focus. Since the drop is a small object, the camera had to be placed as near the column as possible to get a good size picture. A distance of about 21 inches from the center of the column to the camera lens was found to be convenient.

In this connection it may also be mentioned that preliminary attempts at taking pictures when the 6-inch diameter column was not surrounded by the 8 inch square column were not so successful on account of the curved surface of the column. When the 8 inch square column was built around the 6 inch diameter column and both were filled with water, there

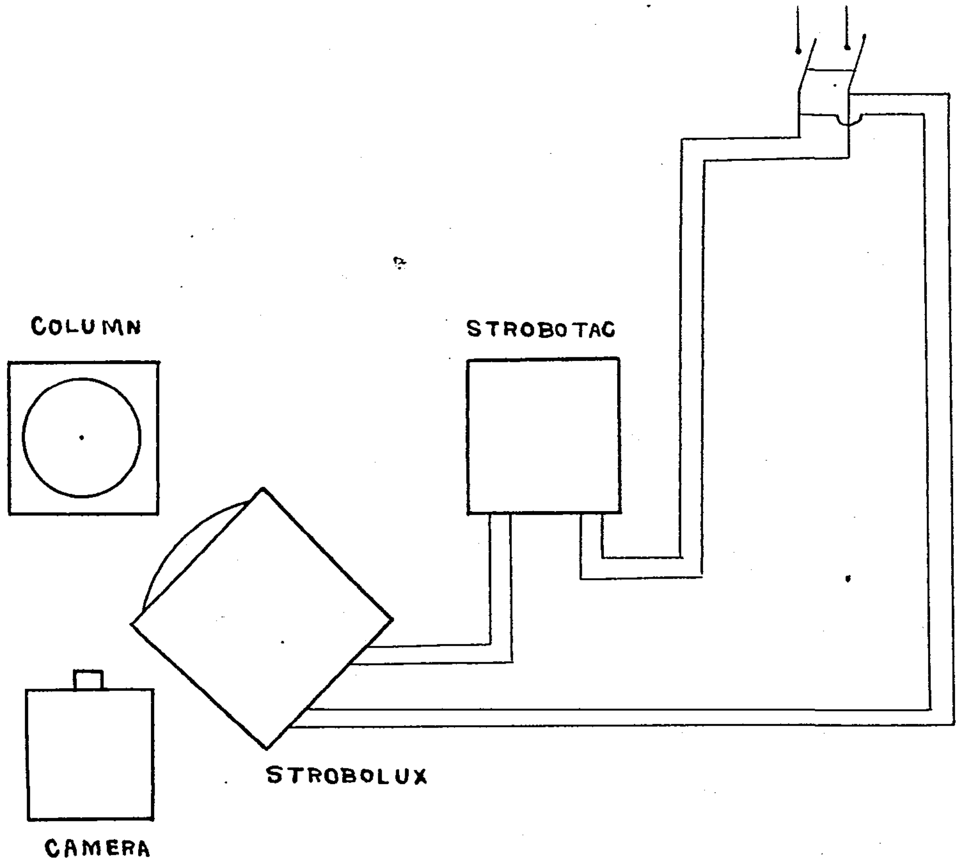


FIG. 7 APPARATUS FOR TAKING PICTURES OF DROPS.

seemed to have been a correction applied for the refractive index of lucite of the round column by that of the square column. In any case, the visibility was actually improved and it became more easy to take the pictures.

Finally the pictures of the drops thus obtained were enlarged and printed on Eastman Kodak Kodabromide F4 paper so as to have the pictures of drops in a reasonable size.

RESULTS

The data obtained on terminal velocities of the various liquids are presented in the tables and graphs as described below.

The fall velocity data in 3 inch and 6 inch diameter columns for carbon tetrachloride drops in water are given in Tables A-1 and A-2 respectively. They are both shown in Fig. 8. A smooth curve was drawn only for the data in the 6-inch column. The values for 3-inch column were plotted and left without curve, so that these points could be compared directly with the smooth curve for the 6-inch column. Also, for every system a smooth curve was drawn so that the smoothed out data could be used for further calculations. As there was no such necessity in the case of the data in the 3-inch column a smooth curve was not drawn.

The fall velocity data for tetrachloroethane drops in water in the 6-inch column are given in Table A-3 and shown in Fig. 9.

The fall velocity data for o-nitrotoluene drops in water in the 6-inch column are given in Table A-4 and shown in Fig. 10.

At this stage it may be mentioned that all the data on carbon tetrachloride drops in 3-and 6-inch columns and o-nitrotoluene drops in 6-inch column and part of the data on tetrachloroethane drops in the 6-inch column were obtained without external temperature regulation. In other words, the column was filled with water whose temperature was adjusted

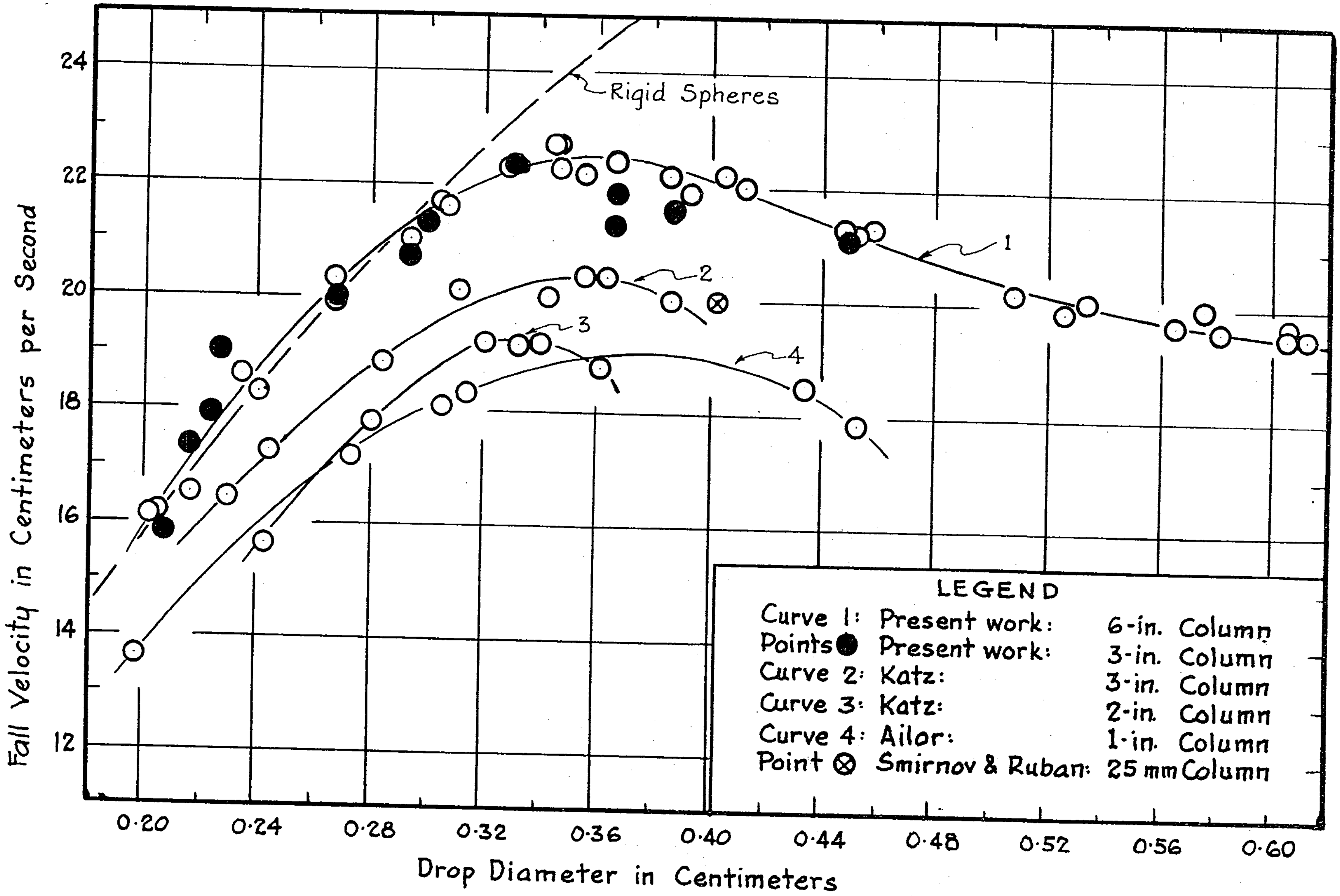


FIG.8 FALL VELOCITIES OF CARBON TETRACHLORIDE DROPS IN WATER

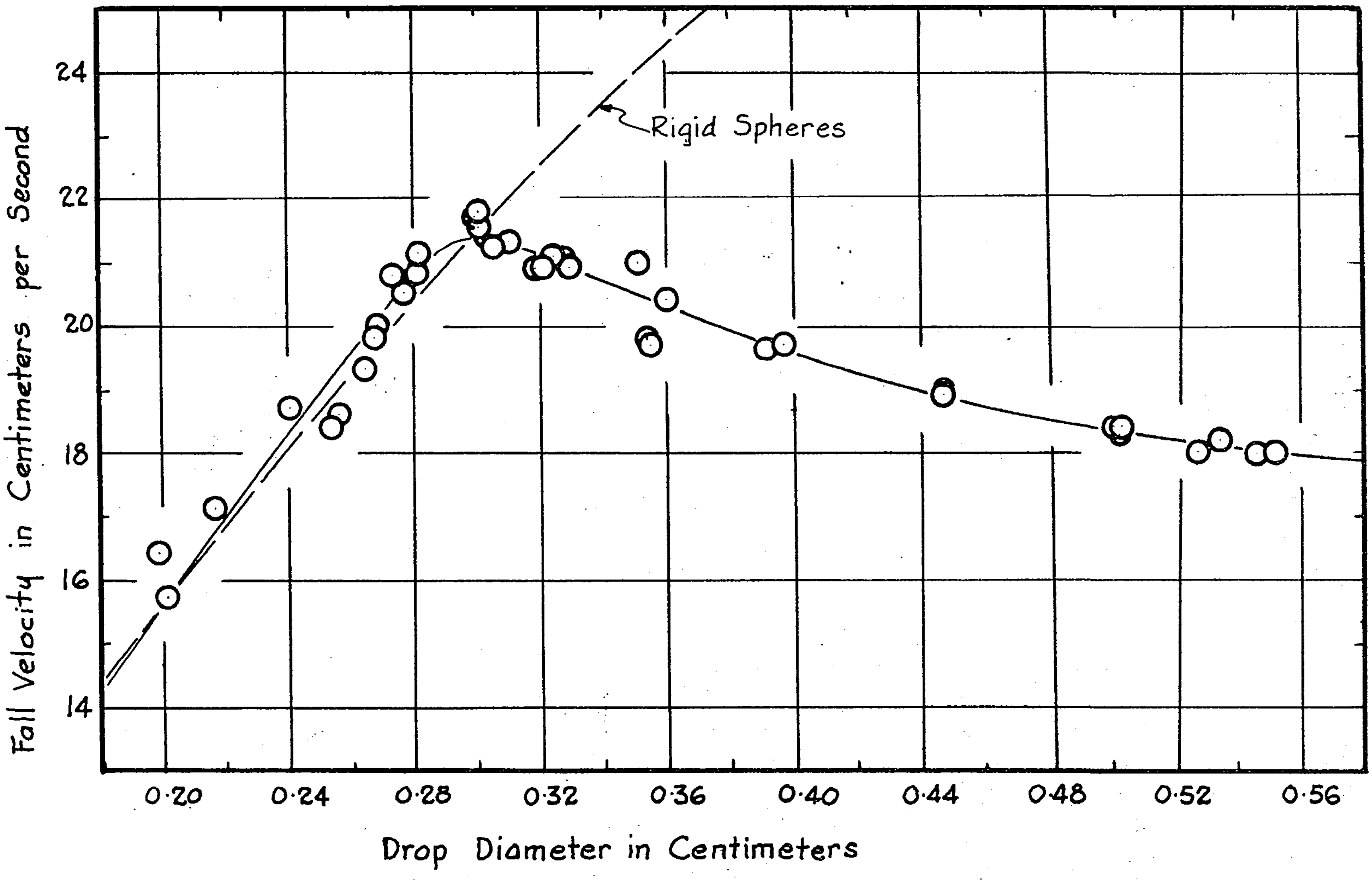


FIG. 9 FALL VELOCITIES OF TETRACHLOROETHANE DROPS IN WATER

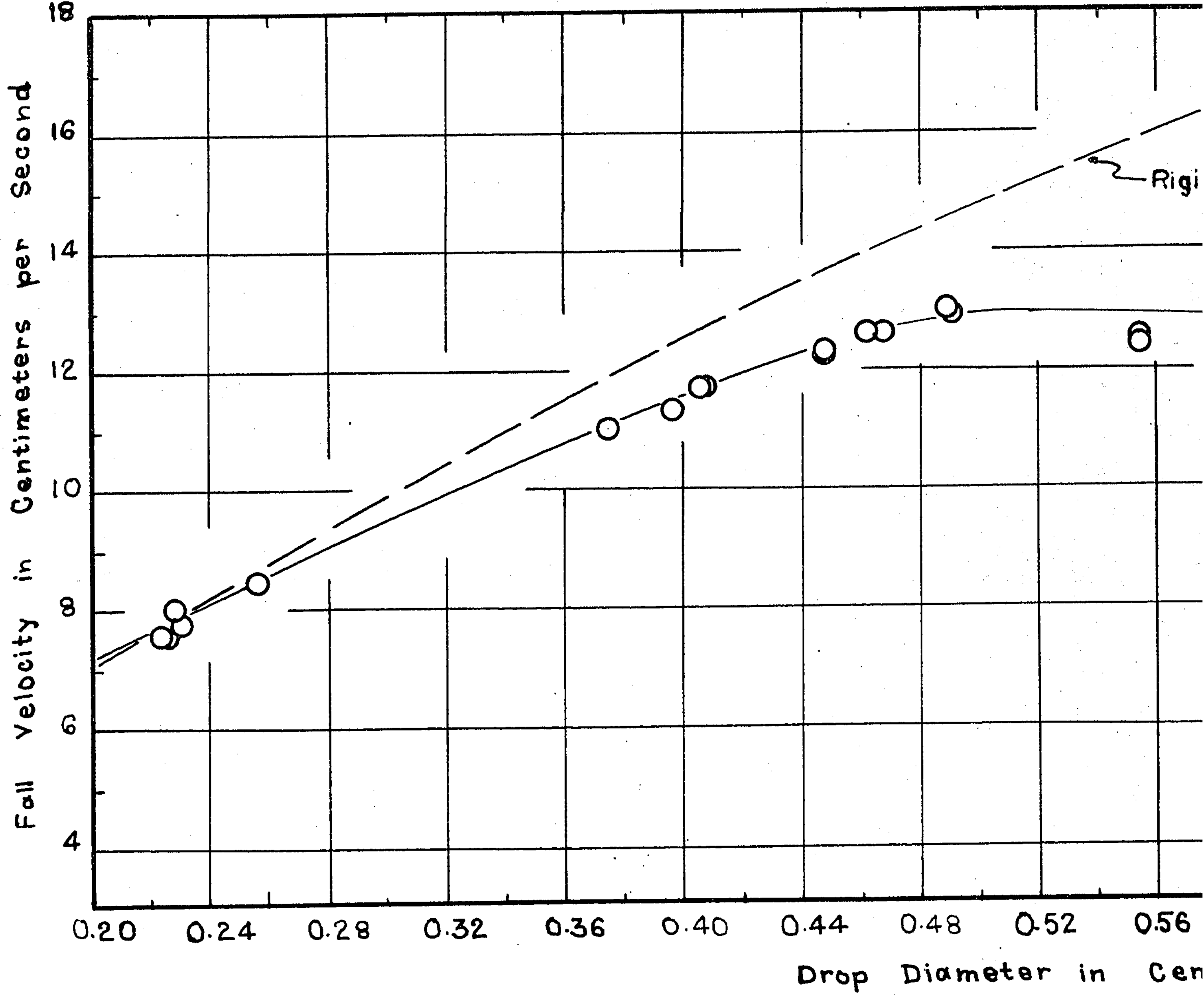
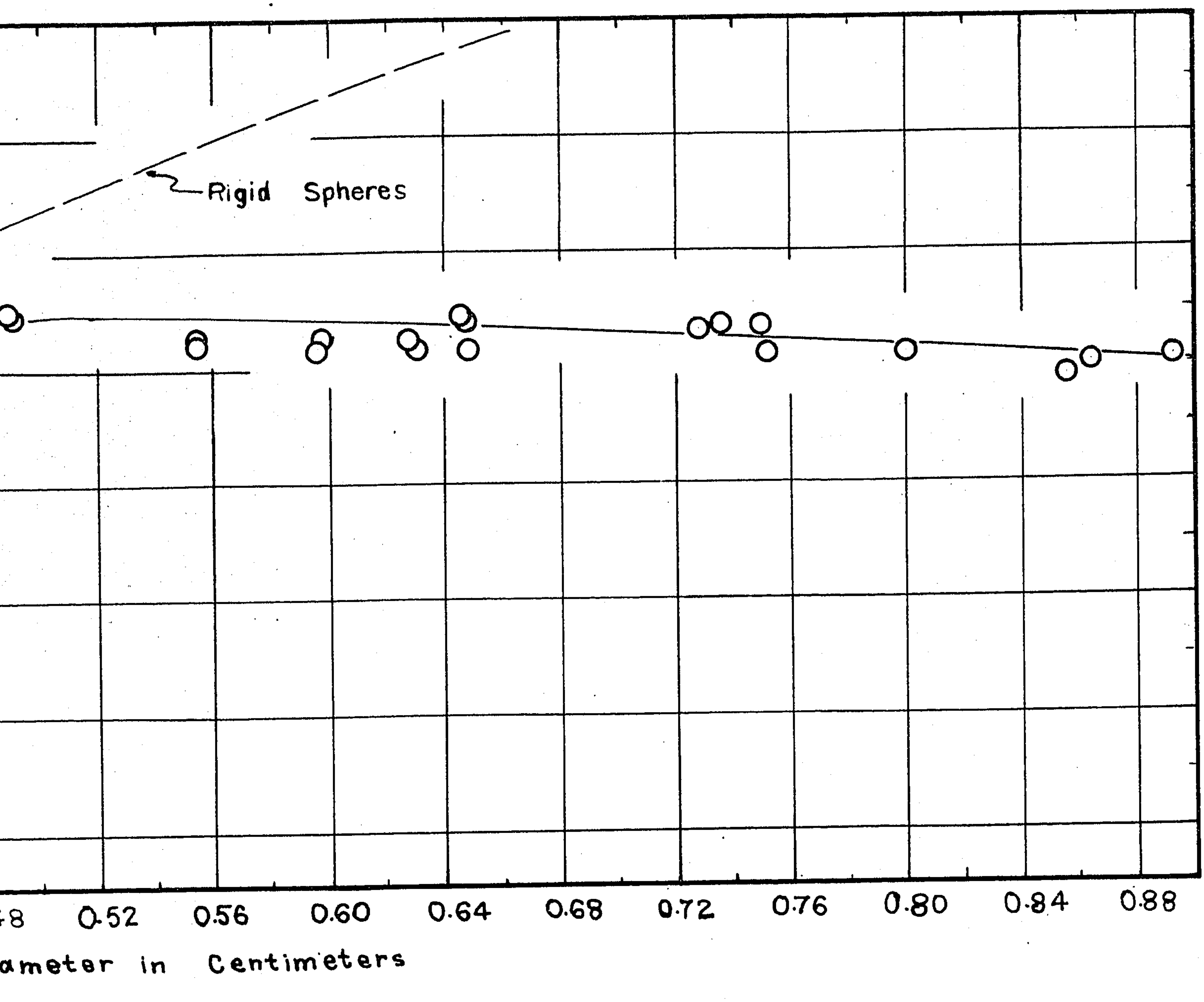


FIG. 10 FALL VELOCITIES OF o-NITROT



o-NITROTOLUENE DROPS IN WATER

to 25.0 ± 0.1°C., and the fall velocity data collected as long as the column temperature was within ± 0.5°C. of 25.0°C. Fortunately, during this time the laboratory temperature was also around the same temperature and hence data could be collected for a sufficiently long time. Part of the data on tetrachloroethane drops was also conducted with the temperature regulation arranged. Several readings were taken after the temperature regulation was brought in. As there was no difference in the fall velocities, the data taken with and without temperature control were not separated. Also some data were collected in the case of tetrachloroethane drops, without the beaker and wires in the 6-inch column. It was found that the presence or absence of the 800 ml beaker and the wires attached to it made no observable difference in the fall velocity data in the 6-inch column.

