

MECHANICS OF DROP MOTION IN STATIONARY
LIQUID MEDIA

by

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INTRODUCTION

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

A number of attempts to analyze such systems mathematically so as to express the speed and motion of a liquid drop moving in a liquid field have been made. The hydrodynamic aspect of the problem is so complex that even for the more simple cases analyzed (33) many simplifying assumptions have to be made in order to render any mathematical solution possible. Unrealistic simplifying assumptions in setting up the model have made most of the analyses of limited accuracy and applicability. In the present situation, therefore, it is more logical to pursue an experimental approach rather than to attempt a mathematical analysis, except in the region of low Reynolds numbers, where viscous forces are completely dominant. With these considerations the present study was initiated with the objective of reviewing and evaluating the relative merits of the mathe-

matical correlations proposed by different workers for predicting the motion of single liquid drops in stationary field fluids.

LITERATURE REVIEW

Rate of Rise or Fall of Single Drops

Several recent papers (23, 28, 29, 36, 53) dealing with the terminal velocities of liquid drops rising or falling through a liquid field have reviewed the existing pertinent literature. Water was, in nearly all cases, used as the continuous phase.

Several efforts have been made to correlate the terminal velocities of drops with their size and the physical properties of the system. Terminal velocity is attained when the forces of drag, buoyancy and weight are in a dynamic balance. Three of these authors (23, 28, 36) have reported the motion and terminal velocity as functions of the drop equivalent spherical diameter and the pertinent physical properties of the system for field liquids of viscosities near that of water. The size of the drop is usually described by the convenient and easily determined equivalent diameter, defined as the diameter of a sphere having the same volume as that possessed by the drop. The terminal velocity of liquid droplets expressed as a function of drop diameter shows two clear regimes. These can be described in terms of Weber numbers and Reynolds numbers, along with the conventional drag coefficient, but the hump or maximum found to exist in the transition zone between the two regimes has not yielded much to analytical description.

Hu and Kintner (23) studied the motion of single drops of ten organic liquids falling in a stationary water field in a glass walled tank 1 ft. square and 4 ft. high and presented a correlation for nine systems with the exception of the aniline-water system, in the form of a single curve relating the drag coefficient, Weber number, Reynolds number, and a physical property group. The interfacial tensions ranged from 24 to 45 dynes/cm. and the drop densities from 1.100 to 2.947 g./ml., the latter resulting in a twenty-fold range of density differences. The range of drop size covered ranged from $(1.6) \cdot 10^{-4}$ to 2.000 mm. in diameter, and Reynolds number varied from 10 to 2200.

They correlated the experimental results by dimensional analysis and introduced a physical property group called the F group. Thus,

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

or,

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

If the equation is solved dimensionally,

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

where

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

In the analysis of their data, however, Hu and Kintner found that a modified form of the M group, called the P group, gave better correlations than the M group:

$$F = \frac{1}{F} \frac{\rho}{\Delta\rho}$$

The authors found that below Re of about of 300 liquid drops behave essentially like solid spheres.

The peak terminal velocity which is the maximum attainable drop terminal velocity is of special interest in view of its direct bearing on practical liquid-liquid contacting operations. Data for these peaks indicate that the following relationship exists:

$$(Re/We)_p = 0.816F^{0.238} \quad (1)$$

On simplification,

$$(U_o)_p = 1.23 \left(\frac{G_1}{F} \right) F^{-0.238} \quad (2)$$

They also found that at peak velocities an average We value of 3.58 seemed to be a good representation for all the systems. Thus,

$$(We)_p = 3.58 \quad (3)$$

When $(d)_p$ is solved for,

$$(d)_p = 3.58 \left(\frac{G_1}{\rho} \right) / (U_o^2)_p \quad (4)$$

The authors found the best correlation of their data was a plot of $\log C_D \cdot We F^{0.15}$ vs. $\log (Re/F^{0.15})$ plot which was capable of bringing all the data into one single curve for the entire Re range covered in this study. The curve shows a distinct break at the Re region approximately corresponding to that where the peak terminal velocity occurs. When the pertinent physical properties of a system are known, this curve can be used directly to obtain the terminal velocity for any given drop size and hence the corresponding C_D , We and Re are readily

calculable. The equations representing the generalized curve are:

$$Y = (4/3) X^{1.275} \text{ for } 2 < Y \leq 70 \quad (5)$$

$$Y = (0.045) X^{2.37} \text{ for } Y \geq 70 \quad (6)$$

where

$$Y = C_D We_F^{0.15}$$

and

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

At the break point $Y = 70$ and $X = 22.25$

For each system there is a definite maximum drop size, or critical size, beyond which the falling drop cannot be preserved without being ruptured due to excessive shear. The maximum, or critical drop size of the systems was found to be dependent on the criterion

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

Thus critical drop size is given by

$$d_c = \left[(1.452) \cdot 10^{-2} \frac{\rho_l}{\Delta \rho} \right]^{\frac{1}{2}} \quad (7)$$

Licht and Harasimhamurty (36) studied six liquid systems and obtained fall velocity data under stationary conditions. They presented a correlation which permits a computation of the velocity in the region of large drops where the velocity is nearly constant. Excellent outline shapes and photographs of the falling drops were included.

They plotted the ratio of the drag coefficient of a liquid drop to that of a rigid sphere of the same volume and density and the same fall velocity as the droplet as ordinate

against the dimensionless group β as abscissa. In the larger drop sizes as represented by higher values of β , the curves were nearly linear on a rectangular coordinate plot. The equation of the mean line representing all six curves was found to be:

$$C_r = \left(\frac{C_D}{C_D^S} \right)_{Re} = 1.87\beta + 0.425 \quad (8)$$

$$\beta = \frac{(\rho_1 - \rho_0)gd^2}{4\sigma_1}$$

This equation can be used for the case of liquid drops in water for the range of $\beta \geq 0.4$, i.e., for the range of drop sizes larger than that corresponding to the maximum fall velocity.

Klee and Treybal (29) investigated eleven liquid systems covering a wide range of physical properties. They divided the velocity versus drop size curve into two regions and arrived at an equation for each. They used water, furfural and aqueous sucrose as field liquids, all of them in the low viscosity range.