In the case of o-nitrotoluene drops, very large drop formation times were tried to find out if the frequency of the drops made any difference in the fall velocity data. In the range of drop frequencies studied there was no such difficulty, and the data were considered to have been collected in a stationary medium without any turbulence effects. Drop velocity data on large size drops of o-nitrotoluene were collected from 60 to 160 cm. marks. This data also showed no appreciable difference.

The fall velocity data for s-tetrabromoethane drops are given in Table A-5 and shown in Fig. 11. The fall velocity data for ethyl chloracetate are shown in Fig. 12 and the data presented in Table A-6. The fall velocity data for chlorobenzene drops are given in Table A-7 and shown in Fig. 13. The data on these three systems were collected using the temperature control for the column.

The physical properties of the liquids used are given in Table A-8. The values of density, viscosity, surface tension and interfacial tension were experimentally determined for each organic liquid separately. The literature values, wherever available, are shown for purposes of comparison. Only the experimental data were used for further calculations. The solubility data were not determined, since those data were not used for any of the calculations afterwards. It was, however, considered advisable to include the available literature data on solubilities, in order to have an idea of the systems under consideration.

The range of drop sizes obtained 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 produced was fixed when the drops fell with almost the same velocity as rigid spheres. In the case of chlorobenzene drops, the

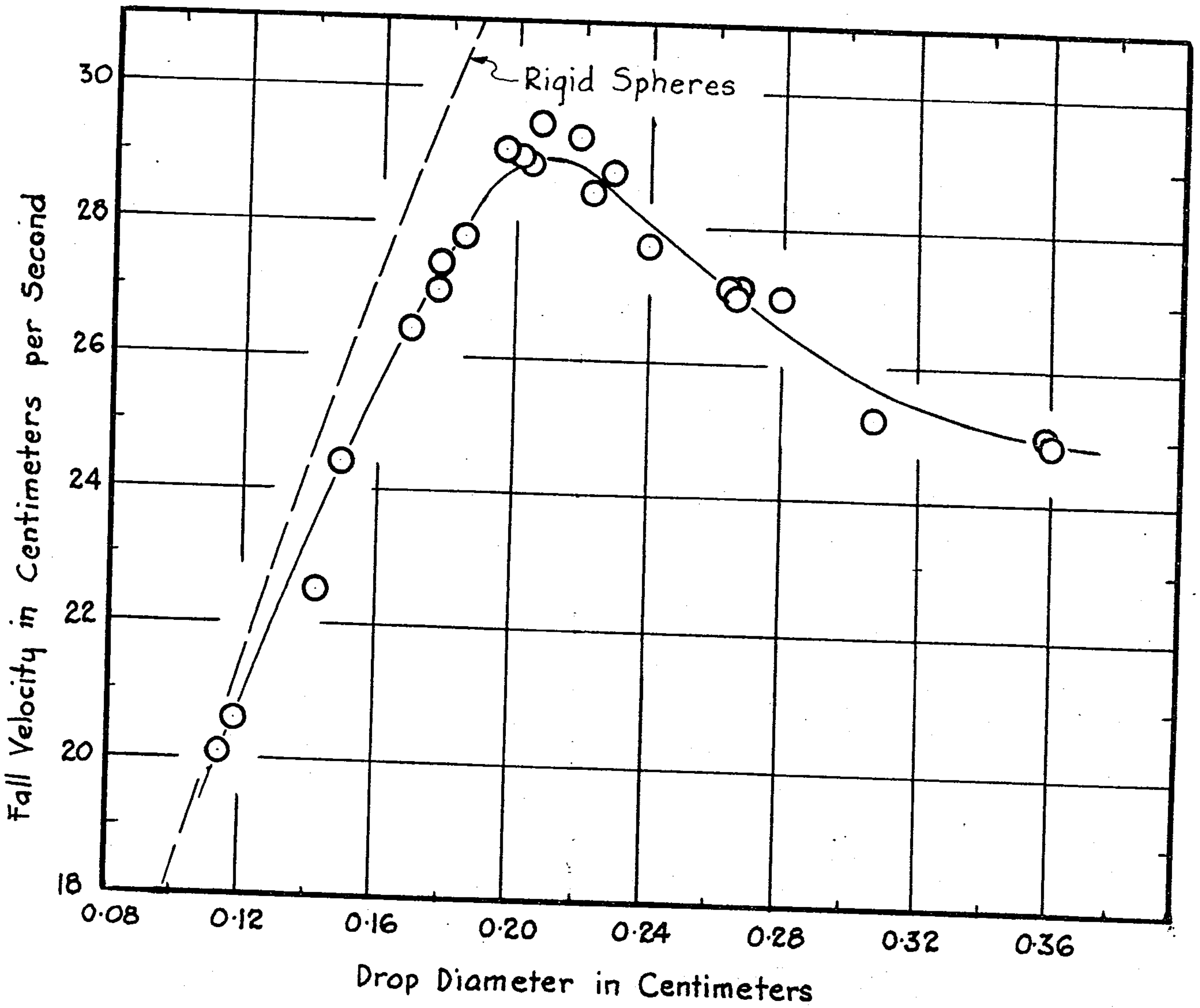


FIG. II FALL VELOCITIES OF s-TETRABROMOETHANE DROPS IN WATER

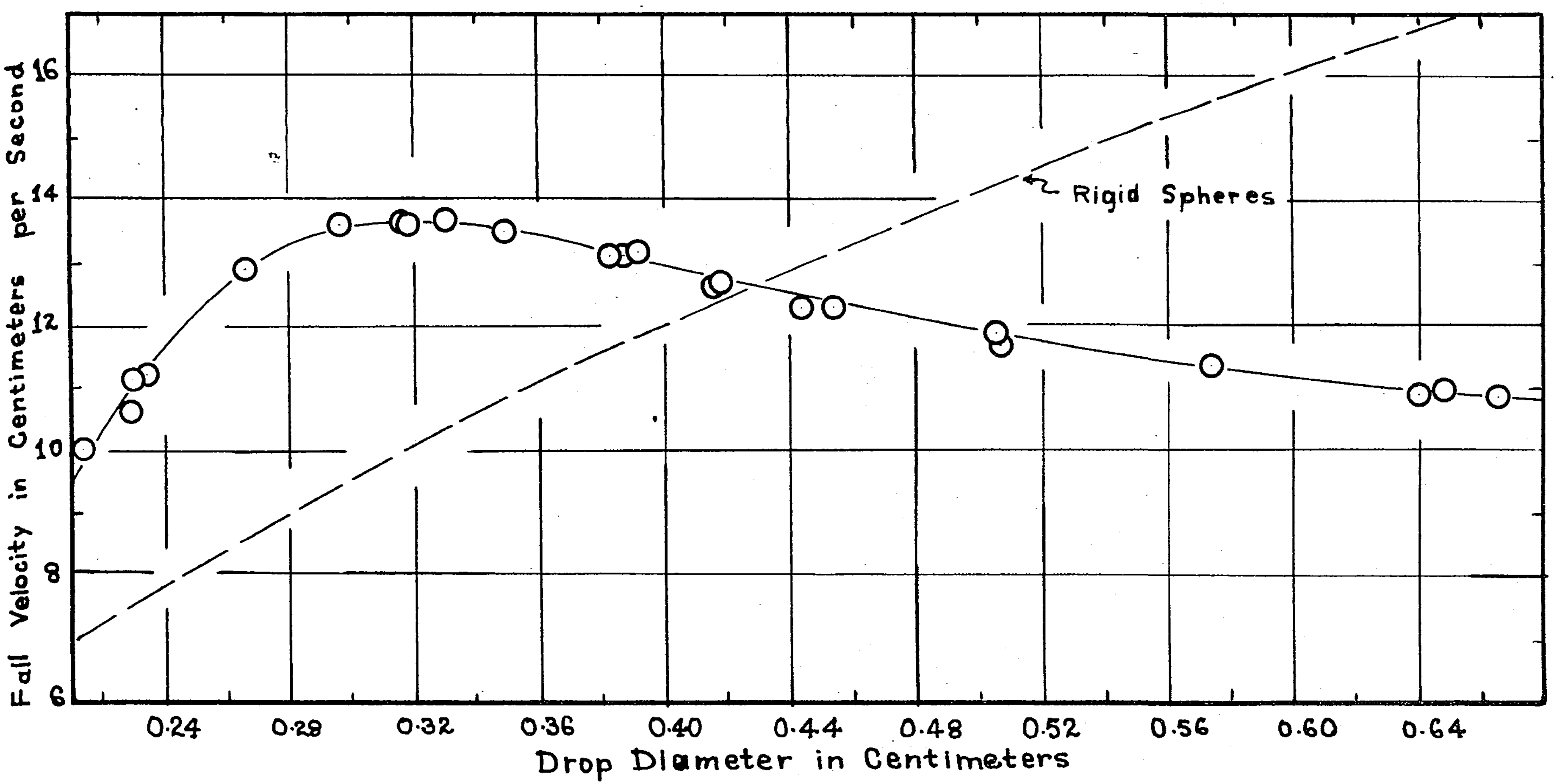


FIG.12 FALL VELOCITIES OF ETHYL CHLOROACETATE DROPS IN WATER

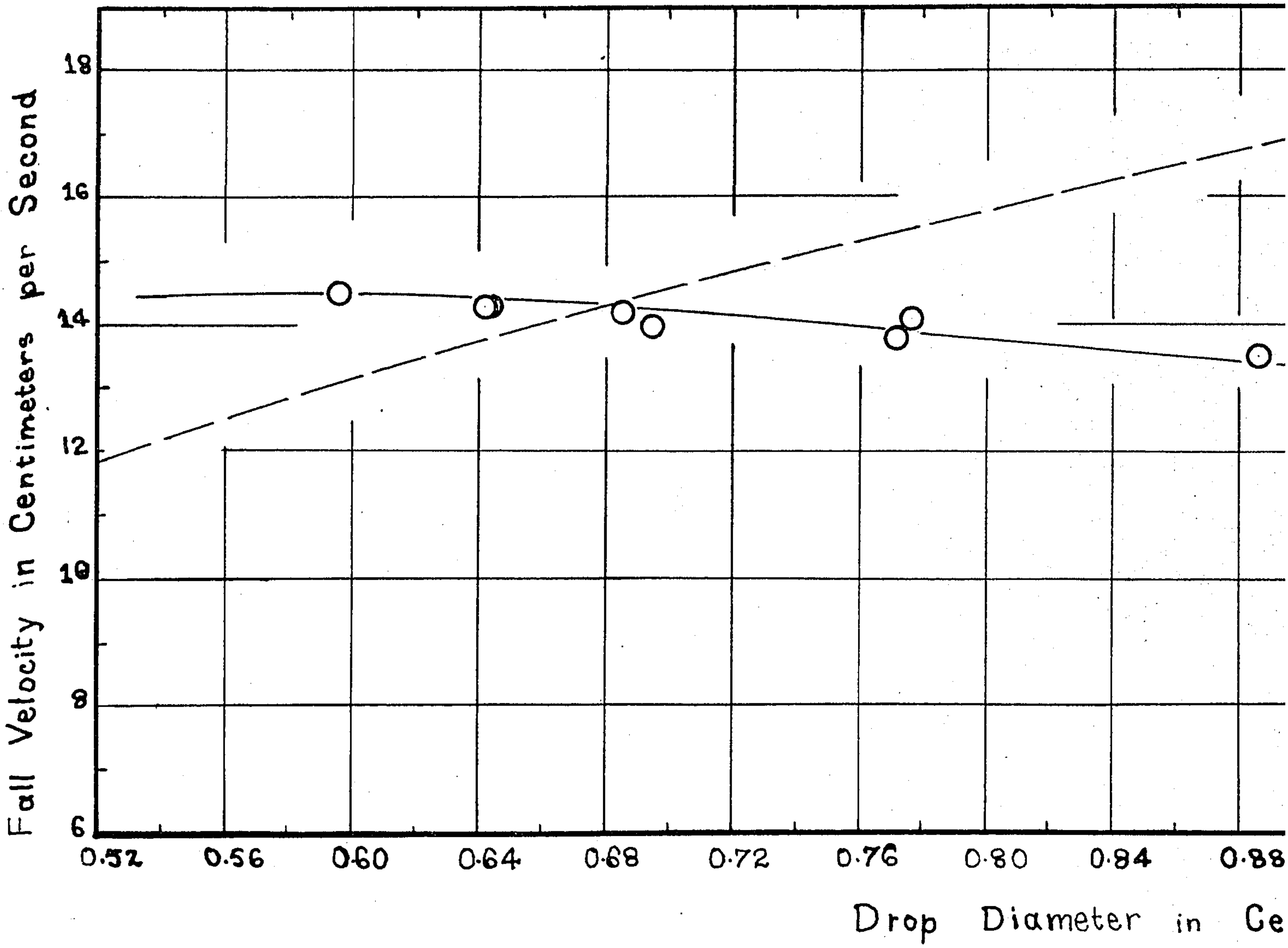
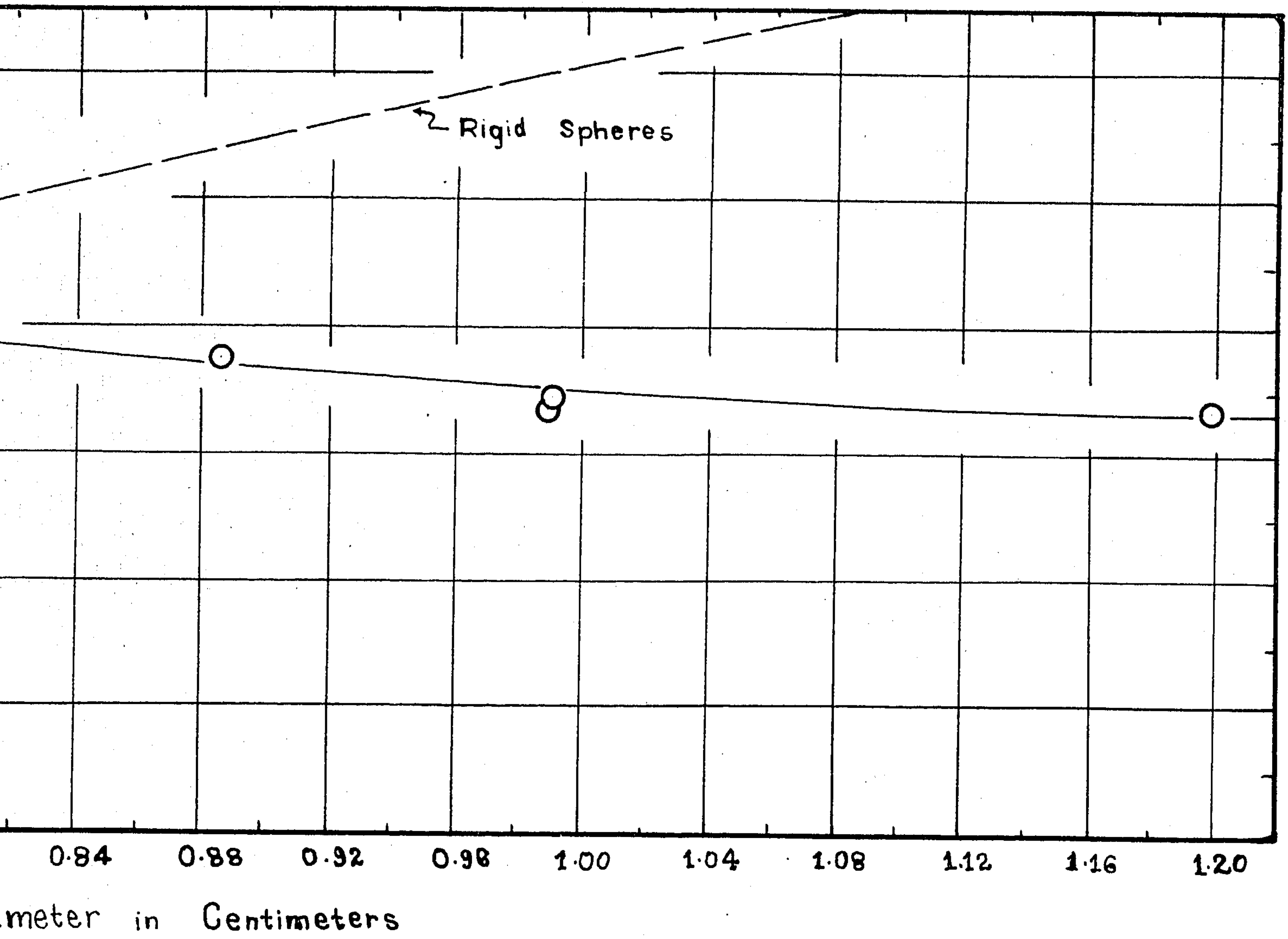


FIG.13 FALL VELOCITIES OF CHLOROBE



CHLOROBENZENE DROPS IN WATER

smallest size drops produced could not be used for taking any data as they drifted greatly and bumped on the walls of the 6-inch column as many as 3 to 4 times in a travel of one meter. (2) The largest size of the drops was limited by the practical difficulties in obtaining a stable drop. There is a maximum limit to the size of a drop that can be stable. Whether that upper limit was reached in the present drop sizes may be questioned, but the present drop sizes attained can be claimed to be in the neighbourhood of the upper size limit for stable drops.

The range of variables studied in the present work may be summarized as follows:

$(\rho_i - \rho_o)$ = 0.1037 to 1.9568 gms/ml.

D = 0.114 to 1.119 cm.

$\frac{\mu_i}{\mu_o}$ = 0.8533 to 10.59

σ = 14.6 to 41.6 $\frac{\text{dynes}}{\text{cm}}$

The results of photographic studies are presented along with discussion of results.

DISCUSSION OF RESULTS AND CORRELATIONS

(a) Discussion of Experimental Results

Let us first consider the system of carbon tetrachloride drops in water. The data are shown in Fig. 8 wherein the data of other workers are also included. Also a dashed line is drawn representing the fall velocities of rigid spheres of the same volume and density as the liquid droplets. This rigid sphere fall velocity line is drawn for all the six systems studied. The following points can be observed in Fig. 8.

1. The data in the present work collected in a 3-inch column seem to fall very near the smooth curve for the 6-inch column. This shows that there is no wall effect in the 3-inch column. The largest size drop measured in a 3-inch column was 0.448 cm. in diameter. Since the 6-inch column is twice the diameter of the 3-inch column, it is reasonable to expect that there will not be any wall effect at least up to a drop diameter of 0.896 cm.

2. The fall velocities of drops are much higher in the present work than in any previous work compared in Fig. 8. The fall velocities in a 3-inch column in the present work are definitely higher than those obtained by Katz in the same size column. The difference can be explained by the fact that Katz used a tygon tube to connect the hypodermic needles to the buret. During preliminary experimental work it was found that tygon lowered the interfacial tension of

carbon tetrachloride to water by 12.8% after contact of 5 minutes. So it can be explained that the lowered interfacial tension thus resulted lowered the fall velocities in Katz's work. Later on it will be shown that a lower interfacial tension causes the drops to fall slower. Though Ailor[3] , Smirnov and Ruban[39] have not clearly stated what tubes they used, their figures of apparatus show some connecting tube on the glass connections. These could be rubber, Tygon, or some other tube and could have lowered the interfacial tension. Also they conducted their experiments in much smaller diameter columns and there could possibly be some wall effect too. Any way it can clearly be stated that the three previous references have not taken any special care about cleanliness of apparatus, avoidance of rubber or plastic tubes, greases etc. Since all these precautions were taken and since the chances of interfacial tension decreasing due to impurities were much smaller, the present data in the 6-inch column were considered to be more accurate. It is of interest to note that the fall velocity curves in all cases including the rigid sphere line seem to converge at smaller drop sizes.

Farmer[12] also obtained data on the fall velocities of carbon tetrachloride drops in water. His data could not be used for direct comparison since the carbon tetrachloride he used contained dissolved acetic acid. The fall velocities noted by Farmer are also lower than those obtained in the present work. Since (1) Farmer used only a 2-inch column,

(2) he took only average velocities and not terminal velocities, (3) he did not take any special precautions to avoid the lowering of interfacial tension and (4) he did not give the interfacial tension data on the carbon tetrachloride containing acetic acid, which he used, it is hard to state whether mass transfer had any part in lowering the fall velocities of the liquid droplets in Farmer's work.

3. The dashed line for rigid spheres indicates the velocity of fall of rigid spheres of the same density and diameter as the drops. Since the rigid spheres have an infinite viscosity and infinite interfacial tension, the difference in the behaviour of the drops may be due to either of the two factors of lowered viscosity and interfacial tension. In the case of smaller drop sizes, the velocities of drops and rigid spheres are very nearly the same. A discerning person can point out that the line of fall velocities of drops does cross over the rigid sphere line, but the difference in this case is not serious and can be left over for discussion at a later stage.

4. As the drop diameter increases, the fall velocity of the drops increases to a maximum and then falls off with further increase in drop diameter, finally approaching an asymptotic value. This general behaviour of fall velocity curves seems to be a characteristic of drop behaviour as it can be seen from the data on other systems also.

Considering next the tetrachloroethane drops in water, one finds that the fall velocity curve is similar to that of carbon tetrachloride. It should be noted that the densities of carbon tetrachloride and tetrachlorethane are very near each other, being 1.5842 and 1.5813 gms/ml., both at 25.0°C. Hence the rigid sphere fall velocity curves are both almost the same. The difference in $(\rho_i - \rho_o)$ is about 0.5% only. But the fall velocity data show that though both of them have approximately the same fall velocities in the smaller drop sizes and are very near to the values for rigid spheres, yet in the larger sizes the tetrachloroethane drops fall more slowly than the carbon tetrachloride drops. This lowering in velocity is much higher than can be expected from the lowering of 0.5% in $(\rho_i - \rho_o)$. Turning to the viscosities of the two liquids, tetrachloroethane has $\left(\frac{\mu_i}{\mu_o}\right) = 1.681$ while carbon tetrachloride has $\left(\frac{\mu_i}{\mu_o}\right) = 1.040$. 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 fall velocity of the tetrachloroethane drops. The interfacial tension of the tetrachloroethane is only 31.3 $\frac{\text{dynes}}{\text{cm.}}$ compared to 41.6 $\frac{\text{dynes}}{\text{cm.}}$ for carbon tetrachloride. It seems appropriate to assign the responsibility for the lowered drop velocity on the lower interfacial tension. It is also consistent, since the rigid spheres with infinite interfacial tension fall faster than both these and the tendency is in the right direction. Hence it was felt that interfacial tension has certainly a hand in controlling the fall velocities of liquid droplets and that an increase in interfacial tension does make the drop fall with velocities approaching that of rigid spheres.

The above can also be explained as follows: A drop with a high interfacial tension is deformed less, while one with a low interfacial tension is deformed more by external forces. If both drops have the same density and same volume, they both tend to fall with the same velocity. As they fall they meet with a resisting force to their motion. This resisting force produces greater deformation in a drop of lower interfacial tension than in the one with higher interfacial tension. As a sphere has the lowest surface area, any deformation increases the surface area of the drop. The greater the deformation, the greater this increase in surface area. As such a greater deformation of the drop increases, the drag on the drop and finally lowers its fall velocity. In this way a lower interfacial tension leads to a lower fall velocity of the drop.

In the smaller size drops, according to equation (36)

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{p_i - p_o}{\sigma}$$

we have $(p_i - p_o)$ becoming larger as the radii of curvature decrease. This will be the result in the case of smaller drops, as such smaller drops are deformed much less from the spherical shape than the bigger drops. Qualitatively, one can argue that $(p_i - p_o)$ is inversely proportional to the diameter of the drop and since the drop deformation is increased by a lowered value of $(p_i - p_o)$, that the deformation of the drop is roughly proportional to the drop diameter. Also since the drag of a drop is proportional to the increase in surface area, one is tempted to feel that the increase in the drag coefficient of drops may in some way be proportional to the square of the drop diameter or at least a function of the drop diameter.

The photographic studies have also shown that the smaller drops are very nearly spherical while the larger drops are deformed appreciably. As such the greater deviation of the fall velocities of the larger size drops from those of rigid spheres is understandable.

The fall velocity curve increases to a maximum as the drop size increases and then falls off asymptotically with a further increase in drop size. It may be mentioned that Conway[10] and Katz[22] found the same type of behaviour with liquid droplets in liquids while Lenard[24] reported a similar behaviour for water drops in air. At this stage we might note that no such maximum point or peak occurs in the case of rigid spheres. In other words, for a given system the terminal velocity of fall is a unique function of the size of the rigid sphere, while it is not so in the case of liquid droplets. This peculiar behaviour of liquid droplets needs some consideration.

In the first instance, the liquid droplets do change their deformation with a change in their size. This explains why the fall velocity of the drops is lower than that of rigid spheres, but does not explain why there should be a peak in the fall velocity curve. In order to understand this phenomena, the drops were carefully observed during their fall and the following observations were made.

Very small size drops deformed less, and were nearly spherical. They did not show any observable vibrations in their shape. As the drop size increased, the drop deformation increased. About the size corresponding to the maximum velocity range on the curve, the drops began to show some oscillations in shape; while a rotation or vibration of the drop as a whole could only be suspected in the case of small size drops, they could be seen very clearly in the case of large size drops. Hence it can be said that a falling drop possesses translational, vibrational and possibly rotational energy and this energy can only be derived by the loss of potential energy. In the case of the small size drops, the translational energy seems to be important,while in the larger size drops the vibrational energy seems to be predominant. As such, in the case of the smaller size drops, the fall velocity increases with an increase of drop diameter. In the case of the large size drops the increased loss of potential energy due to larger sized drop is utilized to supply the vibrational energy of the drop. It is quite possible to have a characteristic vibrational frequency for each system so that the vibrational energy requirements might be proportional to the size of the drop. Since the loss of potential energy is proportional to the product of v $(\rho_i - \rho_o)g$ times $\left(\frac{\text{height fall}}{\text{unit time}}\right)$ it can be seen that the approach of the velocity to a constant velocity is possible. The peak in the velocity curve probably corresponds to the transition stage of translational energy important to vibrational energy becoming prominent.

While observing the drop behaviour, it was also found that the very small drops fall nearly vertically, and as the drop size increases, they begin drifting till at one stage this drifting becomes so much that it may even become impossible to collect the fall velocity data as almost every drop falls on the walls of the container. As the drop size is increased, this drifting decreases and the drops fall much nearer the center line of the column, though they do not keep strictly to any straight line path. There is a feeling that some of the large size drops followed even a spiral path, but it was hard to get it in a photograph. The maximum drifting of the drops as noted above may correspond to the natural frequency of the drop coinciding with the frequency of eddies formed in the wake of the drop,as was referred to by Gunn[16] for the case of water drops in air. The presence of these eddies known as Karmon vortex trail was also observed in a qualitative experiment conducted as follows:

The 3-inch diameter column was filled with distilled water containing 2% $\frac{w}{v}$ of potassium iodide and 0.036% $\frac{w}{v}$ of soluble starch. Drops of carbon tetrachloride containing 2% $\frac{w}{v}$ of iodine were allowed to fall through the column. The iodine in the drop reacted with the starch iodide in the continuous medium and dark rings began peeling off from behind the drop as the drop fell through. These dark rings peeling off from behind the drop clearly show the presence of Karman vortex trail in the wake of the drop.

In view of the peeling off of the vortices from the body, vibrations are likely to be set up in the falling body. In the case of a symmetric body like a rigid sphere, these vibrations may be difficult to observe. As was already mentioned, Prandtl[34] refers to these vibrations in discs in the range of Re = 80 to 3000. So the vibrations in the drops are also in consonance with the behaviour of solids. In the smaller sizes, the drop might be vibrating as a whole while in the larger sizes these vibrations are introducing changes in the drop shape also in addition to the vibrations of the drop as a whole.

The larger size drops could clearly be seen to be tilting or nutating while it became difficult to decide whether the drop was actually rotating or vibrating. In the case of the small size drops, the rotation of the drops was suspected while it became difficult to decide about it in view of the fact that the drop consists of a clear and homogeneous liquid. Even photographs did not help clarify the point on account of the same difficulty. Blanchard's[6] reference to rotation of water drops in air could not be checked clearly for the case of liquid droplets in liquids.

In the smaller ranges of the drop sizes, it may be found that the fall velocities of liquid droplets are greater than those of rigid spheres over a certain range of drop sizes. This criss-crossing of the fall velocity curves needs to be explained. This difference in fall velocities in this particular range really is significant in the case of ethyl chloroacetate drops and chlorobenzene drops in water, though it was not so significant in the case of the other systems. The possible explanation for this behaviour may be:

(1) Goldstein[15] shows that for rotating spheres, the drag coefficient decreases first and then increases as the ratio of equatorial speed to wind speed is increased. In a similar way, if the drops were rotating, during certain size and rotational speed range, their drag will be smaller than that of spheres. Consequently they can fall with a higher velocity than the rigid spheres.

(2) It is possible that the drop in this region may be more or less streamlined and thus having a lesser drag coefficient and consequently falling faster than the spheres.

But the photographic evidence does not lend much support to this idea and in some instances even seems to run counter to this postulate of streamlined shape. Hence, this is not likely to be correct. The question of spinning remains still unverified by direct evidence.

It is also interesting to note that this behaviour of the drag of drops being lower than that of spheres occurs prominently in the case of the two systems which are characterized by a combination of small values for $(\rho_1 - \rho_0)$ and $(\frac{\mu_1}{\mu_0})$. This behaviour probably became prominent because the drops with material having small values of $(\rho_1 - \rho_0)$ and $(\frac{\mu_1}{\mu_0})$ could be more easily influenced by the behaviour of the vortex trail.

Since our knowledge of the variation of drag coefficients with vibrations and rotations of bodies is limited, it looks as if further discussion in this direction may be only in the type of conjectures. Hence let us consider the drop behaviour as revealed by the photographic studies and then proceed to find out some correlations, if possible.

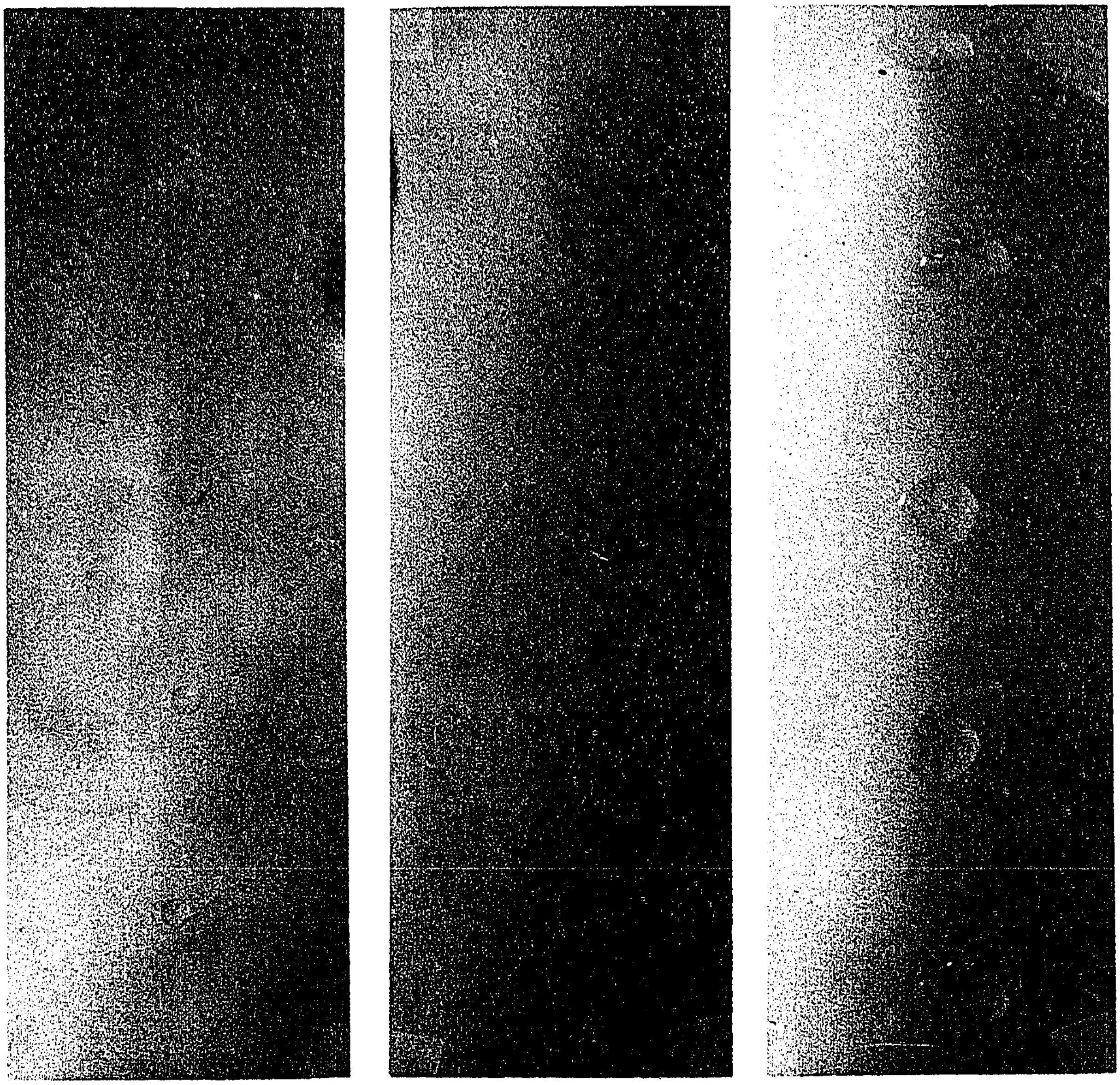
(b) Discussion of Photographic Results

The results of the photographic studies are given in Figs. 14 to 18. In Fig. 14 are shown drops of carbon tetrachloride containing dichlorofluorescein in solution falling through water. Figs. 15 and 16 show o-nitrotoluene and ethyl chloroacetate drops respectively falling in water. Fig. 17 shows drops of ethyl chloroacetate containing dichlorofluorescein falling through water. Fig. 18 shows drops of chlorobenzene containing dichlorofluorescein falling through water. All the pictures were taken on drops after they fell through a height of about 110 cms.

The first impression on looking at these pictures will be that the smaller drops are less deformed than the larger drops, and are sometimes nearly spherical. A close study of each of the systems will show the following features.

The carbon tetrachloride + dichlorofluorescein drops show distortion of shape even in the smaller drop sizes of D = 0.241 cm. The deformation of the drop is greater for drops of 0.384 cm. diameter. For the bigger drops of 0.563 cm. diameter, the violent changes occurring in the shape of the drop can be clearly shown. The drops do not show any equilibrium shape. Indeed they do not seem even to approach any regular geometric shape. They seem to defy any mathematical treatment.

For purposes of comparison Magono's[27] water drop in air and Blanchard's[6] multiple image of a water drop in air are



(a) (b) (c)

Fig. 14. Carbon Tetrachloride + Dichloroflourescein Drops in Water.

(a) D = 0.241 cm., V = 18.9 cm/sec.
(b) D = 0.384 cm., V = 20.9 cm/sec.
(c) D = 0.563 cm., V = 18.6 cm/sec.