They used a dimensional analysis similar to that used by Hu and Kintner (23) for developing their correlations. They considered the velocity-diameter curves as forming two separate regions: region I where the velocity increases with diameter and region II where the velocity remains substantially constant with increasing diameter. By the method of least squares, with the data for all the measurements in each region, the equations obtained were for region I:

$$Re = 22.2 C_D^{-5.18} We^{-0.169} \quad (9)$$

and for region II:

$$Re = 0.00418 C_D^{2.91} We^{-1.81} \quad (10)$$

Equations (9) and (10) when solved for velocity give:

$$U_{oI} = 38.3 \rho^{-0.45} \Delta \rho^{0.58} \mu^{-0.11} d^{0.70} \quad (11)$$

$$U_{oII} = 17.6 \rho^{-0.55} \Delta \rho^{0.28} \mu^{0.10} \sigma_1^{0.18} \quad (12)$$

The critical diameter d_C which determines when the transition from region I to II is to be made is obtained by solving equations (11) and (12) simultaneously, eliminating U_o :

$$d_C = 0.33 \rho^{-0.14} \Delta \rho^{-0.43} \mu^{0.30} \sigma_1^{0.24} \quad (13)$$

Johnson and Braida (26) studied the rate of fall of liquid drops passing through quiescent liquid phases, with an attempt to correlate the velocity data with the circulation and oscillation of the drops. Water and glycerol solution ranging in density from 0.9970 to 1.1919 gms/cc., and viscosity from 0.3937 to 27.73 centipoises were used as the continuous phases. They found that their data lay parallel to the Hu-Kintner curve and that by applying a continuous phase viscosity correction, i.e., by introducing a viscosity ratio term, the points would fall on the curve for drops falling through water. The break point in the curve was found to serve as a criterion for oscillation.

Warshay, Bogusz, Johnson and Kintner (65) extended the range of field fluid viscosity to the 500 centipoise value.

Keith and Hixson (28) investigated over a dozen liquid-liquid systems, six of them in the low interfacial tension

range. In their experiments, the water phase was moving through the containing tube in a direction counter-current to that of the drop.

Smirnov and Ruban (55) analyzed their earlier experimental data, and developed expressions for Reynolds numbers to cover a range of Reynolds numbers from 0.001 to 1,000. They presented three different equations for turbulent, transition and laminar flow, involving the criteria of Galileo, Archimedes and geometric similarity. They worked with tubes of such small diameter that the movement of the drops was affected by the container walls. The field fluid was in continuous countercurrent motion in most of their runs..

Oscillations

The effect of oscillations on the terminal velocity was discussed in some recent papers by Licht and Narasimhamurty (36), Hu and Kintner (23), Garner and Skelland (15) and Lamb (33). Oscillations are present in large drops for systems of low interfacial tensions. Licht and Narasimhamurty (36) pointed out that oscillating drops show unequal velocities of fall when time intervals are reduced. Hu and Kintner observed vigorous oscillations in the falling drops. The deformation in drop shape may or may not be noticeable, depending on the relative magnitudes of the interfacial tension, and the density difference of the system concerned. This indicates that the oscillations induced by the peak velocity is the main reason for the

rapid increase in the drag coefficient and the corresponding decrease in terminal velocity within this short Reynolds number range. After the abrupt rise in the $\log C_D$ versus $\log Re$ curve, deformation of the drops becomes more and more severe and the oscillations progressively more erratic.

Garner and Skelland (15) showed that the oscillation of drops reduces the fall velocity. The extent of these prolate and oblate oscillations increases with drop size. In one case the reduction in fall velocity was found to be at least 12 per cent. The oscillations were found to cause a marked increase in the rate of mass transfer.

Previous workers (33, 49, 57, 61) have also analyzed the deformation of moving drops without oscillation and the oscillation of deformed drops without gross drop motion. Solutions for such ideal cases are expressed in terms of the eccentricity of the drops, the amplitude and frequency of their oscillations, and the amplitude of oscillation decay, as related to drop size and velocity and the physical properties of the system. Despite these mathematical formulations, the exact nature of drop motion still remains to be determined by experimental methods because of the inseparableness of the two interfering factors in the actual situation.

Shapes

This aspect of drops was studied by Licht and Narasimhamurty (36), Hu and Kintner (23), Johnson and Braida (26), Klee and Treybal (29), Null and Johnson (46) and Keith and Hixson (28).

They observed that there is a deformation in the drop and that this is due to the drag forces involved and differences in pressure acting on various parts of the surface.

A photographic study of the drop shapes was made by Licht and Narasimhamurty, Klee and Treybal and Keith and Hixson, which shows that the smaller size drops are less deformed than larger ones. As the size varies, the shape changes from spherical to ellipsoidal. Licht and Narasimhamurty pointed out that in larger sizes the drop shows oscillations in shape. This adds to reduction and nonuniformity of velocity due to an added resistance to the flow produced by oscillation and deformation. They also pointed out that in the case of several drops there was no axial symmetry in the shape of the drops. Keith and Hixson showed that flattening is proportional to drop size and density difference, and inversely proportional to the surface tension. The flattening of a drop of any given volume has an effect on surface area.

Savic (53), studying circulation and distortion of liquid drops falling through a viscous medium, proposed the existence of a transition from non-circulation to circulating drops. He postulated a relatively incompressible surface layer in which the normal interfacial tension must be added to the integrated effect of the surface tractions to the outside fluid, with opposite sign. He reasoned that this would result in a distortion to tear drop shape which is observed for water drops falling through castor oil. As the drop speed increased, the surface layer became unstable and destroyed the rear stagnation

point to result in a trailing filament. Arnold (2), also observed the same phenomenon. Experiments have indicated (65) that the phenomenon might only occur in non-Newtonian field fluids.

Wall Effect

In order that ultimate velocities measured in tubes of small diameter may be interpreted in terms of those in an infinite medium the effect of wall proximity on the fall of liquid drops has to be determined. The reduction in velocity is caused by a reduction in the area available for fluid to flow around the drop. The correlations proposed are based on the area of the annular space with reference to the drop diameter and tube diameter.

Uno and Kintner (63) studied the effect of wall proximity on the rate of rise of single air bubbles rising through various liquids in vertical cylindrical tubes of different diameters and developed an equation to express a velocity-correction factor in terms of the ratio of bubble diameter to tube diameter and an empirical constant. Strom and Kintner (59) measured the rate of fall of drops of five organic liquids through an aqueous phase contained in vertical cylinders of various diameters and presented a correction factor equation with the more convenient equivalent spherical diameter. Its use is limited to d/D ratios less than one half.

Internal Circulation

Internal circulation refers to the presence of a vortex ring inside the droplet which is produced by drag on the outside of the drop caused by motion of the sphere.

A critical value of the equivalent spherical radius below which internal circulation will not occur in a bubble or drop moving freely through a quiescent liquid has been treated by Bond (4), Bond and Newton (5), Garner and Hammerton (12), Garner and Skelland (15, 11), Haberman and Morton (17), Keith and Hixson (23) and others. If no inhibiting surface active substance is present, circulation is said to occur above this critical radius.