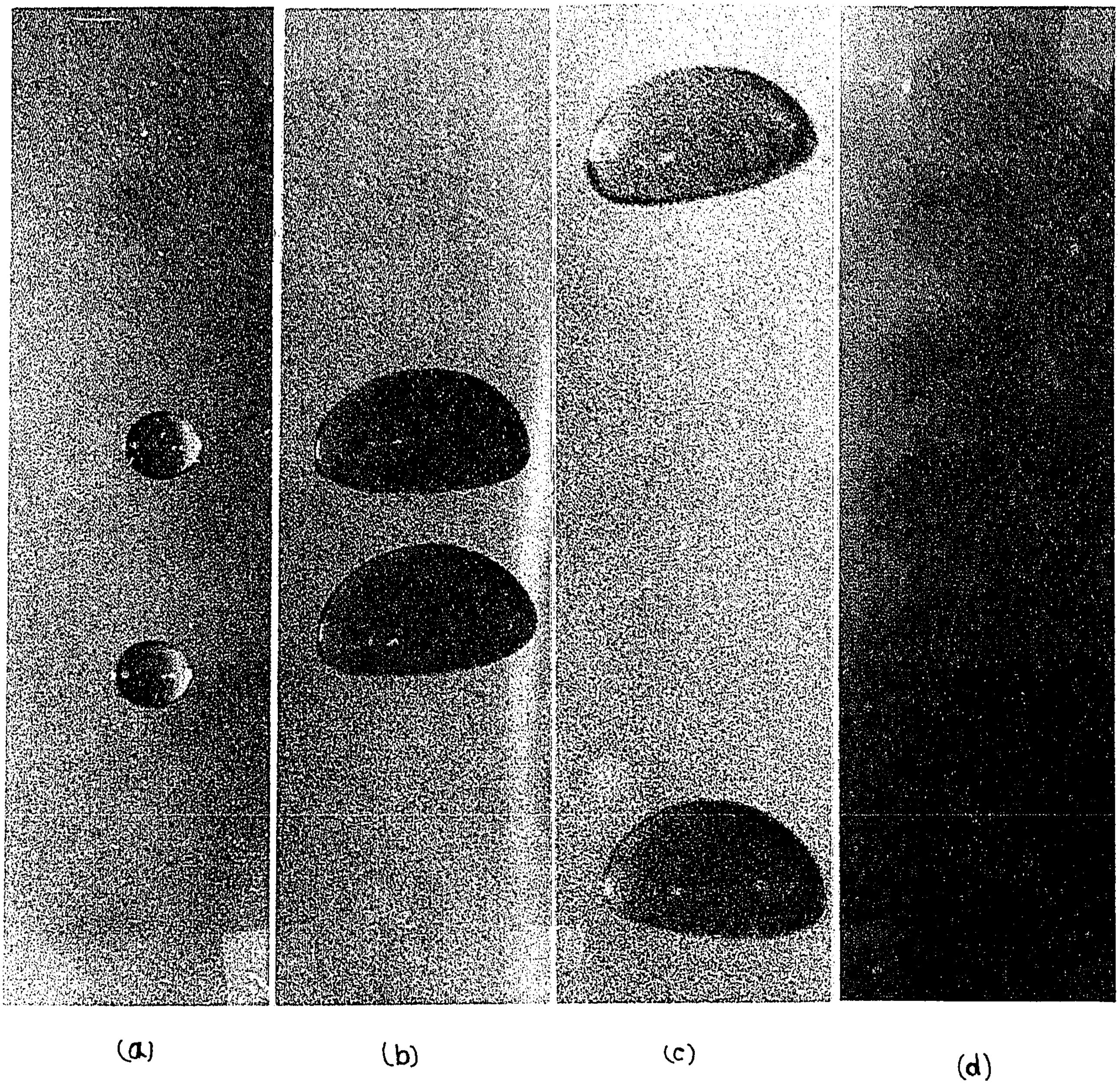
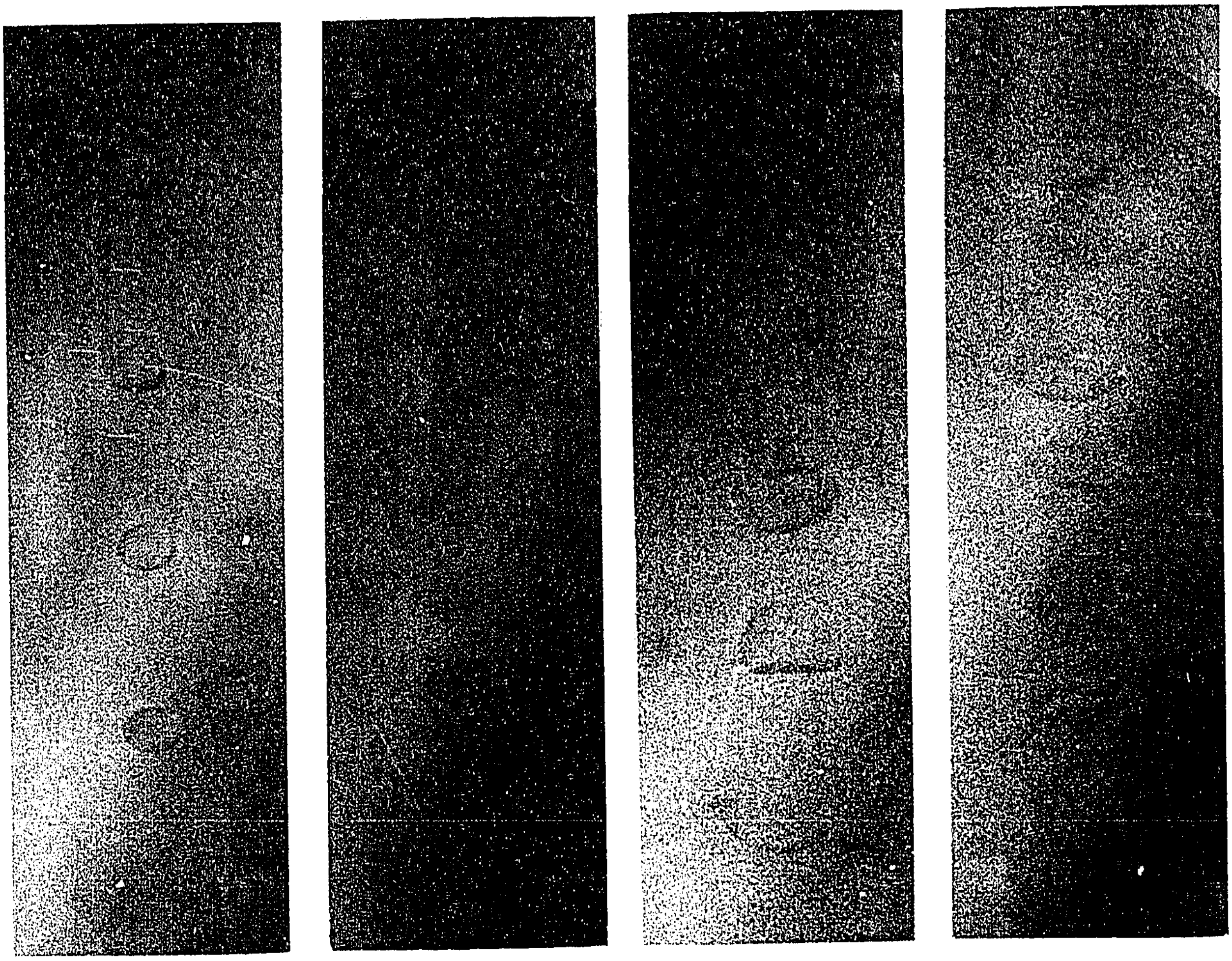


Fig. 15. O-Nitrotoluene Drops in Water.

(a) D = 0.372 cm., V = 11.1 cm/sec.
(b) D = 0.758 cm., V = 13.1 cm/sec.
(c) D = 0.758 cm., V = 13.1 cm/sec.
(d) D = 0.758 cm., V = 13.1 cm/sec.



(a) (b) (c) (d)

Fig. 16. Ethyl Chloroacetate Drops in Water.

(a) D = 0.399 cm., V = 12.3 cm/sec.
(b) D = 0.399 cm., V = 12.3 cm/sec.
(c) D = 0.633 cm., V = 11.1 cm/sec.
(d) D = 0.633 cm., V = 11.1 cm/sec.

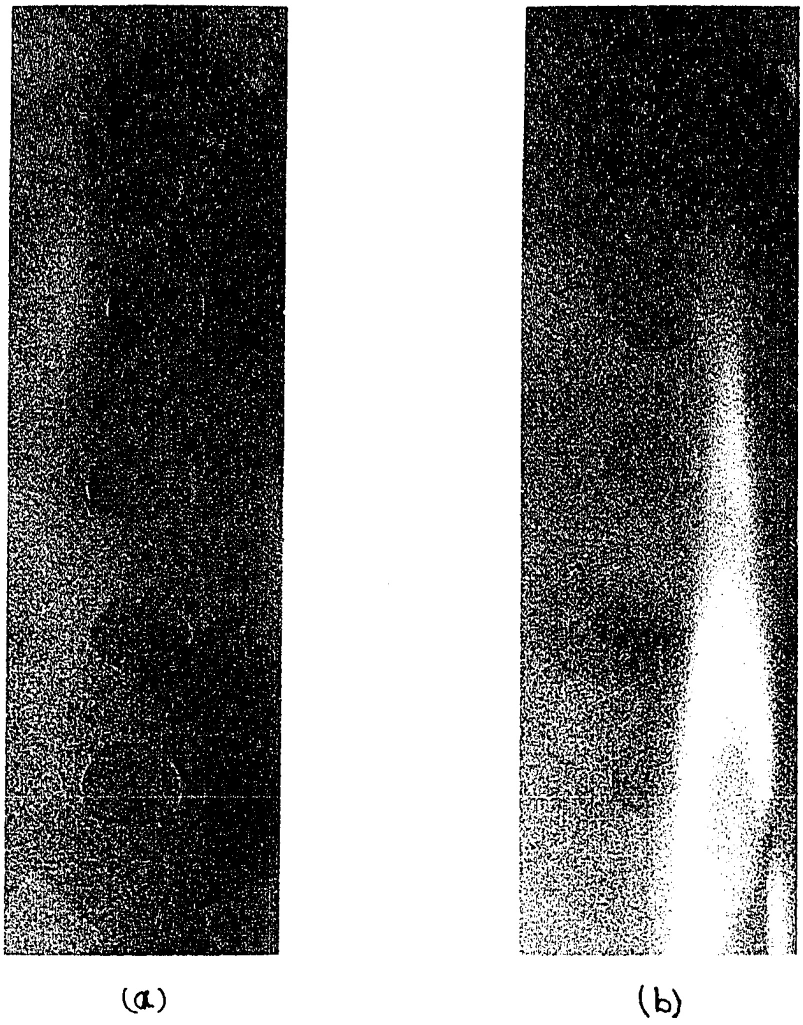
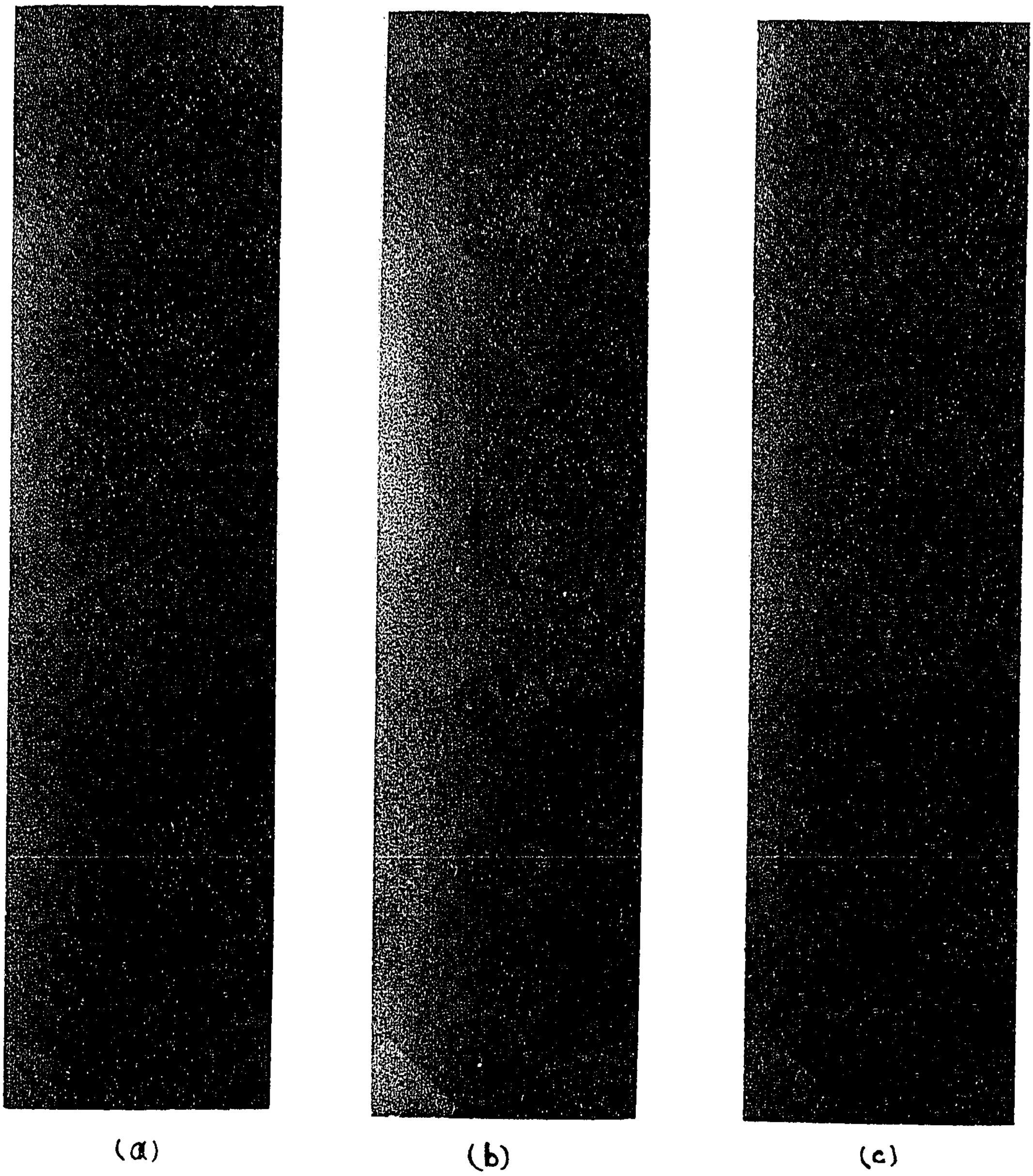


Fig. 17. Ethyl Chloroacetate + Dichlorofluoroscein Drops in Water.

(a) D = 0.638 cm.
(b) D = 0.638 cm.



Figl 18. Chlorobenzene + Dichlorofluoroscein Drops in Water.

(a) D = 0.622 cm., V = 9.5 cm/sec.
(b) D = 0.650 cm., V = 12.1 cm/sec.
(c) D = 0.899 cm., .

reproduced in Figs. 19 and 20. It can easily be seen that the shape of the water drop in Magono's photograph is much similar to that of the large sized drops of O-nitrotoluene. The vibration observed in the present work on carbon tetrachloride + dichlorofluorescein drops in water seem to be much more violent than those observed by Blanchard on water drops in air.

In Fig. 15 for o-nitrotoluene drops slight deformation can be observed even in the case of the drops of 0.372 cm. diameter, while the drops of 0.758 cm. diameter are deformed very much. The drops also seem to be tilting sideways. The vibrations in drop shape can be clearly seen in Fig. 15 (d), though they are not as bad as in Fig. 14.

In the case of ethyl chloroacetate drops, both deformation and vibrations can be clearly seen even in the case of drop diameters of 0.399 cm. and 0.633 cm. Figs. 16(c) and (d) for 0.633 cm. drops clearly show that the distance between the consecutive images are not the same. Of course, this feature is to be expected when the drops are changing their shape continuously. After having observed this feature in this case, an attempt was made to compare the distance between the consecutive drops in other systems also. This type of non-uniform velocity over small intervals of time is to be found in the case of the previous two cases of carbon tetrachloride + dichlorofluorescein as well as o-nitrotoluene drops where multiple images of same drop were taken.

In the case of 0.638 cm. drops of ethyl chloroacetate + dichlorofluorescein, both vibrations as well as unequal

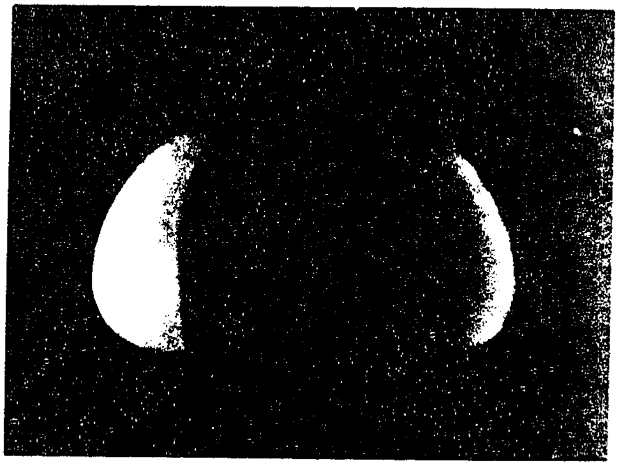


Fig. 19. Water Drop Falling in Air

Photograph by Magono[27]

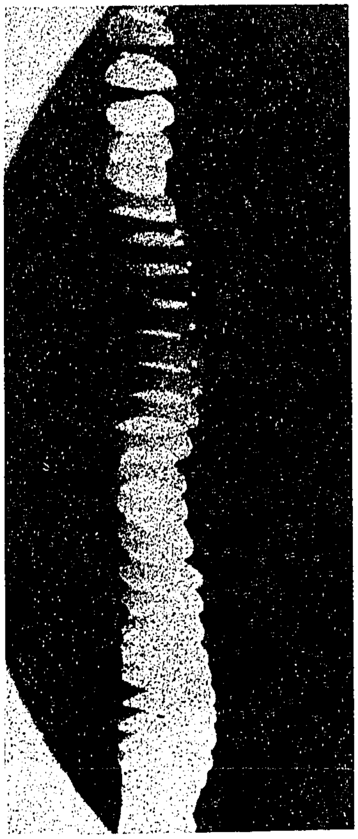


Fig. 20. Stroboscopic Picture of a Water Drop.

Photograph by Blanchard.[6]

velocities of fall can be clearly seen. Though both Figs. 17(a) and (b) were taken under identical conditions, the shapes of the two different drops are clearly different. This phenomenon of different shapes for same size drops under identical conditions of experiment can be observed in the case of Figs. 16(a) and (b) as well as Figs. 16(c) and (d).

In the case of chlorobenzene + dichlorofluorescein drops, the drops with a diameter of 0.422 cm. are very nearly spherical and have no vibrations in their shape. Drops with a diameter of 0.650 cm. are deformed, but do not show any changes in shape. These two sizes show a uniform fall velocity in the small intervals of time. Drops with a diameter of 0.899 cm. are deformed and they also show changes of shape. The distances of fall between consecutive drops are not uniform. A careful review of the cases of non-uniform velocity of drops over small intervals of time indicates that this phenomenon occurred only in those cases where there were changes in shape of the drop going on.

In view of the fact that dichlorofluorescein was added to some of the drops and also because the data are not comprehensive, it is not possible to develop quantitative relationships from these photographs. Yet one can qualitatively say that in the range of drop velocities less than the maximum, the drops seem to retain approximately an equilibrium shape, while in the range beyond the maximum fall velocity, the drops change their shape due to vibrations. The range of maximum fall velocities seems to correspond to the transition from a stage having equilibrium shape to one without any equilibrium shape.

(c) Correlations

The values of fall velocities were taken from the smooth curves in Figs. 8 to 13 for purposes of this section. The drag coefficients and different dimensionless groups used in this section are presented in Tables A-9 to A-14.

The drag coefficients of liquid drops were first plotted against the Reynolds number and are shown in Fig. 21. For purposes of comparison the drag coefficients of rigid spheres are also shown in the Figure with a broken line. It can be seen that the drag coefficients of liquid droplets reach a minimum and then climb up steeply with increasing Reynolds numbers. At a first glance there does not seem to be any parameter connecting them. An attempt was made to see if the C_D vs. Re plot cannot be redrawn with Weber number as a parameter, but it did not prove successful.

The next attempt was to check the correlations proposed by Hughes and Gilliland.[20] As was already mentioned, they have presented curves of terminal velocity group T_v versus Wt. In Fig. 22 are plotted T_v vs. Wt values of carbon tetrachloride and o-nitrotoluene drops in water. Their Sd values are 2,445 and 2,408 respectively. Fig. 23 shows the plot of T_v vs. Wt values for s-tetrabromoethane and ethyl chloroacetate drops which have Sd values of 1,424 and 1,359 respectively. Fig. 24 shows a similar plot for tetrachloroethane and chlorobenzene drops in water. These two have Sd values

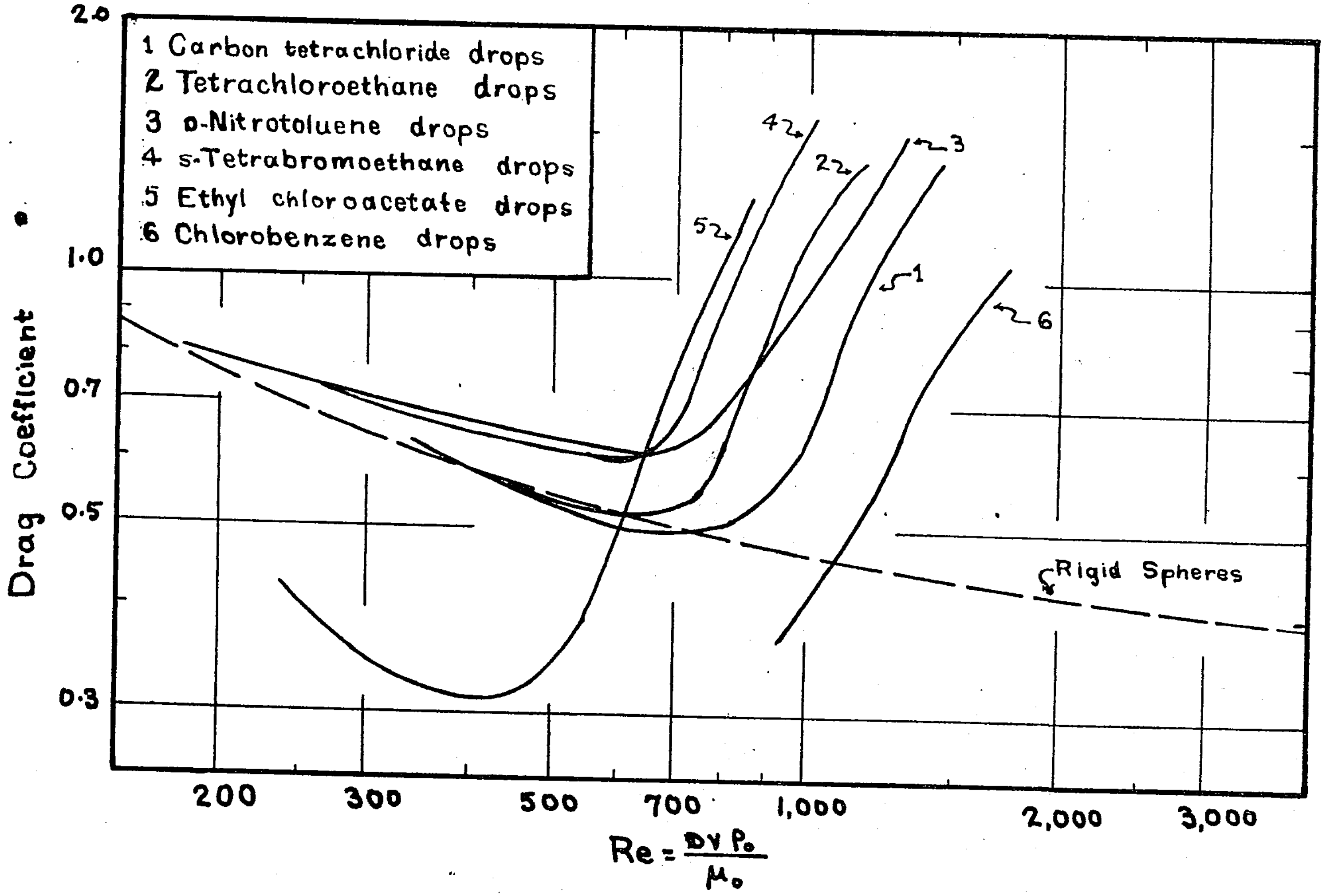


FIG. 21 DRAG COEFFICIENTS OF LIQUID DROPLETS

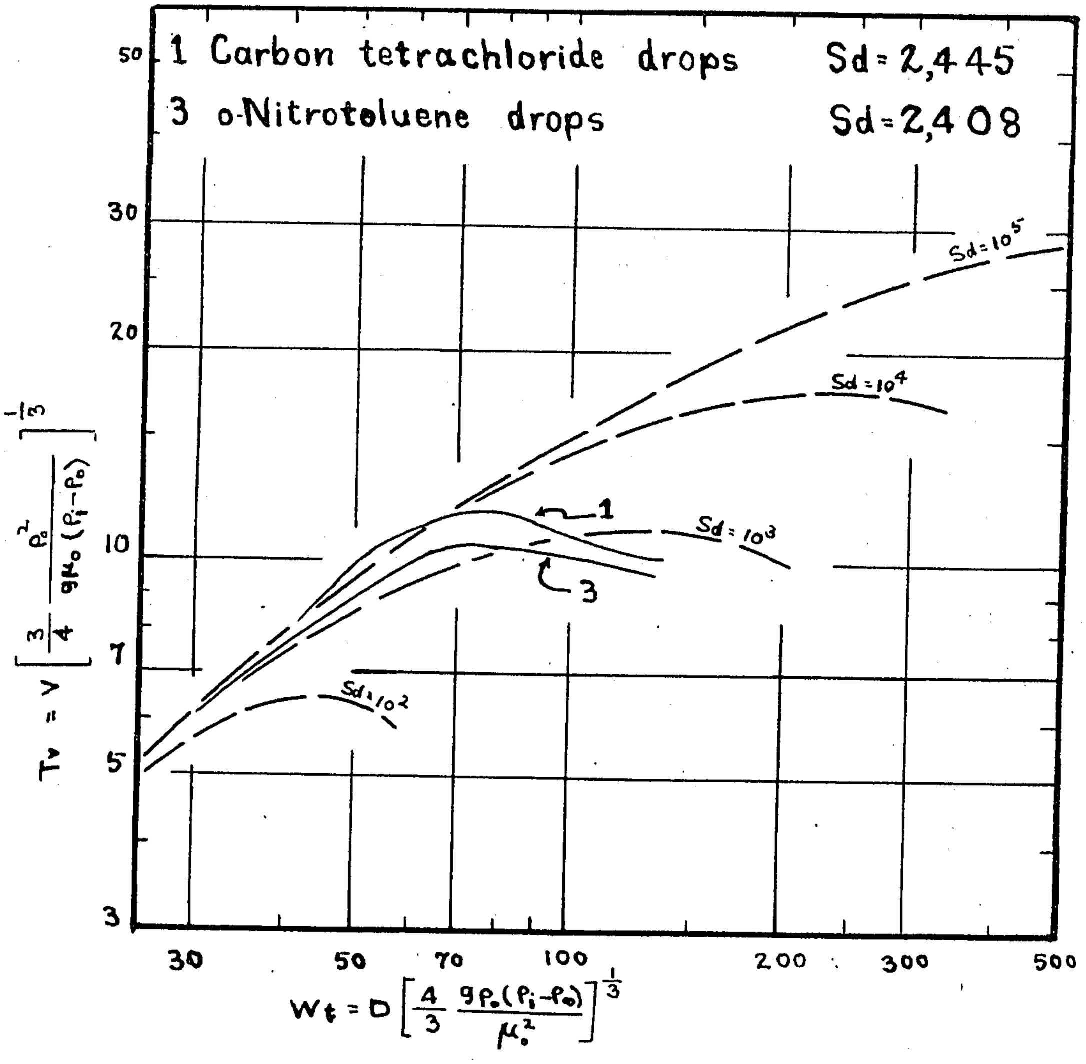


FIG. 22 CHECK OF HUGHES & GILLILAND'S CORRELATION

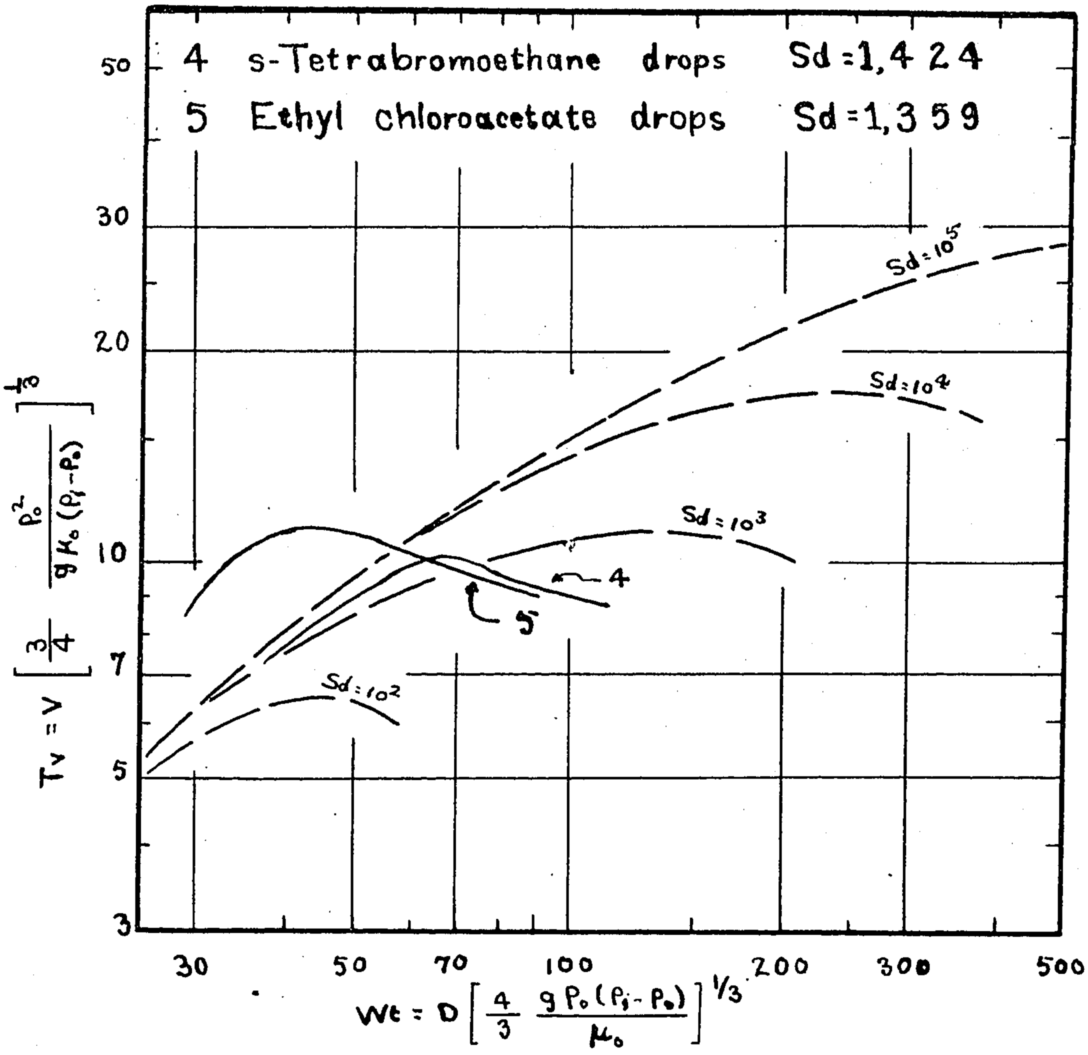


FIG. 23 CHECK OF HUGHES & GILLILAND'S CORRELATION

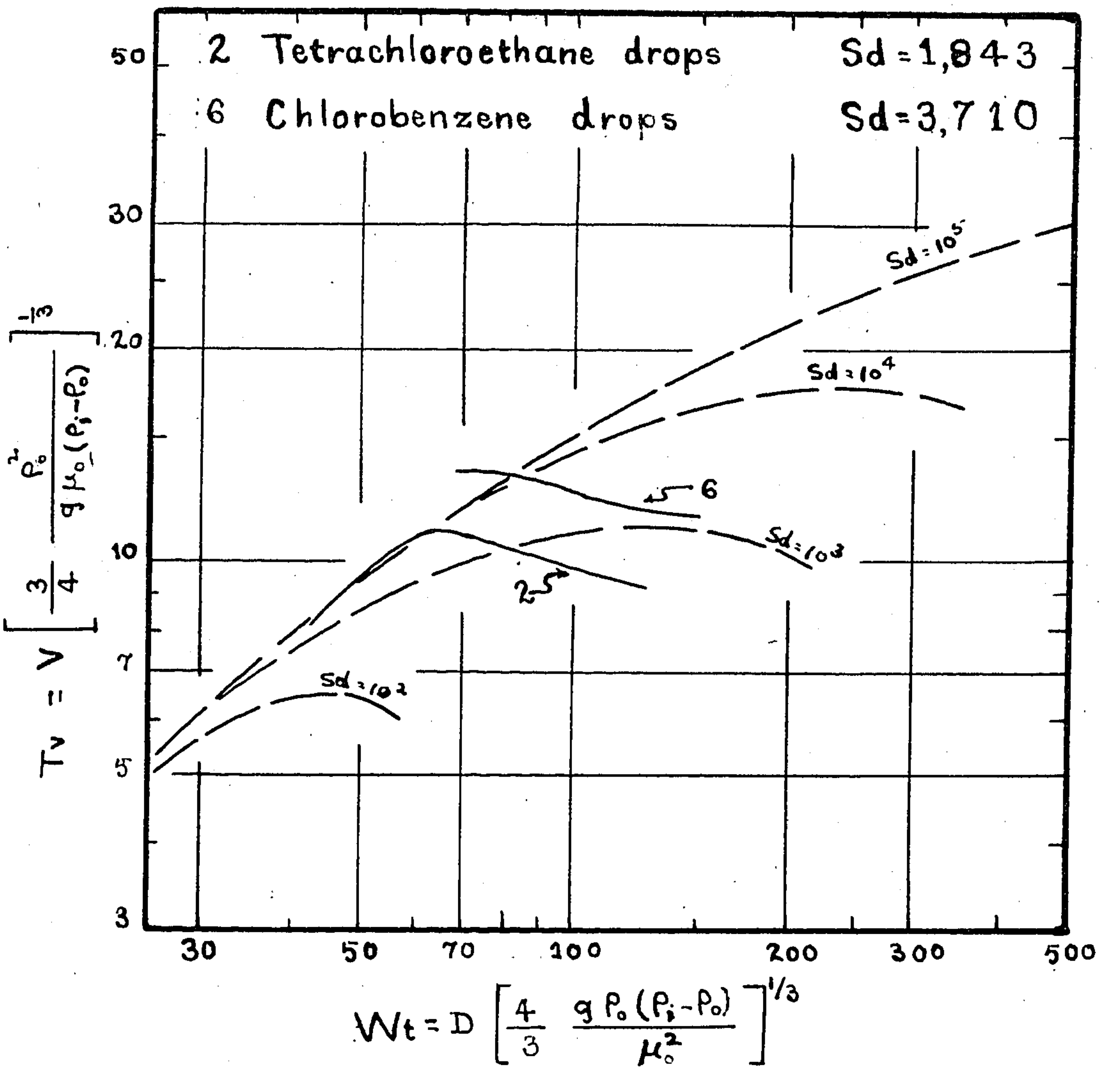


FIG.24 CHECK OF HUGHES & GILLILAND'S CORRELATION

of 1,842 and 3,710 respectively. In all the three Figs. 22 to 24, the curves of Hughes and Gilliland are shown in dotted lines for purposes of comparison. It can easily be seen that Hughes and Gilliland's correlations are not applicable to the case of liquid droplets falling in liquids.

In view of the above, an independent method of approach to the problem appeared to be necessary. For this purpose the assistance of dimensional analysis was taken. The complicated nature of drop behaviour as revealed in the photographic studies also indicates that there probably will not be a simple solution to this problem. Hence it was decided to find a correlation in terms of dimensionless groups. In view of the fact the fall velocity of the liquid droplets passes through a maximum and then falls off asymptotically, it appears as though there are two mechanisms controlling this process of fall of droplets. It has already been pointed out that the vibrations in drop shape are predominant in the larger drops where the fall velocities of drops decrease asymptotically. In view of the above, the following two correlations were attempted:

The Weber numbers of falling droplets are plotted against Reynolds numbers in Fig. 25. It can be seen that each of the curves shows a linear relationship on a log-log plot up to a certain value of Reynolds number and then breaks off from the linear relationship. Also, it is interesting to note that all the straight lines are very nearly parallel.

Hence in the range of smaller values Weber number one can write

$$We = H\,(Re)^{J} \qquad (50)$$

where H and J are arbitrary constants.

Equation (50) is obviously a special case of equation (34) developed under dimensional analysis.

In Fig. 26, the ratio of drag of a droplet to that of a rigid sphere of same volume and density properties and falling with the same velocity as the droplet is plotted against the dimensionless β group. In this higher values of β represent larger drop sizes. In the larger drop sizes, it can be seen that the curves are nearly linear on a rectangular coordinate plot. This shows that each of the curves follows approximately the straight line equation. The dotted line represents the mean of all the six curves.

Let us consider equation (32)

$$C_D = K\,\beta^{k}\,Re^{l}\,\mu_r^{j} \times 4 \qquad (32)$$

We can assume

$$C_D^{s} = K_1\,Re^{l_1} \qquad (51)$$

where K_1 and l_1 are constants.

Then

$$\left(\frac{C_D}{C_D^{s}}\right)_{Re} = \frac{4K}{K_1}\,\beta^{k}\,(Re)^{l - l_1}\,\mu_r^{j} \qquad (52)$$

The present relationship is

$$\left(\frac{C_D}{C_D^{s}}\right)_{Re} = K_2\,\beta + Q \qquad (53)$$

wherein K_2 and Q are both constants.