An increase in terminal velocity (resulting in a decrease of the drag coefficient) as compared to that of an equivalent solid sphere has been generally considered to occur as a result of circulation within the drop or bubble (4, 5, 17, 27, 28).

If the movement of the fluid inside the drop be such that fresh fluid be supplied from the interior to the interface, mass and heat transfer across the interface would be aided (14, 22, 30, 35, 40).

Garner, Skelland and Haycock (14a.) studied the speed of circulation inside the drop and found that, for a spherical droplet, the maximum possible circulation velocity is equal to one and a half times the velocity of fall. The velocity of circulation was found to decrease during the fall of the droplet.

Photographs showing circulation patterns have been published by Garner (10), Spells (56), and MacPhail (39).

Effect of Surface Active Agents

These and some normal impurities can and apparently do become oriented at the interface in a two phase system. The role of surface active agents in inhibiting internal circulation has been discussed by Datta, Napier and Newitt (6), Garner and Hale (13), Garner and Hammerton (12), Garner and Skelland (15), Haberman and Morton (17), Hughes and Gilliland (24), Keith and Hixson (28), Lindland and Terjesen (37) and MacPhail (39). MacPhail photographed a circulation pattern from which he deduced that the impurities segregated at one pole. Savic (53) stated that in small drops the suppression of circulation is due to a surface active layer, the extent of which is governed by the balance between interfacial tension and the integral of viscous surface shear.

Fall or Rise of Multiple Droplets

Passage of multiple droplets of a liquid through another partially or totally immiscible liquid due to a difference in density is a familiar phenomenon. The process is used in both industrial and laboratory spray columns for countercurrent liquid-liquid extraction. Design of such equipment requires a knowledge of flow rates, concentrations, and the rate of material transfer for the system. Few quantitative data are available to show the effect on transfer area of changes in flow rates,

number and size of nozzles and physical characteristics of the solvents and the solute (28). The ultimate effect of changing these physical variables is a change in average drop size of the dispersed phase. The transfer area depends on holdup, rate of drop formation, and the average surface area per drop. These factors, in turn, are functions of drop size, rate of rise of drops and the distortion and extent of coalescence of drops (28). It has also been shown that transfer is dependent on convection as well as diffusional forces and that the proportion due to convection is affected by drop size (30, 54). Unusually large material transfer has been found at the dispersed phase inlet (54) or outlet (16); these and other effects are also partially dependent on drop size and number.

Drop Sizes

A number of investigators have studied drop size resulting from the breakup of liquid-into-air jets and gas-into-liquid jets. Some papers on extraction spray towers have made qualitative note of drop sizes (1, 25, 54). Hayworth and Treybal (21) studied drop size in liquid-liquid systems for a static continuous phase at flow rates mostly below the jetting point. A correlation for drop diameter was developed for these systems. The recommended range of rates for this correlation, however, is too low for practical application in most spray towers. Photographs of their flow range were presented and also some data for the largest drops at higher flow rates, but the results are too scattered to fix a peak condition. Several

studies on extraction at very low flow rates have determined drop sizes (8, 34, 54, 67).

Johnsen and Bliss (25) found that spray tower efficiency was increased by avoiding nonuniform drop sizes in the dispersed phase. Plate type distributors tend to give irregular drop sizes, as noted by Blanding and Elgin (3); a picture by Laddha and Smith (31) shows the nonuniformity of drop sizes from a hemispherical distributor at low throughput.

Sherwood, Evans and Longcor (54) gave the first quantitative data on calculated interfacial areas and their effect on transfer coefficients for single drop transfer. They found that application of a calculated area term resulted in a coefficient showing an increase in transfer with drop size, as would be expected because of the increased internal convection. Other data (8) show little effect of drop size on the coefficient or show the coefficient becoming constant above a certain drop size.

Keith and Hixson (23) investigated quantitatively the factors affecting drop size in liquid-liquid systems and obtained data on other factors contributing to the calculation of interfacial transfer area. The results can serve two purposes-- to allow calculation of more fundamental transfer coefficients from laboratory extraction tower results and to permit more accurate estimation of the effect on transfer efficiency of various changes in the physical characteristics of extraction columns and their flow systems. The formation and break-up of liquid jets directed upward into an immiscible continuous

water phase were studied for twelve organic solvents in glass nozzles. The jet breakup was coordinated in terms of the nozzle diameter, the size and number of drops formed, and the physical properties of the mutually saturated phases. They found that for each nozzle with each solvent there was a particular flow rate that gave a minimum drop size and a maximum interfacial transfer area per unit of dispersed phase flow. Rates of drop rise were found to be generally below the rates predicted by Newton's law or the drag coefficient for spheres; flattening of the drops and coalescence also affected the interfacial area. They also defined a most suitable range of flow rates for liquid-liquid extraction spray towers and evaluated quantitatively the conditions determining the interfacial transfer area.

Uniformity of Drop Sizes

Keith and Hixson (28) found that the uniformity of the drop sizes obtained on jet breakup could be related to the flow ratios as defined for jet length. Below the jetting point the drops formed were found to be very uniform, and in the varicose region they showed almost as much uniformity. In the region of the critical flow a decrease in uniformity was noted and in the sinuous region an increasingly wide range of drop size was obtained. As the disruptive stage was reached, the sizes became somewhat more uniform and usually tended to be fairly small. When operation was extended into the spray region, which was found to occur at a fairly low velocity for the larger nozzles,

Limits of Suitable Operating Range

Keith and Hixson (28) defined a most suitable range of operation for the dispersion nozzles of liquid-liquid spray towers. The lower end of the range is set by the jetting point velocity since this combines the lowest economical flow rate with high uniformity. The upper limit of the suitable range is determined by the onset of appreciable non-uniformity of drop sizes because this factor gives rise to a reduction of transfer area.

For the best operation of a spray tower, several factors appear to be important (28). Flow of the dispersed phase must be even and steady; each of the dispersion nozzles must receive an equal portion of the total feed; and the nozzle length should be two inches or greater.

The results of several investigators (8, 34, 54, 67) show up to 40 per cent of the extraction in a tower occurring at the dispersed phase inlet. All of these tests were made below the jetting point; with a jet there is no expansion time and a very short formation time, so that no extra transfer would be expected at this point for a tower under normal operating conditions, as reported by Geankoplis and Hixson (35).

Coalescence

Keith and Hixson (28) observed that coalescence is promoted by low interfacial tension and by increasing nozzle size and therefore by increasing drop size; the last also generally im-

plies an increase with higher rates of drop rise. These trends are opposite to those noted by Appel and Elgin (1). The findings of Keith and Hixson (28) seem reasonable because increasing drop size and decreasing interfacial tension both promote oscillation and vibration of the drops and thus increase the chances of contact between drops. One criticism of large spray towers is that in long columns the dispersed phase may coalesce so much that the transfer area is greatly reduced.

Column Holdup and Slip Velocity

In spray towers, the mass transfer area per unit volume is determined by the holdup and by the size of the particles of the dispersed phase formed at the distributor nozzles. Several papers dealing with column holdups and slip velocities have been published recently (3, 7, 20, 38, 43, 44, 50, 52, 62, 63). The most recent of these is the paper published by Weaver, Lapidus, and Elgin (66). They have presented a method for estimating the behavior of the liquid-in-liquid spray column in which droplets of one phase move through a second quiescent phase.

The degree of contact between two phases clearly affects the overall rate of any process occurring across the phase boundary. For most practical transfer operations, however, where one fluid is dispersed through the other it is possible to estimate the contact area or the system holdup only through a difficult direct measurement. Consequently, most earlier studies of transfer processes have been forced to combine the

contact area with intrinsic rate characteristics.

In a previous publication (38), Lapidus and Elgin had presented a detailed theoretical analysis for predicting the behavior of all possible types of vertical moving fluidized systems. The basic postulate of this development was the proposal that a unique relationship exists between the slip velocity and the holdup for a system comprising one particle size. For such a system the holdup is related directly to the interfacial area. Subsequent publications (47, 48, 60) have tested this theory with rigid glass spheres fluidized by water. Under all possible flow arrangements the agreement between theory and experiment was excellent, thereby substantiating the unique relationship mentioned above.

Using this previous work as a model, Weaver, Lapidus and Elgin (66) presented a method for estimating the behavior of the liquid-in-liquid spray column in which droplets of one phase moved through a second quiescent phase. The problem is complicated by the nature of the liquid droplets of being susceptible to internal circulation, oscillation and distortion. The holdup in this case was shown to be related to a slip velocity ratio, a ratio of the relative velocity between the two phases in the column to the terminal velocity of a representative single droplet. Furthermore, this relationship approaches that exhibited by rigid particles of the same size and density. By combining solids fluidization results with information on single liquid droplet terminal velocity one can thus obtain a design estimate of the holdup or interphase contact

area for a liquid-liquid spray tower without recourse to experimentation.

The authors (66) presented holdup data for the systems methyl-isobutyl ketone-water, isobutanol-water, toluene-water and isocamyl alcohol-water operating in a 1 7/8 inch Elgin type spray tower. In each case the organic solvent was the dispersed phase. The holdup was measured directly in the tower as a function of the system parameters by sealing off a part of the tower proper. The system parameters included the dispersed phase flow rate, the droplet size in the column and the physical properties of the organic solvents. Detailed data were also presented to show that the droplet size distribution in the tower was normal in the Gaussian sense.

The compilation of Zenz (69) which correlates fluidization information in three parameters, representing the particle size, velocity and holdup, was the source of information used by Weaver, Lapidus and Elgin (66). The important observation stressed by Lapidus, Elgin and co-workers, that the slip velocity is the pertinent flow variable in all types of solids fluidized systems, allows the extension of the above batch fluidization information to cocurrent and countercurrent operation of both phases. The generalized flow diagram for the fluidization of a given particle size in all modes may thus be deduced entirely from batch-bed data and the slip velocity definition.

$$V_s = \frac{Q_f}{A\epsilon} - \frac{Q_d}{A(1-\epsilon)} \quad (14)$$

On the basis of this generalization, one can suggest the use of the solids fluidization background in characterizing the behavior of masses of moving droplets in a spray column.

It was observed (66), that the holdup increased with flow rate in a nonlinear fashion. The authors also studied the droplet size distribution and found that straight lines which are characteristic of a normal or Gaussian distribution described the cumulative distributions data on probability paper. Taking the 50 per cent mean diameter of the cumulative distribution as the appropriate size parameter the authors illustrated the effect of the slip velocity on the droplet size. The single droplet terminal velocity information was obtained from the experimental studies of Keith and Hixson (28) supplemented when necessary with the correlation of Klee and Treybal. Specifically the 10, 30, 50, 70 and 90 per cent points of the distribution were selected, and the corresponding terminal velocities were averaged. It was found that the results predicted on the basis of the solid particle model agreed with the experimental liquid-liquid results with an average deviation of 11 per cent.

The main weakness of the model is the apparent tendency of all the experimental data to curve somewhat more than the predicted behavior would indicate. This may be due to the involvement of the droplets in gross circulation patterns and to the virtual mass effect which is involved with accelerated (or decelerated) motion when the mass of the displaced fluid relative to that of the moving body is significant.

Terminal Velocities of Clouds of Drops

A study was made of the motion of clouds of liquid drops in a spray column, by this author and Kintner, using water as stationary field fluid. Shower-head type perforated plate nozzles were used for forming the drops. The densities of the drop liquids ranged from 1.04 to 1.6 gms/cm³, the viscosities from 0.926 to 5.15 centipoises and the interfacial tensions from 4.25 to 45 dynes/cm. Drop velocities were obtained by motion picture techniques and compared with single-droplet terminal velocities. Drop sizes were obtained by taking still pictures of the column while in actual operation. The drop size distributions thus obtained were found to be normal. Plots of cumulative size distributions were constructed using arithmetic probability paper. Straight lines characteristic of a normal or Gaussian distribution were obtained. The average drop velocities were found to correspond with the single droplet terminal velocities for a drop size corresponding to the 50 per cent point of the cumulative distributions which is also the volume weighted mean drop size for the distribution. The drop velocity distribution across the column was found to be parabolic.

Plots of column holdup versus dispersed phase flow rate and column holdup versus slip velocity ratio were obtained. The column holdup was found to increase with the dispersed phase flow rate in a nonlinear fashion. A given holdup also defines a unique slip velocity ratio, V_s/V_t . Thus average

droplet terminal velocity can be calculated at any given value of column holdup using the plots of volumetric holdup versus disperse phase flow rate and volumetric holdup versus the slip velocity ratio. A graphical correlation was thus obtained between the drop size distributions, column holdup and the average droplet terminal velocity for the spray column.

THEORETICAL CONSIDERATIONS

Stokes' law

The translation of a solid sphere in an infinite mass of a perfect liquid was first analyzed by Stokes (53) in 1845 and was later reduced to the case of viscous flow from which the famous law bearing his name resulted. In its simplest form, Stokes' law may be written as

$$F = 3\pi\mu d U_0 \quad (15)$$

The assumptions involved in the derivation are:

1. The discontinuities of the fluid are small compared with the size of the sphere.
2. The fluid is infinite in extent.
3. The sphere is smooth and rigid.
4. There is no slip at the interface between the sphere and the fluid.
5. The velocity of the sphere is small.