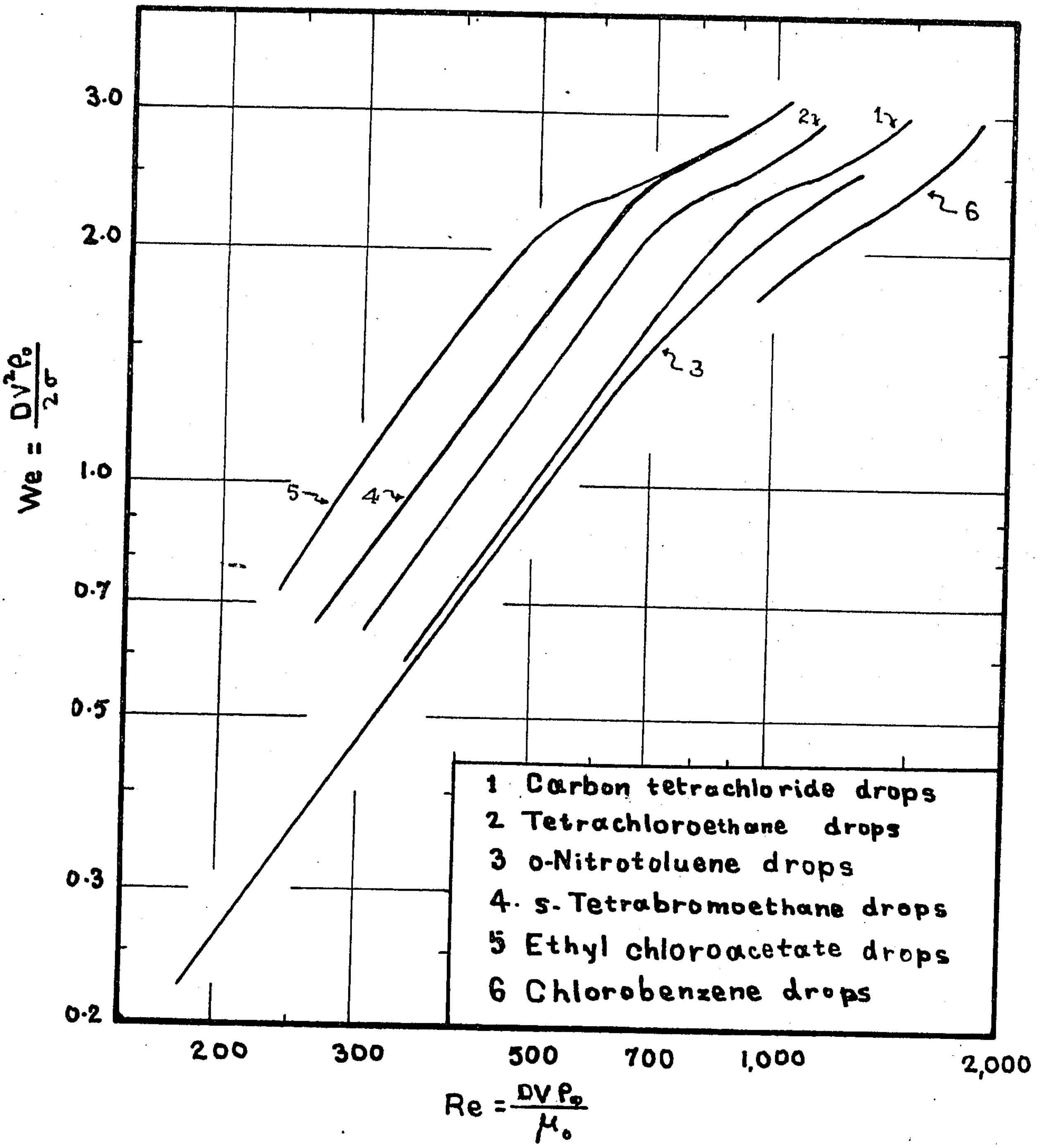


FIG. 25 CORRELATION OF WEBER NUMBER WITH REYNOLDS NUMBER

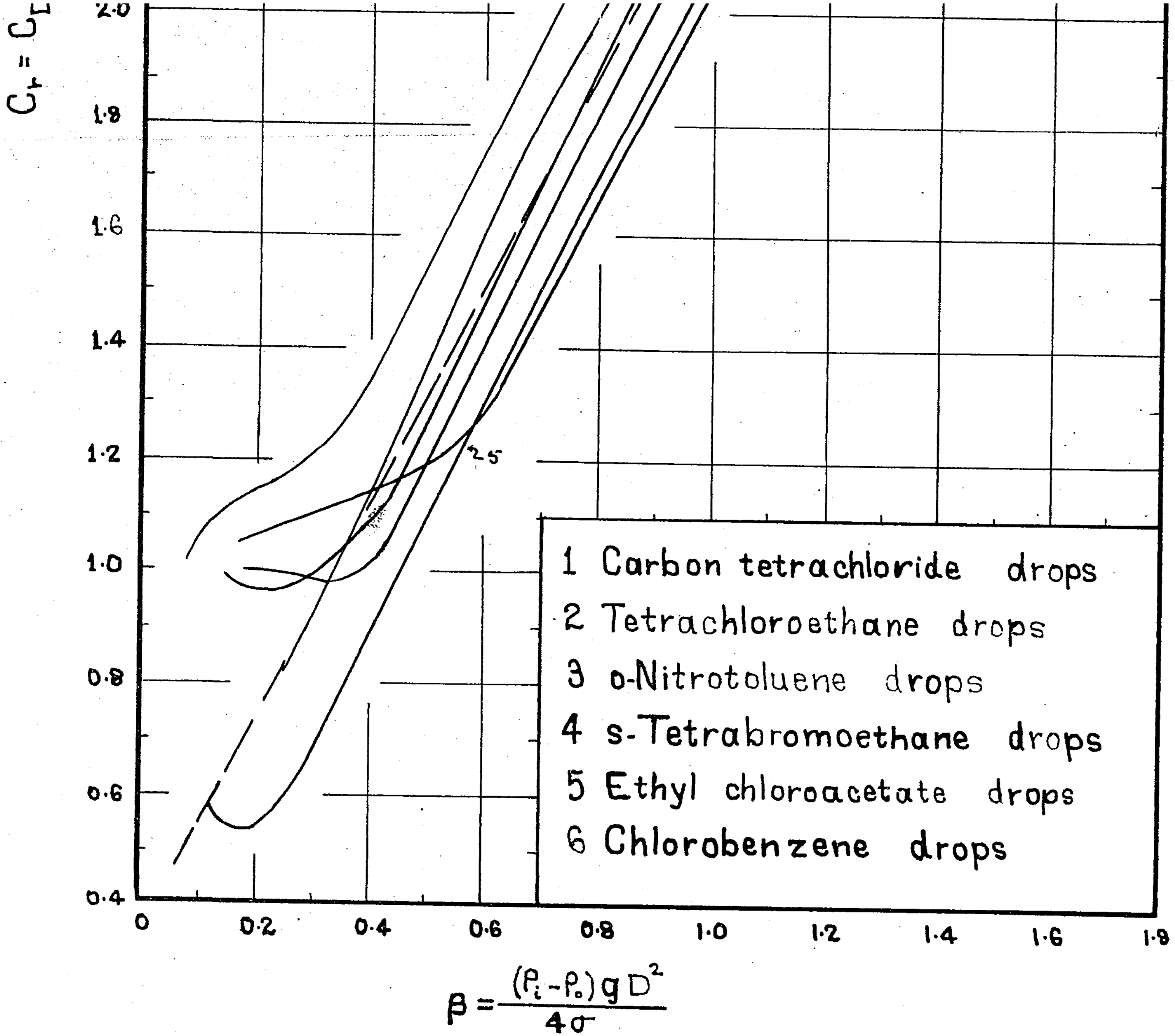
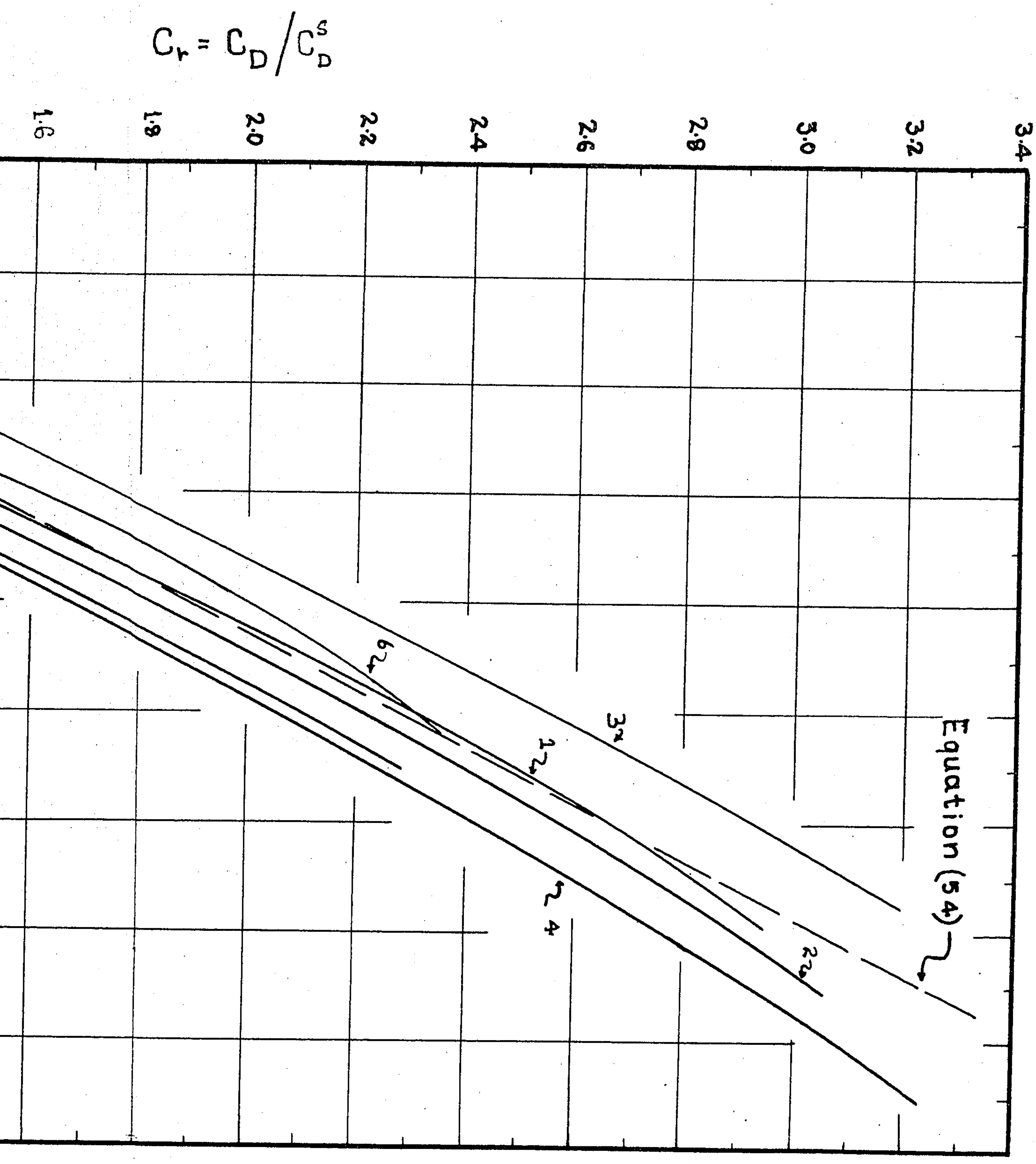


FIG. 26 CORRELATION OF C_r WITH β GROUP


$C_r = C_D / C_D^s$
1.6
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4
6
3
1
4
2
Equation (54)

Obviously equation (53) is a special case of equation (51). Also it appears as though

$$l \approx l_1$$

In both the correlations presented herein, the nature of μ_r^j could not be decided. It may need further refinement and accuracy of work as well as a more extensive range of experimental conditions to determine exactly the role played by the viscosity of the discontinuous phase. Circulations inside liquid droplets are known to exist and Spells[40] has shown them photographically. So the viscosity of the drop material must have something to do with the drag of the drop though it is not clear from the present work how exactly it enters the picture in the range of Reynolds numbers under consideration.

Going back to Figures 25 and 26, it is to be seen that the straight line relationship in Fig. 26 begins at about the same point where the straight line relationship ends in Fig. 25. Thus Figs. 25 and 26 used in conjunction clearly show that there are two separate mechanisms controlling the rate of fall of liquid droplets in liquids. The first mechanism holds up to We = 1.5 to 2.0, and becomes less important. The other mechanism is from $\beta = 0.2$ to 0.4 and is controlling afterwards. Putting it in other words, the relationship

$$We = H\,(Re)^J \qquad (50)$$

fails at a point slightly before the drop attains the maximum velocity. At this point the relationship

$$\left(\frac{C_D}{C_D^s}\right)_{Re} = K_2\beta + Q \qquad (53)$$

begins to become important and is valid in the range afterwards. Thus the maximum velocity point in the fall velocity curves is a point of transition from one mechanism to the other.

Looking more carefully at Figure 25, it can be seen that the curves are so arranged that for any given Reynolds number the Weber numbers are increasing as the value of the Sd group decreases. The curves for carbon tetrachloride and orthonitrotoluene are here inverted but the actual deviation is small. It may be possible to find a suitable parameter if more extensive data were collected.

In Fig. 26 no such general pattern for the deviations of the different curves could be found. Hence a dashed line indicating an average for all the curves was drawn. The average line can be represented by:

$$C_r = \left(\frac{C_D}{C_D^s}\right)_{Re} = 1.87\beta + 0.425 \qquad (54)$$

This equation may be used with reasonable confidence for the case of liquid drops in water for the range of

$\beta \geqq 0.4$.

It should, however, be noted that the continuous medium in the case of all the six systems was water. It will be of great interest to study the behaviour of liquid droplets in other liquids. Preliminary comparisons with the data for water drops in air and of air bubbles in liquids show similar behaviour for the drops and bubbles as is found in the present work. However, as direct comparison of the data could not be made, they were not included in the present discussion and correlations. Further work in this direction may prove fruitful.

CONCLUSIONS

1. Liquid drops falling in liquids are influenced by the interfacial tension property in addition to the four properties of diameter and density of particle as well as density and viscosity of the continuous medium which are important in the case of rigid spheres. A lower interfacial tension value lowers the fall velocity of the drop.

2. For any given system, the liquid drops falling in another liquid are nearly spherical in the smaller sizes and their deformation increases with an increase in the size of the drop.

3. Large size drops show vibrations in their shape and apparently do not have any equilibrium shape.

4. The drops though deformed, if they do not show vibrations in shape (i.e. if they have an equilibrium shape) fall with a uniform terminal velocity. Drops which show vibrations in shape do not fall with a uniform velocity if the time interval for the measurement of velocity is made small (1/10 sec. to 1/20 sec. in the present work). However, they fall with a uniform terminal velocity if a large interval of time is considered.

5. For any given system, as the drop diameter is increased, the fall velocity of the drop increases first to a maximum and then falls off asymptotically.

6. The above phenomenon has been explained by the assumption of two separate mechanisms taking part, one controlling in the range of increasing fall velocities, and the other in the range of decrease of fall velocities.

The maximum velocities of fall are attained during the transition from one to the other.

(a) The range of increasing drop velocities is associated with the range where the drop has an equilibrium shape though deformed. The range of decreasing drop velocities is associated with the region where the drop does not show any equilibrium shape.

(b) The drop behaviour in the region of increasing fall velocities can be represented by an equation of the type:

$$We = H\ (Re)^{J} \qquad (50)$$

This equation fails in the range of We 1.5-2.0. For $We \geqq 2.0$, the drop behaviour can be represented by an equation of the type:

$$\left(\frac{C_D}{C_D^s}\right)_{Re} = K_2 \qquad + Q \qquad (53)$$

Above $\beta = 0.4$, the behaviour of all liquid droplets insoluble in water and falling through water can be represented by

$$\left(\frac{C_D}{C_D^s}\right)_{Re} = 1.87\,\beta + 0.425 \qquad (54)$$

7. The correlations of Hughes and Gilliland[20] are not applicable to the case of liquid droplets falling in liquids.

SUGGESTIONS FOR FURTHER WORK

The present work can be used as a basis to start further work in the study of two phase flow problems especially when deformable bodies are moving through fluids. This work can properly be called only a beginning in the direction. During the course of the work the procedure for obtaining reproducible and reliable data on the fall velocities of liquid droplets in liquids has been carefully worked out and standardized. Hence further work in this direction should be easier to that extent. Also the new method of approach to the analysis of drop behaviour and the correlations obtained may be considered as a beginning and not an end. Hence the following suggestions are offered for further work in this direction.

1. The correlations developed in the course of the present work offer scope for further improvement. They have been worked only for the case of water as a continuous medium. It will be advisable to use other liquids as continuous media and see if further generalizations may not be obtained on similar lines on the present equations.

2. When more data are available over wider range of conditions, an attempt should be made to obtain a general correlation for the three cases of liquids in gases, liquids in liquids, and gases in liquids. At that stage it may be possible to have the case of motion of rigid spheres as a special case of the general two phase flow problem. Experiments should be designed to provide data over widely varying conditions.

3. The present work could not lead to any positive conclusions regarding the influence of viscosity of the drop material. An experiment conducted on the same lines as here but using two isomeric liquids with different melting points may prove useful for the purpose. The two isomers may be so chosen that they have nearly the same density as well as interfacial tension, but a different viscosity.

Now that a method has been worked out, it should be possible to improve the accuracy of the results by further improvements in the apparatus. But there is a limit to the improvement of this accuracy. Any such improvement should make the influence of viscosity of the drop much clearer. Obviously the viscosity of the drop seems to enter as a second order correction only.

4. Further photographic studies of the behaviour of falling drops of liquids should prove very helpful in throwing more light both on drop behaviour as well as its motion.

5. In order that this work can be made useful for the liquid liquid extraction in single drop extraction towers, experimental work should be undertaken on the rate of fall of liquid droplets with mass transfer between the phases.

6. For purposes of application in actual practice as in spray towers, much more work will have to be done and probably statistical methods of approach adopted along with the experimental work.

LITERATURE CITED

1. Adam, N.K., The Physics and Chemistry of Surfaces, 3rd ed., p. 8, Oxford University Press, London (1941).
2. Ibid., p. 365.
3. Ailor, R.M., Velocities of Liquid Droplets in Liquids, B.S. Thesis in Ch.E., University of Tennessee (1950).
4. Allawala, M.A., Drag Coefficients of Gas Bubbles Moving through Liquids, M.S. Thesis in Ch.E., Illinois Institute of Technology (1952).
5. Bashforth, F., and Adams, J.C., An Attempt to Test the Theories of Capillary Action by Comparing the Theoretical And Measured Forms of Drops of Fluid, Cambridge University Press, Cambridge, U.K. (1883).
6. Blanchard, D.C., Trans. Amer. Geophys. Union, 31, 836 (1950).
7. Bond, W.N., Phil. Mag. Ser. 7, 4, 889 (1927).
8. Bond, W.N., and Newton, D.A., Phil. Mag. Ser. 7, 5, 794 (1928).
9. Brown, G.G., et. al., Unit Operations, Chap. 7, John Wiley & Sons, New York, N.Y. (1950).
10. Conway, J.B., Liquid-liquid Extraction: The Mechanism of Solute Transfer in Spray Towers, Ph.D. Thesis in Ch.E., University of Cincinnati (1949).
11. Edgerton, E.K., and Killian, J.R., Flash: Seeing the Unseen by Ultra High Speed Photography, Hale Publishing Co. (1939).
12. Farmer, W.S., Controlling Variables in Liquid-liquid Extraction from Single Drops, M.S. Thesis in Ch.E., University of Tennessee (1949).
13. Flower, W.D., Proc. Phys. Soc. (London), 40, 167 (1928).
14. Gaudin, A.M., Principles of Mineral Dressing, McGrawhill Book Co., New York, N.Y. (1939).
15. Goldstein, S., Modern Developments in Fluid Dynamics, Vol.II, p. 492, Oxford University Press, London (1938). Also p. 504.
16. Gunn, R., Jour. Geophys. Res. 54, 383 (1949).
17. Gunn, R., and Kinzer, G.D., Jour. Meteorol., 6, 243 (1949).
18. Hadamard, J., Compt. rend., 152, 1735 (1911).

19. Henrickson, F., Jr., S.M. Thesis in Ch.E. Pract., Massachusetts Institute of Technology (1941).

20. Hughes, R.R., and Gilliland, E.R., Chem. Eng. Progress, 48, 497 (1952).

21. Imai, I., Jour. Meteorol. Soc. Japan, Series 2, 28 (4), 113 (April 1950).

22. Katz, H.M., Studies of Falling Velocities of Liquid Droplets, M.S. Thesis in Ch.E., University of Cincinnati (1950).

23. Laws, J.O., Trans. Amer. Geophys. Union, 22, part III, 709 (1941).

24. Lenard, P., Meteorol. Zeitschr., 21, 248 (1904).

25. Liznar, J., Meteorol. Zeitschr., 31, 339 (1914).

26. McDonald, J.E., Theoretical Cloud Physics Studies (Final Report), Office of Naval Research, U.S. Navy Dept., Project NR082 093.

27. Magone, C., Photograph of Water Drop Given by McDonald (See Ref. 26) on p. 72.

28. Meinzer, O.E., Hydrology (Physics of the Earth IX), P. 40, McGrawhill Book Co., New York, N.Y. (1942).

29. Newton, Sir Isaac, Mathematical Principles of Natural Philosophy, translated by Motte, A., in 1729, University of California Press, Berkeley, Calif. (1934).

30. Oseen, C.W., Neure Methoden und Ergebmisse in der Hydrodynamik, Akadamische Vergsgesellschaft, Leipzig (1927).

31. Perry, J.H., Chemical Engineer's Handbook, 3rd ed., p. 1018, McGrawhill Book Co., New York, N.Y. (1950).

32. Pansing, W.F., Liquid-liquid Extraction: The Mechanism of Solute Transfer from Single Drops, Ph.D. Thesis in Ch.E., University of Cincinnati (1951).

33. Peebles, F.N., and Garber, H.J., Chem. Eng. Progress, 49, 88 (1933).

34. Prandtl, L., Essentials of Fluid Dynamics (Authorized Translation in English), p. 188, Blackie & Son Ltd., London (1952).

35. Rouse, H., Nomogram for the Settling Velocity of Spheres, Natl. Res. Council, Div. Geol. and Geog., Com. on Sedimentation, Report 1936-37, p. 57 (1937).

36. Rybczyncki, W., Bull. Acad. Sci., Cracovie, 1, 40 (1911).

37. Saito, S., Science Reports, Tohoku Imperial University, Sendai, Japan, Series I, 2, 179 (1913).

38. Schmidt, W., Sitzber. Math. Naturwiss. Klasse, K. Akad. Wiss. Bd 118, Abt IIa, 71 (1909).

39. Smirnov, N.I., Ruban, V.L., Zhur. Priklad. Khim (J. Applied Chem.), 22, 1068 (1949).

40. Spells, K.E., Proc. Phys. Soc. (London), B, 65, 541 (1952).

41. Spilhaus, A.F., Jour. of Meteorology, 5, 108 (1948).

42. Spronck, R., Science et Industrie, Jan. 1934.

43. Squires, L., and Squires, W., Trans. Amer. Inst. Chem. Engrs., 33, 1 (1937).

44. Stokes, G.G., Math. and Phys. Papers, Vol. 3, Cambridge University Press (1901).

45. Wadell, H., Jour. Franklin Inst., 217, 459 (1934).

46. Zahm, A.F., Natl. Adv. Com. Aeronautics, Tech. Rept. No. 253 (1927).

47. Zuidema, H.H., and Pilz, G.P., Anal. Chem., 20, 1244 (1948).

48. Zuidema, H.H. and Waters, G.W., Ind. Eng. Chem., Analytical Edn., 13, 312 (1941).

49. Watson. This reference is reported by Hughes and Gilliland (see Ref. 20) without giving the original reference.

APPENDIX

Estimation of Errors

An estimation of the probable errors is useful in evaluating the validity of the results and judging the conclusions obtained therefrom.

(a) Errors involved in the determination of physical properties: These are generally negligible. An error of 0.1 mg. in weighing the pyknometer can only make an error in 10 ml. capacity pyknometer of the order of 0.001%. If we consider that two weighings are involved for getting the weight of a liquid, the error will be doubled. If it is considered that the density of the liquid is obtained by comparing with that of water, then we get the error still doubled. We then get a total error of 0.004% which is still very low.

An error of 0.1 sec. in timing the stopwatch when taking viscosity measurements will involve an error of 0.20%, if the total time is 50 seconds.

An error of 0.1 $\frac{\text{dyne}}{\text{cm.}}$ in taking the interfacial tension will be 0.40% if the interfacial tension is of the order of 25 $\frac{\text{dynes}}{\text{cm.}}$.

(b) Errors in measurement of the volume of the drop:

(i) If the total volume measured was 0.50 ml and an error of 0.005 ml each were made in taking the initial and final readings, then the error in the measurement of drop volume would be 2%.

(b) (ii) In the case of large size drops, it was found that a small tail drop was formed behind a big drop. There will certainly be an error introduced in the estimation of the volume of the drops and the calculated volume of the drops will be higher. But these secondary drops were very small in comparison to the big size ones, so that the error introduced by neglecting them is small. Laws who worked on water drops in air estimated that the volume of the secondary drop was less than 1% of the big main drop. This error is only half as much as the error involved in the actual measurement of the drop volume. It is also fortunate that this secondary drop formation occurs only in the case of large drops where the velocities of fall do not vary much with size of the drops for any particular system. Hence this did not seem to be a serious error and no actual measurements were made to estimate the errors involved therein.

Thus the total error in the measurement of volume may be estimated at 3% or less.

(c) Errors in measurement of time of fall of the drop: If an error of 0.1 sec. was made in timing the stopwatch, the error would be 2.0% when the total time of fall was 5 sec. However, the actual time is greater than 5 sec. in most cases. The stopwatches were checked against standard time signals and were found to be accurate to less than 0.1 sec. in 10 minutes, which is less than 0.17%. Then the total error in timing is about 2.17%.

(d) Errors in the measurement of distance of fall of the drop. If an error of 0.1 cm. was made in measuring the distance of 130 cms., then the error would be 0.078%.

(e) Errors due to temperature variation: Let the maximum temperature variation allowed be ± 0.5°C. Then the effect of this temperature variation on the different properties of the liquids would be as follows:

	Variation/°C	Variation for 0.5°C.
Density of water	0.0025%	0.0013%
Viscosity of water	2.27%	1.24%
Surface tension of water	0.22%	0.11%
Total variation for 0.5°C. variation in temperature		1.35%

In the absence of any information, the variation in the properties of the organic liquids may be assumed to be of the same order. The variation in the interfacial tension values may also be assumed to be of the same order as for the surface tension of water. Fortunately, the variation of density and viscosity of both water and organic liquid are in the same direction for any variation in temperature, so that the total errors are likely to be less than the above. But it was felt safe to retain the above values which are indicative of the order of errors and not of the absolute errors.

(f) Errors due to turbulence: There can be turbulence created and left unnoticed in the experiment on account of the following reasons: (1) When the column was originally filled by pouring water into the column, there is enough turbulence caused in the operation. In order to let the turbulence die down, about 1/2 hr. to 1 hr. time was allowed after the column was filled, before a run was started. In each run the times of fall of the drops when noted did not show any appreciable difference to suspect that the turbulence did not die down. Hence it was considered that the time allowed was sufficient for the purpose.

(2) Turbulence may also be left behind in the wake of a drop and it may distort the motion of the next drop falling behind it. To avoid this difficulty, and to keep a check on it, the frequency of formation of drops was noted. This was recorded indirectly in Tables A-1 to A-7 as drop formation time, which was a direct measurement. This gives the time interval between successive drops. A minimum of 30 sec. was chosen for the drop formation time and it was generally increased for the bigger size drops. The data so collected seemed to be free from any errors due to residual turbulence left by the previous drops.

(g) Errors due to solubility effects: This error has been minimized by the choice of systems with low mutual solubility. Though this varies for each system, the error introduced by this should never be more than 1%. That there is no such error at least in the properties of water was checked by collecting fall velocity data with same size drops both when the column was freshly filled with distilled water and also at the end of the day's runs. Another check was also obtained by varying the drop formation time. The first one is a check on change of properties of water, while the second one is a check on the change of properties of the discontinuous phase. Actual measurements made on carbon tetrachloride water mixtures shaken in a bottle over time did not show any appreciable differences in the physical properties of either of them. Hence this error is considered to be less than 1%. It may be mentioned here that mixtures of liquids, as well as wetting agents were purposefully avoided as they might bring in additional complications.

(h) Errors in drop velocities due to zigzag path of the drops: The zigzag path seems to be inherent in the fall of liquid droplets. So it was felt that an average of large numbers of fall velocity observations should be taken instead of accepting any single value.

(i) Errors due to oscillations of drops: This also was accepted as part of the drop behaviour and the only method to get over the difficulty was considered as taking an average of a number of observations.

TABLE A-1

FALL VELOCITIES OF CARBON TETRACHLORIDE DROPS IN WATER IN A 3-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
24.8	20	0.00466	53.5-54.6	9	8.23	0.207	15.8
24.9	16	0.00527	36.6-44.0	15	7.51	0.216	17.3
25.3	15	0.00575	75.1-83.2	12	7.26	0.223	17.9
24.9	16	0.00601	38.5-39.3	12	6.83	0.226	19.0
25.1	15	0.0101	62.1-62.3	12	6.50	0.268	20.0
25.0	15	0.0101	53.0-54.2	9	6.48	0.268	20.1
25.2	10	0.0133	100.9-124.2	7	6.27	0.294	20.7
25.2	12	0.0142	85.0-86.1	11	6.11	0.300	21.3
25.3	10	0.0168	126.4-130.8	6	5.92	0.318	22.0
24.8	10	0.0191	53.8-54.7	5	5.84	0.332	22.3
25.0	7	0.0256	196-221	6	6.10	0.366	21.3
24.7	7	0.0257	155.2-162.0	2	5.95	0.366	21.9
25.2	6	0.0302	140.6-147.3	3	6.03	0.387	21.6
25.3	6	0.0308	153.1-178.0	4	5.90	0.389	22.0
25.5	4	0.0469	244-255	3	6.17	0.448	21.1

* Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-2

FALL VELOCITIES OF CARBON TETRACHLORIDE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
25.5	17	0.00433	41.2-44.2	12	8.07	0.202	16.1
25.4	20	0.00445	40.8-43.0	15	8.04	0.204	16.2
24.8	18	0.00535	70.3-94.4	16	7.88	0.217	16.5
25.5	15	0.00673	67.4-79.9	14	6.98	0.234	18.6
24.9	15	0.00726	87.6-101.9	13	7.10	0.240	18.3
25.2	10	0.00990	34.6-35.8	8	6.52	0.266	19.9
25.1	10	0.0100	35.9-37.1	9	6.43	0.267	20.3
24.8	10	0.0132	82.2-89.5	9	6.18	0.293	21.0
24.8	30	0.0148	27.5-37.0	24	5.99	0.304	21.7
25.3	8	0.0151	92.5-97.1	6	6.02	0.307	21.6
24.7	35	0.0151	37.9-42.4	27	6.02	0.307	21.6
25.0	30	0.0185	33.5-38.4	25	5.83	0.328	22.3
24.9	30	0.0186	36.1-40.8	25	5.82	0.329	22.3
25.3	9	0.0190	66.5-68.5	8	5.80	0.331	22.4
24.6	25	0.0214	37.6-41.8	22	5.72	0.345	22.7
25.1	8	0.0219	139.4-143.7	7	5.82	0.347	22.3
25.2	25	0.0220	46.7-60.1	21	5.70	0.348	22.8
24.9	7	0.0234	200.7-239.0	5	5.86	0.355	22.2
24.7	20	0.0256	32.2-42.4	18	5.81	0.366	22.4
24.8	20	0.0258	28.3-32.2	18	5.82	0.367	22.3

(Continued)

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-2 (Continued)

FALL VELOCITIES OF CARBON TETRACHLORIDE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
24.7	20	0.0301	40.6-45.1	18	5.86	0.386	22.2
24.8	20	0.0302	40.4-44.8	17	5.88	0.386	22.1
25.1	20	0.0317	49.4-56.1	19	5.94	0.393	21.9
25.1	15	0.0347	60.9-67.2	14	5.86	0.405	22.2
25.0	15	0.0366	59.8-156.3	14	5.91	0.412	22.0
25.2	10	0.0470	59.6-66.8	9	6.11	0.448	21.3
25.3	10	0.0472	65.5-72.4	8	6.16	0.448	21.1
25.5	10	0.0480	64.5-110.2	9	6.09	0.451	21.3
24.8	10	0.0488	76.6-88.8	8	6.13	0.453	21.2
25.1	10	0.0510	94.2-184.8	9	6.10	0.458	21.3
25.4	8	0.0681	42.0-61.0	6	6.45	0.508	20.2
25.4	7	0.0757	83.0-115.9	5	6.52	0.526	19.9
25.4	7	0.0793	124.1-193.8	6	6.47	0.534	20.1
25.0	5	0.0950	83.5-235	4	6.58	0.565	19.7
25.0	5	0.0996	229-520	4	6.48	0.575	20.0
25.4	5	0.1024	239-257	4	6.64	0.581	19.6
25.1	5	0.116	57.9-95.2	4	6.59	0.605	19.7
25.1	4	0.116	116.1-121.8	3	6.67	0.605	19.5
25.0	4	0.120	107.4-213.4	3	6.65	0.612	19.5

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-3

FALL VELOCITIES OF TETRACHLOROETHANE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
25.0	20	0.00411	42.3-44.3	18	7.94	0.199	16.4
25.0	20	0.00426	57.8-61.7	18	8.27	0.201	15.7
25.0	20	0.00533	48.7-49.8	17	7.60	0.217	17.1
25.0	18	0.00734	49.3-51.2	17	6.96	0.241	18.7
25.0	15	0.00860	86.7-89.4	14	7.09	0.254	18.4
25.0	21	0.00894	51.4-52.4	20	6.99	0.257	18.6
25.0	15	0.00978	35.5-36.2	13	6.73	0.265	19.3
25.0	15	0.0101	51.5-52.7	14	6.56	0.268	19.8
25.0	18	0.0102	107.8-110.1	17	6.49	0.269	20.0
24.9	30	0.0108	28.9-48.0	27	6.25	0.274	20.8
25.0	15	0.0113	100.3-105.5	12	6.34	0.278	20.5
24.8	30	0.0118	35.6-65.6	29	6.26	0.282	20.8
24.8	25	0.0118	31.0-53.3	24	6.17	0.282	21.1
25.0	25	0.0138	29.6-64.6	23	5.99	0.298	21.7
24.5	40	0.0141	31.9-45.1	36	5.99	0.300	21.7
25.1	20	0.0145	31.1-78.3	18	6.04	0.302	21.5
24.85	20	0.0145	47.3-78.7	18	5.97	0.302	21.8
24.55	35	0.0146	41.0-87.7	31	6.10	0.303	21.3
25.05	35	0.0149	33.2-57.8	34	6.13	0.306	21.2
25.0	35	0.0157	39.8-51.3	33	6.11	0.311	21.3

(Continued)

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-3 (Continued)

FALL VELOCITIES OF TETRACHLOROETHANE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
25.0	11	0.0170	131.1-134.1	9	6.22	0.319	20.9
25.1	6	0.0173	60.3-61.2	5	6.23	0.321	20.9
25.0	30	0.0178	38.9-53.4	28	6.22	0.324	20.9
24.9	10	0.0179	107.6-109.6	6	6.17	0.325	21.1
25.0	25	0.0180	35.3-53.6	22	6.21	0.325	20.9
25.0	15	0.0185	44.9-222.6	14	6.17	0.328	21.1
25.0	15	0.0187	36.3-86.9	14	6.24	0.330	20.9
24.8	10	0.0188	66.8-69.0	4	6.26	0.330	20.8
24.7	20	0.0225	36.4-60.4	19	6.18	0.351	21.0
24.7	25	0.0230	59.0-75.4	24	6.56	0.354	19.8
24.8	25	0.0234	45.2-59.2	24	6.61	0.355	19.7
24.8	8	0.0244	165.1-173.8	3	6.37	0.360	20.4
24.8	20	0.0315	30.8-106.1	18	6.62	0.392	19.6
24.6	10	0.0318	50.2-110.6	9	6.63	0.394	19.6
24.9	15	0.0330	34.9-82.3	14	6.59	0.398	19.7
24.8	10	0.0470	48.0-122.6	9	6.87	0.448	18.9
24.7	10	0.0470	38.0-75.1	9	6.84	0.448	19.0
25.0	8	0.0653	93.1-205.7	7	7.07	0.500	18.4
24.6	8	0.0663	69.0-108.6	7	7.09	0.503	18.3
25.0	8	0.0665	73.8-113.8	7	7.05	0.503	18.4
24.6	8	0.0669	82.9-152.5	7	7.05	0.504	18.4
24.6	8	0.0669	40.6-60.4	7	7.14	0.504	18.2
24.9	7	0.0771	53.6-131.5	6	7.22	0.528	18.0
24.9	6	0.0797	151.1-268.4	5	7.15	0.535	18.2
25.0	7	0.0854	94.2-116.3	6	7.24	0.547	18.0

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop

(Continued)

TABLE A-3 (Continued)

FALL VELOCITIES OF TETRACHLOROETHANE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*			Drop Diameter Cm.	Drop Velocity
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.			
24.9	7	0.0857	158.4-257.0	6	7.21		0.548	18.0
24.9	7	0.0879	85.6-133.0	6	7.23		0.552	18.0

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-4

FALL VELOCITIES OF O-NITROTOLUENE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observa-tions	Average Time, Sec.		
25.5	10	0.00592	31.5-52.4	9	17.27	0.224	7.52
25.3	10	0.00603	32.7-171.8	8	17.25	0.226	7.53
25.0	10	0.00628	107.1-108.7	7	16.23	0.229	8.00
25.2	10	0.00642	83.6-85.8	9	16.73	0.231	7.75
25.2	10	0.00644	81.6-82.1	7	16.74	0.231	7.75
25.4	10	0.00885	140.5-145.4	8	15.36	0.257	8.46
25.4	10	0.00892	139.6-145.9	9	15.33	0.257	8.47
24.8	20	0.0275	45.8-90.2	18	11.87	0.375	11.0
24.9	20	0.0275	32.8-54.7	19	11.85	0.375	11.0
25.4	15	0.0323	36.9-55.9	14	11.49	0.396	11.3
25.2	15	0.0327	51.5-85.7	14	11.47	0.397	11.3
25.0	15	0.0351	39.3-53.5	12	11.15	0.406	11.7
25.1	15	0.0357	35.0-42.2	14	11.14	0.408	11.7
25.1	13	0.0469	56.5-102.6	12	10.61	0.448	12.3
24.9	10	0.0472	102.7-128.1	9	10.69	0.448	12.2
25.1	10	0.0515	52.6-58.3	8	10.35	0.462	12.6
24.8	10	0.0535	73.4-102.7	9	10.36	0.468	12.6
25.0	10	0.0612	63.9-80.4	9	9.96	0.489	13.0
25.0	8	0.0619	89.6-119.3	7	10.04	0.491	12.9
24.9	10	0.0622	99.5-132.3	9	10.04	0.492	12.9

(Continued)

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-4 (Continued)

FALL VELOCITIES OF O-NITROTOLUENE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
25.2	6	0.0892	114.7-149.8	5	10.44	0.555	12.4
25.2	6	0.0892	88.6-119.7	5	10.42	0.555	12.5
25.0	5	0.111	149.3-208.3	4	10.53	0.597	12.3
24.9	4	0.1125	112.8-245.3	3	10.40	0.599	12.5
24.9	5	0.130	102.3-132.5	4	10.38	0.628	12.5
25.0	4	0.1313	215-357	3	10.60	0.631	12.3
24.8	5	0.141	102.6-149.9	4	10.03	0.646	12.9
24.8	5	0.1426	169-317	5	10.22	0.648	12.8
24.6	3	0.1433	403-573	3	10.57	0.649	12.3
25.2	3	0.202	628-687	3	10.30	0.728	12.6
25.5	3	0.203	621-683	3	10.27	0.729	12.7
25.4	3	0.210	422-489	3	10.23	0.737	12.7
25.2	4	0.2205	446-888	3	10.28	0.750	12.7
25.2	4	0.223	1122-2183	3	8.18#	0.752	12.2
25.4	3	0.269	2340-2760	3	8.17#	0.800	12.2
24.7	2	0.329	1659-2340	2	8.45#	0.856	11.8
25.0	3	0.337	3000-4500	3	8.30#	0.864	12.0
25.1	4	0.374	1380-3300	4	8.28#	0.893	12.1
25.4	2	0.428	730-1500	2	8.30#	0.935	12.0
25.4	2	0.430	1020-1500	2	8.40#	0.936	11.9

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.
#Time of fall of 100 cms. after an initial fall of 60 cms. of the drop.