By keeping the velocity of the sphere small, as required by the last assumption above, the inertial forces due to the motion of the sphere may be neglected in comparison with the

viscous forces involved. The Navier-Stokes equations of motion are then solved to give the Stokes' law of resistance (33). On account of these assumptions applicability of Stokes' law is limited to Reynolds numbers of less than two.

Hadamard-Rybczynski Equation

The case of a liquid sphere was analyzed by Rybczynski (51) in 1911 and independently by Hadamard (18) in the same year. Lamb (33) has given a detailed discussion by considering the internal as well as external motions and has given the following correction factor for Stokes' law when steady state conditions prevail.

$$F = K_1 3\pi \mu d U_0 \quad (16)$$

$$\text{and } U_0 = (1/K_1) g d^2 \Delta \rho / 18 \mu \quad (17)$$

$$\text{where } K_1 = (3\mu_0 + 2\mu) / (3\mu_0 + 3\mu) \quad (18)$$

μ and μ_0 are dynamic viscosities of the external and internal fluids, respectively. If the viscosity of the internal fluid is large compared to that of the external fluid, as for a droplet of water in air, the value of K_1 is approximately unity for the solid sphere. At the other extreme, for example, for an air bubble in a liquid, K_1 is equal to two-thirds. Thus the velocity of ascent of a bubble would be fifty per cent greater than that for the movement of a solid.

Terminal Velocity

Under steady state conditions, the gravity force on a falling spherical drop is exactly balanced by the resistance it

encounters.

$$\frac{\pi d^3}{6} \Delta \rho g = C_D \frac{\pi d^2}{4} \frac{U_o^2 \rho}{2} \quad (19)$$

from which,

$$U_o^2 = \left(\frac{4}{3}\right) \left(\frac{\Delta \rho}{\rho}\right) \left(\frac{dg}{C_D}\right) \quad (20)$$

and,

$$\begin{aligned} C_D &= \left(\frac{4}{3}\right) \left(\frac{\Delta \rho}{\rho}\right) \left(\frac{dg}{U_o^2}\right) \\ &= \left(\frac{4}{3}\right) \left(\frac{\Delta \rho}{\rho}\right) \left(\frac{1}{Fr}\right) \end{aligned} \quad (21)$$

The last relation is the definition of the drag coefficient. If the drop starts to fall from rest it will accelerate for a short distance before reaching its terminal velocity. In this case the force balance becomes,

$$\frac{\pi d^3 \rho_c}{6} \cdot \frac{du}{dt} = \frac{\pi d^3 \Delta \rho g}{6} - C_D \frac{\pi d^2}{4} \cdot \frac{u^2 \rho}{2} \quad (22)$$

However, for a sphere or an ellipsoid moving in a fluid under accelerating conditions, the inertia of the moving body is higher than its actual mass by one-half the amount of the fluid displaced. Hence, a more rigorous expression for the motion of the accelerating drop is,

$$\frac{\pi d^3}{6} \left(\rho_o + \frac{\rho}{2}\right) \frac{du}{dt} = \frac{\pi d^3}{6} \Delta \rho g - C_D \frac{\pi d^2}{4} \cdot \frac{u^2 \rho}{2} \quad (23)$$

when solved,

$$u = U_o \tanh \left(\frac{U_o t}{a}\right) \quad (24)$$

$$\text{where } a = (2/3) (d/C_D) (2 \rho_o + \rho)/\rho \quad (25)$$

for a distance of fall S,

$$\frac{dS}{dt} = u = U_0 \cdot \tanh\left(\frac{U_0 t}{a}\right) \quad (26)$$

Integrating,

$$S = 2.3 a \log \cosh\left(\frac{U_0 t}{a}\right) \quad (27)$$

To find the relation between u and S , the following transformations can be made:

$$\cosh\left(\frac{U_0 t}{a}\right) = e^{S/a} \quad (28)$$

Let $U_0 t/a = x$

From equation (24) we get

$$\begin{aligned} u/U_0 &= \tanh x = \frac{\sinh x}{\cosh x} \\ &= \frac{\sqrt{\cosh^2 x - 1}}{\cosh x} \end{aligned} \quad (29)$$

Squaring,

$$u^2/U_0^2 = 1 - e^{-2S/a}$$

$$u^2 = U_0^2 (1 - e^{-2S/a}) \quad (30)$$

$$u = U_0 \left[1 - \exp\left(-\frac{3 C_D \rho S}{(2 \rho_0 + \rho) d}\right) \right]^{\frac{1}{2}} \quad (31)$$

It can be seen that u approaches U_0 asymptotically as S approaches infinity. For all practical purposes, $u = 0.995 U_0$. Thus it is possible to calculate S , the distance of fall starting from rest that is required for a drop to reach its terminal velocity. If $u = 0.995 U_0$, then $0.995 = \tanh x$. From mathematical tables, $x = 2.99$ and $\log \cosh x = 0.9986$. Therefore,

$$\begin{aligned} S &= 2.3 a \log \cosh x \\ &= (2.3) (0.9986) (2 d/3 C_D) (2 \rho_0 + \rho)/\rho \\ &= (1.53) (d/C_D) (2 \rho_0 + \rho)/\rho \end{aligned} \quad (32)$$

The value of C_D to be used in the above equations is the drag coefficient for steady state motion. Knowing the pertinent physical constants of the system and an approximation of C_D , the distance S required for attaining the true terminal velocity U_0 can be readily calculated.

Wall Effect

Mathematical analysis of the effect of boundary proximity on the rate of fall of solid spheres assuming negligible inertia effects and a low d/D ratio has been carried out by Iadenburg (32), Faxen (9), Happel and Byrne (19), and Wakiya (64). The resulting equation is,

$$\frac{U}{U_0} = \frac{1}{K_w} = 1 - 2.105 (d/D) + 2.087(d/D)^3 \quad (33)$$

A binomial expansion of $[1 - (d/D)]^{2.1}$ leads to

$$1/K_w = 1 - 2.1 (d/D) + 1.155(d/D)^2 - 0.0385(d/D)^3 \quad (34)$$

The difference in the two expressions is small in the range over which they may be applied. Hence the correction factor equation may be expressed in a general form:

$$1/K_w = \Phi (1 - d/D)^n \quad (35)$$

McNown, et al. (41) obtained two different equations for low and high values of (d/D) respectively, based on different assumptions, both of which may be said to be approximated by equation (35) above.