TABLE A-5

FALL VELOCITIES OF S-TETRABROMOETHANE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
25.0	40	0.000786	24.6-28.4	35	6.47	0.114	20.1
25.0	40	0.000865	24.8-27.7	36	6.30	0.118	20.6
25.0	40	0.00121	28.0-32.7	37	5.77	0.132	22.5
25.0	40	0.00174	35.6-40.1	36	5.32	0.149	24.4
25.0	30	0.00253	36.7-38.3	29	4.93	0.169	26.4
25.0	25	0.00291	40.2-42.0	24	4.81	0.177	27.0
25.0	30	0.00294	47.7-53.2	28	4.75	0.178	27.4
25.0	30	0.00332	45.9-52.0	25	4.67	0.185	27.8
25.0	20	0.00401	65.0-69.5	19	4.46	0.197	29.1
25.0	25	0.00431	51.5-54.7	23	4.47	0.202	29.0
25.0	25	0.00448	73.4-79.6	23	4.49	0.204	28.9
25.0	20	0.00464	53.3-55.3	17	4.41	0.207	29.5
25.0	20	0.00547	114-127	18	4.43	0.219	29.3
25.0	20	0.00577	50.8-54.5	17	4.56	0.223	28.5
25.0	15	0.00629	87.6-91.7	13	4.52	0.229	28.8
25.0	20	0.00728	72.7-75.5	18	4.69	0.240	27.7
25.0	15	0.00965	94.1-104.4	14	4.80	0.264	27.1
24.9	15	0.00999	104-108	14	4.81	0.267	27.0
25.0	40	0.0101	49.2-99.4	36	4.80	0.268	27.1
25.0	40	0.0116	41.7-53.2	38	4.81	0.280	27.0
25.0	30	0.0154	46.3-68.3	27	5.16	0.308	25.2
25.0	20	0.0244	74.5-99.3	18	5.20	0.360	25.0
25.0	20	0.0246	119-150	18	5.22	0.361	24.9

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-6

FALL VELOCITIES OF ETHYL CHLOROACETATE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature °C	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
24.8	20	0.00512	28.9-31.3	15	12.99	0.214	10.0
24.8	12	0.00626	62.1-66.1	10	12.21	0.229	10.6
25.0	15	0.00639	28.6-30.1	14	11.75	0.230	11.1
24.8	12	0.00681	64.5-73.9	10	11.66	0.235	11.2
25.0	16	0.00989	53.8-60.7	15	10.04	0.266	12.9
25.0	10	0.0137	54.4-58.6	9	9.54	0.297	13.6
24.8	10	0.0166	159-171	9	9.53	0.316	13.6
24.9	10	0.0171	163-166	9	9.56	0.319	13.6
24.9	10	0.0188	158.5-159.3	9	9.46	0.330	13.7
25.0	25	0.0223	36.0-39.8	24	9.61	0.349	13.5
25.0	20	0.0295	45.7-54.5	19	9.91	0.383	13.1
25.0	20	0.0308	99.3-111.4	18	9.90	0.388	13.1
25.0	17	0.0315	90.3-109.3	15	9.87	0.392	13.2
25.0	15	0.0375	58.6-65.6	13	10.31	0.415	12.6
25.0	15	0.0385	56.5-66.8	13	10.22	0.418	12.7
25.0	12	0.0458	117-131	9	10.58	0.444	12.3
25.0	10	0.0490	135-164	9	10.59	0.454	12.3
24.8	10	0.0675	168-198	9	10.97	0.505	11.9
25.0	10	0.0685	134-167	9	11.07	0.507	11.7
25.0	8	0.0990	116-167	7	11.41	0.574	11.4
25.0	5	0.138	148-165	4	11.93	0.640	10.9
25.0	6	0.143	190-265	4	9.10#	0.648	11.0
25.0	5	0.155	374-495	4	11.98	0.665	10.9

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.
#Time of fall of 100 cms. after an initial fall of 60 cms. of the drop.

TABLE A-7

FALL VELOCITIES OF CHLOROBENZENE DROPS IN WATER IN A 6-INCH COLUMN

Water Temperature ^oC	Drop Volume		Drop Formation Time, Sec.	Time of Fall of Drops*		Drop Diameter Cm.	Drop Velocity Cms/Sec.
	No. of Drops Counted	Average Volume ml.		No. of Observations	Average Time, Sec.		
24.9	10	0.111	78.6-104.2	5	8.98	0.596	14.5
25.0	10	0.139	158-222	4	9.10	0.642	14.3
25.0	10	0.141	186-241	7	9.07	0.645	14.3
25.0	9	0.170	251-331	6	9.13	0.686	14.2
25.0	9	0.176	156-208	7	9.26	0.695	14.0
25.0	4	0.2425	92.8-540	4	9.4	0.772	13.8
25.0	4	0.2475	141-600	4	9.25	0.777	14.1
25.0	5	0.3654	334-586	4	9.60	0.886	13.5
25.0	2	0.510	480-820	2	10.2	0.990	12.7
25.0	3	0.5117	186-363	3	10.07	0.991	12.9
25.0	2	0.7325	481-1500	2	10.25	1.119	12.7

*Time of fall of 130 cms. after an initial fall of 30 cms. of the drop.

TABLE A-8

PROPERTIES OF LIQUIDS USED

Property		Carbon-tetrachloride	Tetrachloroethane	O-Nitrotoluene	s-Tetrabromoethane	Ethylchloroacetate	Chlorobenzene
Density gms/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 Centi-poises	(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.	$41.6^{24.6}$	$31.3^{24.9}$	$26.6^{24.2}$	$36.2^{23.2}$	$14.6^{25.0}$	$35.4^{25.4}$
	(Lit.	45.0 ± 1.0^{20}	-	27.19^{20}	38.82^{20}	$15.8^{25.0}$	37.41^{20}
Solubility % $\left(\frac{w}{w}\right)$ (lit.)	(Liq. in Water	0.077^{25}	0.288^{25}	0.0652^{30}	0.0651^{30}	insoluble	0.0488^{30}
	(Water in Liq.	0.010^{24}	-	-	-	-	4.4
Re		111.5DV	111.5DV	111.5DV	111.5DV	111.5DV	111.5DV
C_D		$\frac{769.5D}{V^2}$	$\frac{765.6D}{V^2}$	$\frac{210.6D}{V^2}$	$\frac{2,564D}{V^2}$	$\frac{194.0D}{V^2}$	$\frac{135.9D}{V^2}$
β		$3.457D^2$	$4.573D^2$	$1.480D^2$	$13.24D^2$	$2.484D^2$	$0.7176D^2$
Tv		0.5254V	0.5262V	0.8093V	0.3518V	0.8318V	0.9365V
Wt		212.3D	212.0D	137.9D	317.3D	134.2D	119.1D
We		$0.01199DV^2$	$0.01593DV^2$	$0.01874DV^2$	$0.01377DV^2$	$0.03414DV^2$	$0.01408DV^2$
$\frac{\mu_i}{\mu_o}$		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

TABLE A-9

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS CARBON TETRACHLORIDE DROPS IN WATER

D, Cms.	V, Cms/Sec.	Re	C_D	C_r		T_V	Wt	We
0.20	15.91	355.0	0.608	0.984	0.1382	8.37	42.5	0.608
0.22	17.41	427.0	0.559	0.963	0.1672	9.15	46.7	0.800
0.24	18.77	502.5	0.525	0.955	0.199	9.86	51.0	1.014
0.26	19.90	577.0	0.506	0.958	0.234	10.44	55.2	1.237
0.28	20.80	649.0	0.498	0.978	0.271	10.92	59.5	1.455
0.30	21.52	719.0	0.499	1.012	0.311	11.30	63.7	1.668
0.32	22.09	788.0	0.505	1.047	0.354	11.60	67.9	1.872
0.34	22.42	850.0	0.521	1.108	0.399	11.79	72.1	2.05
0.36	22.55	905.0	0.545	1.160	0.448	11.84	76.4	2.20
0.38	22.42	952.0	0.582	1.252	0.499	11.79	80.6	2.29
0.40	22.12	987.0	0.630	1.368	0.553	11.62	85.0	2.35
0.42	21.80	1021.0	0.681	1.480	0.610	11.47	89.2	2.39
0.44	21.41	1052.0	0.739	1.612	0.668	11.25	93.4	2.42
0.46	21.05	1080.0	0.800	1.758	0.730	11.08	97.5	2.45
0.48	20.71	1110.0	0.861	1.902	0.795	10.89	101.9	2.47
0.50	20.42	1140.0	0.923	2.045	0.864	10.72	106.2	2.50
0.52	20.18	1170.0	0.982	2.183	0.935	10.60	110.3	2.54
0.54	19.97	1203.0	1.040	2.32	1.008	10.49	114.8	2.59
0.56	19.78	1237.0	1.101	2.465	1.047	10.39	118.9	2.63
0.58	19.61	1270.0	1.160	2.615	1.162	10.30	124.1	2.68
0.60	19.52	1308.0	1.211	2.74	1.242	10.26	127.3	2.74
0.62	19.50	1349.0	1.256	2.86	1.329	10.24	131.8	2.83

TABLE A-10

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS TETRACHLOROETHANE DROPS IN WATER

D,Cms.	V, Cms/Sec.	Re	C_D	C_r		T_v	Wt	We
0.20	15.68	349.5	0.623	1.003	0.183	8.25	42.4	0.784
0.22	17.02	418.0	0.582	0.997	0.221	8.96	46.6	1.013
0.24	18.35	491.0	0.546	0.991	0.263	9.66	50.8	1.289
0.26	19.60	568.0	0.518	0.983	0.309	10.31	55.1	1.591
0.28	20.79	649.0	0.496	0.975	0.358	10.96	59.4	1.931
0.30	21.40	715.0	0.502	1.014	0.412	11.28	63.6	2.19
0.32	21.10	753.0	0.550	1.124	0.468	11.11	67.8	2.27
0.34	20.70	785.0	0.607	1.258	0.528	10.90	72.0	2.33
0.36	20.33	816.0	0.667	1.392	0.592	10.70	76.3	2.37
0.38	19.98	846.0	0.729	1.532	0.661	10.51	80.5	2.42
0.40	19.61	875.0	0.796	1.685	0.731	10.31	84.8	2.46
0.42	19.29	903.5	0.865	1.84	0.806	10.16	89.0	2.49
0.44	18.99	931.0	0.935	2.00	0.885	10.00	93.1	2.53
0.46	18.72	961.0	1.002	2.17	0.967	9.86	97.4	2.57
0.48	18.49	989.0	1.074	2.33	1.052	9.75	101.8	2.62
0.50	18.30	1020.0	1.142	2.49	1.142	9.65	106.0	2.67
0.52	18.17	1054.0	1.205	2.64	1.238	9.56	110.1	2.74
0.54	18.04	1088.0	1.270	2.80	1.332	9.51	114.3	2.80
0.56	17.98	1123.0	1.328	2.94	1.432	9.46	118.8	2.88

TABLE A-11

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS O-NITROTOLUENE DROPS IN WATER

D, Cms.	V, Cms/Sec.	Re	C_D	C_r		T_v	Ψt	We
0.22	7.6	186.5	0.801	1.009	0.0716	6.15	30.3	0.238
0.24	8.08	216.2	0.774	1.043	0.0852	6.54	33.1	0.294
0.26	8.55	248.0	0.748	1.063	0.1000	6.92	35.8	0.356
0.28	9.00	281.5	0.727	1.081	0.1160	7.28	38.6	0.426
0.30	9.43	315.2	0.710	1.099	0.1332	7.63	41.4	0.500
0.32	9.90	353.5	0.687	1.110	0.1515	8.01	44.1	0.588
0.34	10.32	391.8	0.672	1.122	0.1710	8.35	46.8	0.680
0.36	10.75	432.0	0.655	1.132	0.192	8.70	49.6	0.780
0.38	11.16	473.0	0.642	1.148	0.214	9.04	52.4	0.888
0.40	11.55	515.0	0.631	1.161	0.237	9.35	55.1	1.000
0.42	11.90	557.0	0.625	1.178	0.261	9.64	57.8	1.118
0.44	12.22	600.0	0.620	1.192	0.286	9.90	60.6	1.234
0.46	12.52	644.0	0.618	1.215	0.313	10.14	63.4	1.350
0.48	12.80	686.0	0.617	1.234	0.341	10.38	66.1	1.475
0.50	12.90	720.0	0.632	1.282	0.370	10.44	68.8	1.560
0.52	12.92	751.0	0.656	1.341	0.400	10.48	71.6	1.628
0.54	12.90	777.0	0.683	1.405	0.432	10.44	74.4	1.688
0.56	12.90	806.0	0.708	1.475	0.464	10.44	77.1	1.750
0.58	12.88	834.0	0.735	1.54	0.498	10.42	79.9	1.805
0.60	12.85	861.0	0.764	1.613	0.533	10.40	82.6	1.860
0.62	12.80	884.0	0.796	1.691	0.569	10.38	85.4	1.902
0.64	12.73	910.0	0.831	1.772	0.606	10.30	88.2	1.947
0.66	12.70	936.0	0.860	1.843	0.645	10.28	90.9	2.000
0.68	12.63	958.0	0.896	1.937	0.684	10.23	93.6	2.035
0.70	12.60	985.0	0.928	2.014	0.725	10.20	96.5	2.080
0.72	12.55	1008.0	0.962	2.091	0.766	10.16	99.2	2.120
0.74	12.51	1033.0	0.995	2.17	0.810	10.13	102.0	2.170
0.76	12.48	1059.0	1.028	2.25	0.855	10.10	104.8	2.222
0.78	12.4	1080.0	1.068	2.35	0.900	10.04	107.5	2.25
0.80	12.38	1106.0	1.098	2.43	0.947	10.02	110.1	2.30
0.82	12.30	1126.0	1.140	2.53	0.995	9.96	113.0	2.325
0.84	12.25	1149.0	1.179	2.62	1.042	9.93	115.9	2.37
0.86	12.20	1172.0	1.218	2.71	1.093	9.88	118.5	2.40

(Continued)

TABLE A-11(Continued)

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS O-NITROTOLUENE DROPS IN WATER

D,Cms.	V, Cms/Sec.	Re	C_D	C_r		T_v	Wt	We
0.88	12.10	1188.0	1.264	2.82	1.145	9.80	121.2	2.42
0.90	12.03	1208.0	1.310	2.93	1.199	9.75	124.0	2.443
0.92	11.99	1230.0	1.348	3.02	1.252	9.70	126.8	2.480
0.94	11.90	1248.0	1.399	3.13	1.308	9.65	129.5	2.495

TABLE A-12

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS
s-TETRABROMOETHANE DROPS IN WATER

D,Cms.	V, Cms/Sec.	Re	C_D	C_r		T_v	Wt	We
0.12	20.86	279.7	0.707	1.052	0.191	7.34	38.1	0.718
0.14	23.28	364.0	0.662	1.088	0.2595	8.20	44.4	1.042
0.16	25.52	455.2	0.630	1.112	0.339	8.97	50.8	1.432
0.18	27.53	552.0	0.609	1.146	0.428	9.67	57.1	1.878
0.20	28.91	643.0	0.614	1.202	0.530	10.18	63.4	2.300
0.22	28.88	708.5	0.676	1.358	0.641	10.17	69.8	2.525
0.24	28.01	749.9	0.784	1.598	0.762	9.85	76.1	2.593
0.26	27.21	790.0	0.900	1.858	0.895	9.58	82.5	2.645
0.28	26.52	828.0	1.021	2.13	1.039	9.33	88.8	2.71
0.30	25.93	868.7	1.142	2.40	1.192	9.12	95.2	2.77
0.32	25.48	908.7	1.263	2.69	1.358	8.96	101.4	2.86
0.34	25.13	950.2	1.380	2.96	1.531	8.84	107.8	2.96
0.36	24.92	1000.0	1.487	3.23	1.716	8.76	114.1	3.08

TABLE A-13

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS ETHYL CHLOROACETATE DROPS IN WATER

D,Cms.	V Cms/Sec.	Re	C_D	C_r		T_v	Wt	We
0.22	10.35	254.3	0.398	0.569	0.1202	8.62	29.55	0.805
0.24	11.68	313.1	0.341	0.526	0.1431	9.72	32.20	1.118
0.26	12.65	367.5	0.316	0.517	0.168	10.52	34.90	1.426
0.28	13.28	414.9	0.308	0.527	0.195	11.05	37.60	1.686
0.30	13.64	456.9	0.313	0.551	0.2239	11.35	40.25	1.905
0.32	13.71	490.0	0.330	0.598	0.2545	11.41	42.90	2.050
0.34	13.62	517.0	0.355	0.654	0.2875	11.32	45.60	2.155
0.36	13.42	539.5	0.388	0.723	0.322	11.18	48.40	2.215
0.38	13.20	559.7	0.421	0.795	0.359	10.99	51.00	2.260
0.40	12.98	579.0	0.461	0.880	0.3975	10.80	53.70	2.30
0.42	12.75	598.0	0.501	0.963	0.438	10.60	56.40	2.33
0.44	12.54	615.1	0.542	1.048	0.482	10.43	59.10	2.365
0.46	12.33	631.5	0.587	1.147	0.525	10.27	61.75	2.385
0.48	12.13	649.8	0.632	1.242	0.572	10.10	64.40	2.410
0.50	11.93	666.7	0.681	1.353	0.621	9.93	67.10	2.430
0.52	11.76	681.8	0.728	1.454	0.673	9.78	69.80	2.458
0.54	11.58	696.6	0.780	1.563	0.724	9.63	72.50	2.470
0.56	11.42	712.3	0.831	1.678	0.778	9.50	75.20	2.495
0.58	11.30	730.0	0.881	1.79	0.836	9.40	77.80	2.525
0.60	11.18	748.5	0.930	1.90	0.895	9.30	80.50	2.56
0.62	11.07	766.2	0.980	2.01	0.956	9.20	83.20	2.59
0.64	10.98	782.5	1.030	2.13	1.018	9.14	85.90	2.63
0.66	10.90	801.0	1.078	2.24	1.082	9.07	88.60	2.675

TABLE A-14

DRAG COEFFICIENTS AND DIMENSIONLESS GROUPS
CHLOROBENZENE DROPS IN WATER

D,Cms.	V, Cms/Sec.	Re	C_D	C_r		T_v	Wt	We
0.60	14.50	971.0	0.388	0.840	0.2583	13.60	71.5	1.771
0.64	14.41	1030.0	0.419	0.915	0.2940	13.50	76.4	1.868
0.68	14.29	1085.0	0.453	0.998	0.332	13.39	81.0	1.960
0.72	14.12	1134.0	0.491	1.089	0.372	13.22	85.8	2.01
0.76	13.97	1180.0	0.529	1.178	0.414	13.08	90.6	2.09
0.80	13.78	1230.0	0.573	1.282	0.459	12.90	95.4	2.14
0.84	13.59	1273.0	0.618	1.392	0.506	12.72	100.1	2.185
0.88	13.40	1315.0	0.666	1.510	0.555	12.55	105.0	2.225
0.92	13.20	1353.0	0.716	1.630	0.607	12.38	109.7	2.26
0.96	13.06	1398.0	0.765	1.742	0.661	12.23	114.5	2.305
1.00	12.93	1442.0	0.812	1.86	0.7176	12.12	119.1	2.355
1.04	12.85	1490.0	0.856	1.97	0.776	12.03	124.0	2.42
1.08	12.79	1541.0	0.896	2.08	0.836	11.98	128.8	2.490
1.12	12.71	1589.0	0.941	2.19	0.902	11.91	133.5	2.545
1.16	12.70	1644.0	0.976	2.27	0.966	11.90	138.2	2.64
1.20	12.70	1700.0	1.010	2.36	1.034	11.90	143.0	2.725