Newton (45) discussed the resistance to a sphere (or cylinder) moving axially in a vertical cylindrical vessel

containing a quiescent incompressible fluid. His expression can be translated into a wall effect correction factor for the case of large inertial effects:

$$\frac{u}{U_o} = \frac{1}{K_w} = \left[1 - \left(\frac{d}{D} \right)^2 \right] \left[1 - \frac{1}{2} \left(\frac{d}{D} \right)^2 \right]^{\frac{1}{2}} \quad (36)$$

McNown and Newlin (42) obtained an equation for the case of (d/D) nearly equal to unity and negligible viscous shear which seemed to be valid for Reynolds numbers above 20,000 and for diameter ratios of 0.75 and higher.

Each of the equations noted indicates that a plot of $\log (1/K_w)$ versus $\log [1 - (d/D)]$ would result in a straight line.

Strom and Kintner (63) presented a correction factor equation with the more convenient equivalent spherical diameter, for drops of organic liquids falling through water. Its use is limited to d/D ratios less than one half.

The equation is,

$$\frac{u}{U_o} = \frac{1}{K_w} = \left[1 - (d/D)^2 \right]^{1.43} \quad (37)$$

The ultimate velocity of a drop of specific size in an infinite medium can thus be calculated from that measured in a small tube by multiplying the latter by the ratio of the tube cross sectional area to the area of the annular space between the tube wall and the drop.

HU-KINTNER CORRELATIONS

Procedure

The data obtained on terminal velocities of single drops

in stationary field fluids by Licht and Narasimhamurty (36), Klee and Treybal (29) and Johnson and Braida (26) were treated using the Hu-Kintner correlation (23).

Using the physical properties and the drop diameter-terminal velocity data, the drag coefficients, Reynolds numbers and Weber numbers were calculated. The physical property group, F , introduced by Hu and Kintner, was calculated from the pertinent physical properties alone for each system. Plots of $C_D We F^{0.15}$ versus $Re/F^{0.15}$ were obtained for each of the systems.

The peak terminal velocities and the drop sizes corresponding to the peak velocity were calculated using empirical relations (equations 2 and 4,) developed by Hu and Kintner and the data of Licht and Narasimhamurty and those of Klee and Treybal.

Finally, the data of Johnson and Braida obtained using 23.5 per cent solution of glycerol in water as the field fluid are considered. A Hu-Kintner plot of the data was obtained and the curve so obtained was made to fall on the water line using the proper correction factor.

Discussion of Results

Terminal velocity data obtained by Hu and Kintner (33) is presented in graphical form in figure 1. All of their systems exhibited high interfacial tensions in excess of 20 dynes per cm., and all the terminal velocity-drop diameter curves show a characteristic hump near peak velocity. They found that a plot of $\log C_D We F^{0.15}$ versus $\log Re/F^{0.15}$ resulted in a single

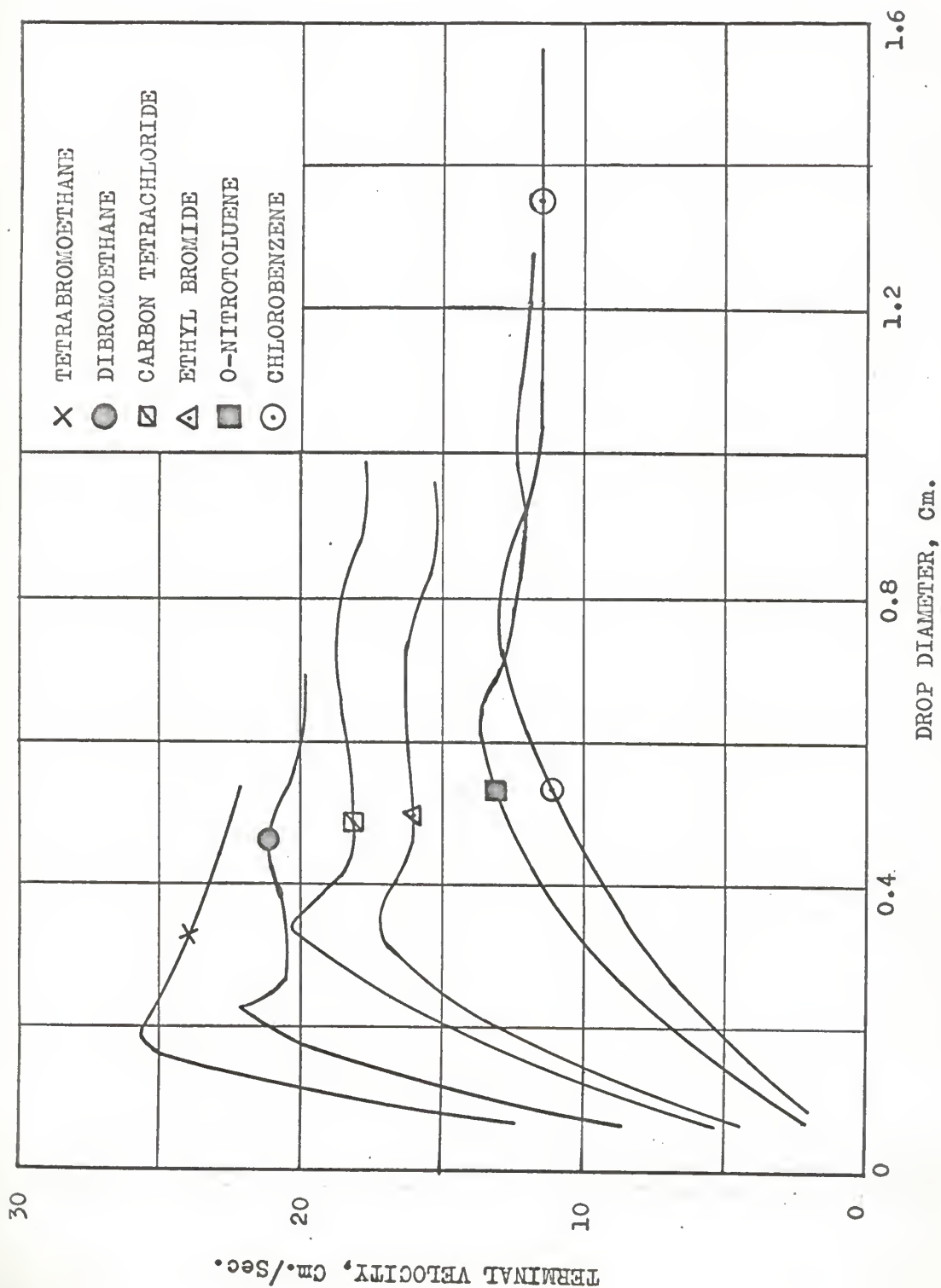


FIG. 1. VARIATION OF TERMINAL VELOCITY WITH DROP SIZE (DATA OF HU AND KINTNER)

Table 1. Physical Properties: Data of Licht and Narasimham-
urty. $\rho = 0.9971$ g/cc.; $\mu = 0.003937$ poises; $T = 25 \pm 0.1^\circ\text{C}$.

Drop Liquid	: ρ_c	: $\Delta\rho$: μ_o	: σ_i	: F
Carbon tetra- chloride	1.5842	0.5871	0.9296	41.6	$(1.95)10^{10}$
Tetrachlorethane	1.5313	0.5342	1.502	31.3	$(0.835)10^{10}$
O-Nitrotoluene	1.1573	0.1607	2.093	26.6	$(1.86)10^{10}$
Tetrabromoethane	2.9539	1.9568	9.464	36.2	$(0.385)10^{10}$
Ethylchloroacetate	1.1451	0.1480	1.101	14.6	$(3.34)10^9$
Chlorobenzene	1.1008	0.1037	0.7625	35.4	$(6.81)10^{10}$

ρ = density, g./ml.

μ = viscosity, centipoises

σ_i = interfacial tension, dynes/cm.

Sub o = organic phase

Others = continuous phase

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

Table 2. Carbon tetrachloride--water

No. :	Drop : Drop	Re :	C_D :	We :	$Re/F^{0.15}$:	$C_D We F^{0.15}$:
:	Diameter: Velocity :	:	:	:	:	:
:	d : U_c	:	:	:	:	:
:	(cm.) : (cm/sec)	:	:	:	:	:
1	0.202 16.1	362	0.6	1.25	10.37	26.2
2	0.217 16.5	400	0.614	1.42	11.46	30.4
3	0.234 18.6	435	0.521	1.94	13.90	35.3
4	0.240 18.3	490	0.552	1.93	14.05	37.2
5	0.266 19.9	590	0.517	2.53	16.9	45.7
6	0.304 21.7	735	0.497	3.43	21.1	59.5
7	0.328 22.3	875	0.509	3.92	23.4	69.5
8	0.331 22.4	825	0.508	3.98	23.65	71.5
9	0.355 22.2	880	0.555	4.20	25.2	81.4
10	0.367 22.3	912	0.568	4.38	26.2	86.9
11	0.386 22.1	950	0.608	4.52	27.2	96.7
12	0.405 22.0	993	0.645	4.70	28.4	106.0
13	0.451 21.3	1070	0.755	4.91	30.6	129.0
14	0.503 20.2	1145	0.96	4.97	32.8	166.4
15	0.534 20.1	1195	1.018	5.17	34.2	183.5
16	0.565 19.7	1240	1.12	5.26	35.5	206.0
17	0.581 19.6	1270	1.16	5.35	36.4	217
18	0.605 19.5	1316	1.22	5.52	37.7	235

Table 3. O-Nitrotoluene--water

No. :	Drop : Drop	Re :	C_D :	We :	$Re/F^{0.15}$:	$C_D We F^{0.15}$:
:	Diameter: Velocity :	:	:	:	:	:
:	d : U_c	:	:	:	:	:
:	(cm.) : (cm/sec)	:	:	:	:	:
1	0.224 7.52	188	0.834	0.475	5.44	13.7
2	0.231 7.75	200	0.81	0.52	5.78	14.56
3	0.257 8.46	243	0.754	0.69	7.03	18.0
4	0.375 11.0	460	0.652	1.7	13.3	38.3
5	0.396 11.3	499	0.653	1.9	14.4	42.9
6	0.408 11.7	534	0.628	2.1	15.4	45.7
7	0.448 12.3	615	0.624	2.54	17.8	54.8
8	0.468 12.6	658	0.621	2.79	19.04	60.0
9	0.489 13.0	709	0.621	3.1	20.5	66.6
10	0.555 12.4	767	0.76	3.2	22.2	84.0
11	0.599 12.5	835	0.806	3.51	24.2	97.9
12	0.631 12.3	866	0.88	3.58	25.1	109.0
13	0.649 12.3	890	0.904	3.68	25.8	115.0
14	0.729 12.7	1030	0.952	4.4	29.8	144.7
15	0.750 12.7	1060	0.979	4.53	30.7	153.4
16	0.800 12.2	1087	1.13	4.46	31.4	174.4
17	0.856 11.8	1127	1.295	4.47	32.6	200.0
18	0.893 12.1	1204	1.285	4.9	34.8	218.0
19	0.935 12.0	1250	1.37	5.05	36.2	239.0

Table 4. Tetrabromoethane--water

No. :	Drop : Diameter	Drop : Velocity	Re :	C_D :	We :	$Re/F^{0.15}$:	$C_{DWe}F^{0.15}$:
:	d	U_0	:	:	:	:	:
:	(cm.)	(cm/sec)	:	:	:	:	:
1	0.114	20.1	255	0.72	1.27	9.3	25.1
2	0.132	22.5	332	0.67	1.84	12.1	33.8
3	0.149	24.4	405	0.64	2.44	14.8	42.8
4	0.169	26.4	497	0.62	3.12	18.1	53.0
5	0.177	27.0	532	0.622	3.55	19.4	60.5
6	0.185	27.8	574	0.612	3.93	21.0	65.9
7	0.202	29.0	652	0.615	4.67	23.8	78.6
8	0.207	29.5	681	0.608	4.95	24.9	82.5
9	0.219	29.3	715	0.652	5.17	26.1	92.4
10	0.229	28.8	735	0.707	5.23	26.3	101.3
11	0.240	27.7	741	0.80	5.06	27.1	111.0
12	0.264	27.1	797	0.92	5.33	29.1	134.2
13	0.268	27.1	810	0.935	5.42	29.6	138.8
14	0.280	27.0	832	0.984	5.62	30.4	151.2
15	0.308	25.2	865	1.24	5.38	31.6	183.0
16	0.360	25.0	1002	1.47	6.15	36.6	248.0

Table 5. Ethyl chloroacetate--water

No. :	Drop : Diameter	Drop : Velocity	Re :	C_D :	We :	$Re/F^{0.15}$:	$C_{DWe}F^{0.15}$:
:	d	U_0	:	:	:	:	:
:	(cm)	(cms/sec)	:	:	:	:	:
1	0.214	10.0	238	0.415	1.46	9.2	16.3
2	0.230	11.1	284	0.366	1.94	11.0	19.0
3	0.266	12.9	392	0.31	3.02	15.2	25.0
4	0.297	13.6	444	0.312	3.75	17.2	31.4
5	0.319	13.6	485	0.334	4.03	18.8	36.1
6	0.330	13.7	504	0.342	4.23	19.5	39.0
7	0.349	13.5	526	0.372	4.35	20.4	43.5
8	0.383	13.1	560	0.435	4.49	21.7	52.5
9	0.392	13.2	577	0.436	4.66	22.4	54.5
10	0.415	12.6	583	0.507	4.50	22.6	61.2
11	0.444	12.3	608	0.57	4.6	23.6	70.4
12	0.505	11.9	670	0.692	4.88	26.0	90.7
13	0.574	11.4	730	0.855	5.1	28.3	117.0
14	0.640	10.9	778	1.045	5.2	30.2	146.0
15	0.665	10.9	807	1.085	5.4	31.3	158.0

Table 6. Chlorobenzene--water

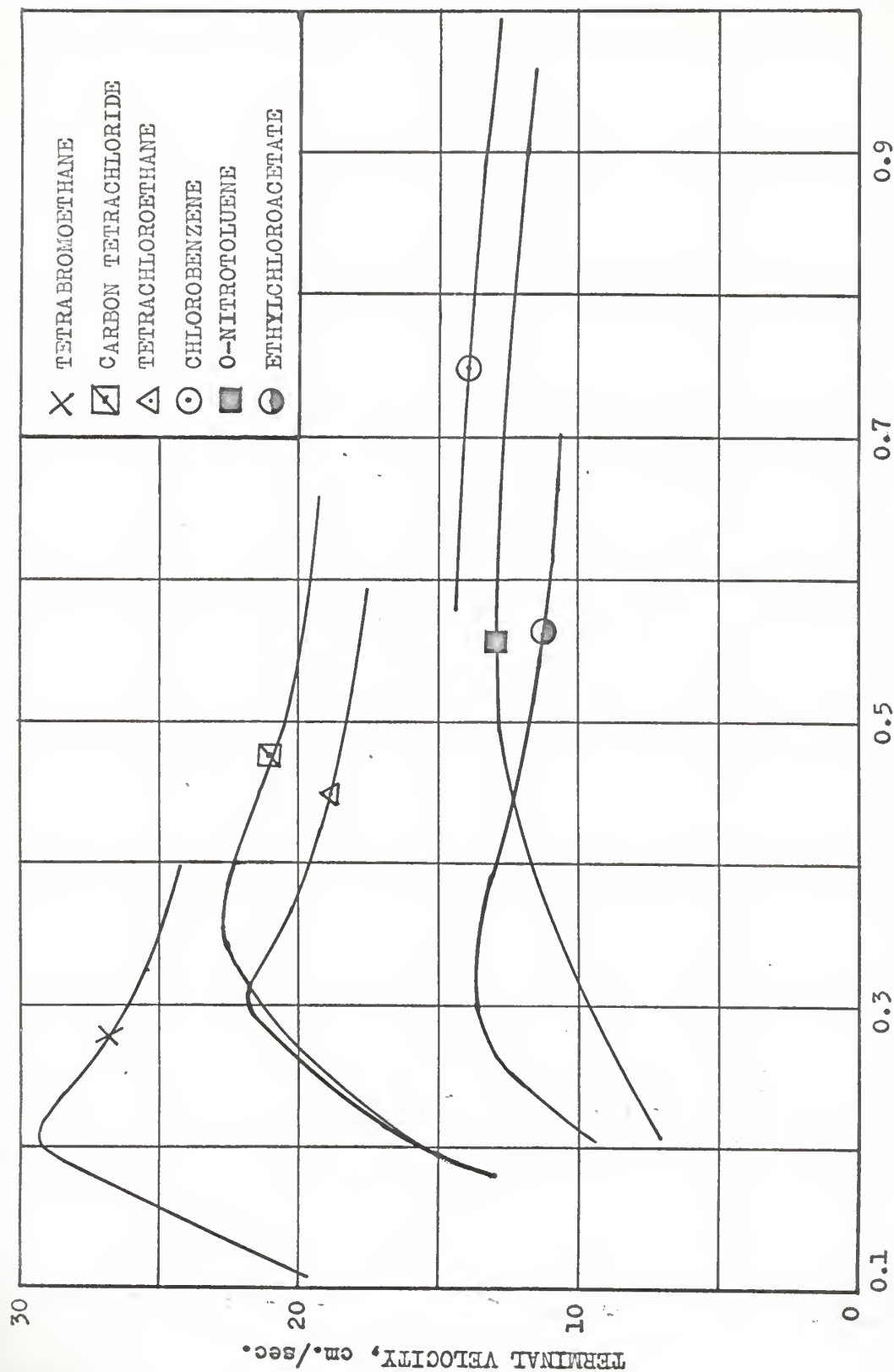
No.	Drop : Diameter : d : (cm.)	Drop : Velocity : U_c : (cm/sec)	Re	C_D	We	$Re/F^{0.15}$	$C_D We F^{0.15}$
1	0.596	14.5	964	0.427	3.54	22.8	63.7
2	0.642	14.3	1024	0.427	3.70	24.3	66.6
3	0.645	14.3	1030	0.429	3.72	24.4	67.3
4	0.636	14.2	1036	0.468	3.90	25.3	77.0
5	0.695	14.0	1035	0.482	3.84	25.8	78.0
6	0.772	13.8	1190	0.552	3.97	28.2	92.5
7	0.777	14.1	1220	0.531	4.35	28.9	97.5
8	0.836	13.5	1335	0.661	4.56	31.7	127.2
9	0.990	12.7	1400	0.834	4.50	33.2	153.0
10	0.991	12.9	1426	0.810	4.65	33.8	158.7
11	1.119	12.7	1530	0.943	5.08	37.4	202.0

Table 7. Comparison of observed and calculated peak velocities and corresponding drop diameters.

System	$(U_o)_p$: obs. : (cm/sec)	$(d)_p$: obs. : (cm.)	F	$(U_o)_p$: calc. : cm/sec	$(d)_p$: calc. : cm.	Dev. : d%	Dev. : U_o %
Carbon Tetra- chloride	22.75	0.355	1.95×10^{10}	21.6	0.320	-9.85	-5.28
Tetra- chloro- ethane	21.8	0.31	0.835×10^{10}	18.9	0.314	1.33	-13.3
C-Nitro- toluene	12.9	0.492	1.86×10^{10}	13.25	0.545	10.8	2.71
Tetra- bromo- ethane	29.5	0.207	0.385×10^{10}	26.3	0.188	-9.17	-10.85
Ethyl- chloro- acetate	13.7	0.330	3.34×10^9	13.9	0.271	1.46	-18.2
Chloro- benzene	14.5	0.596	6.81×10^{10}	13.0	0.752	26.2	-10.34

$$(U_o)_p = 1.23 \left(\frac{61}{\mu} \right) F^{-0.238}$$

$$(d)_p = 3.58 \left(\frac{61}{\rho} \right) \frac{1}{(U_o)_p}$$



DROP DIAMETER, cm.

FIG. 2. VARIATION OF TERMINAL VELOCITY WITH DROP SIZE
(DATA OF LIGHT AND NARASIMHAMURTY)

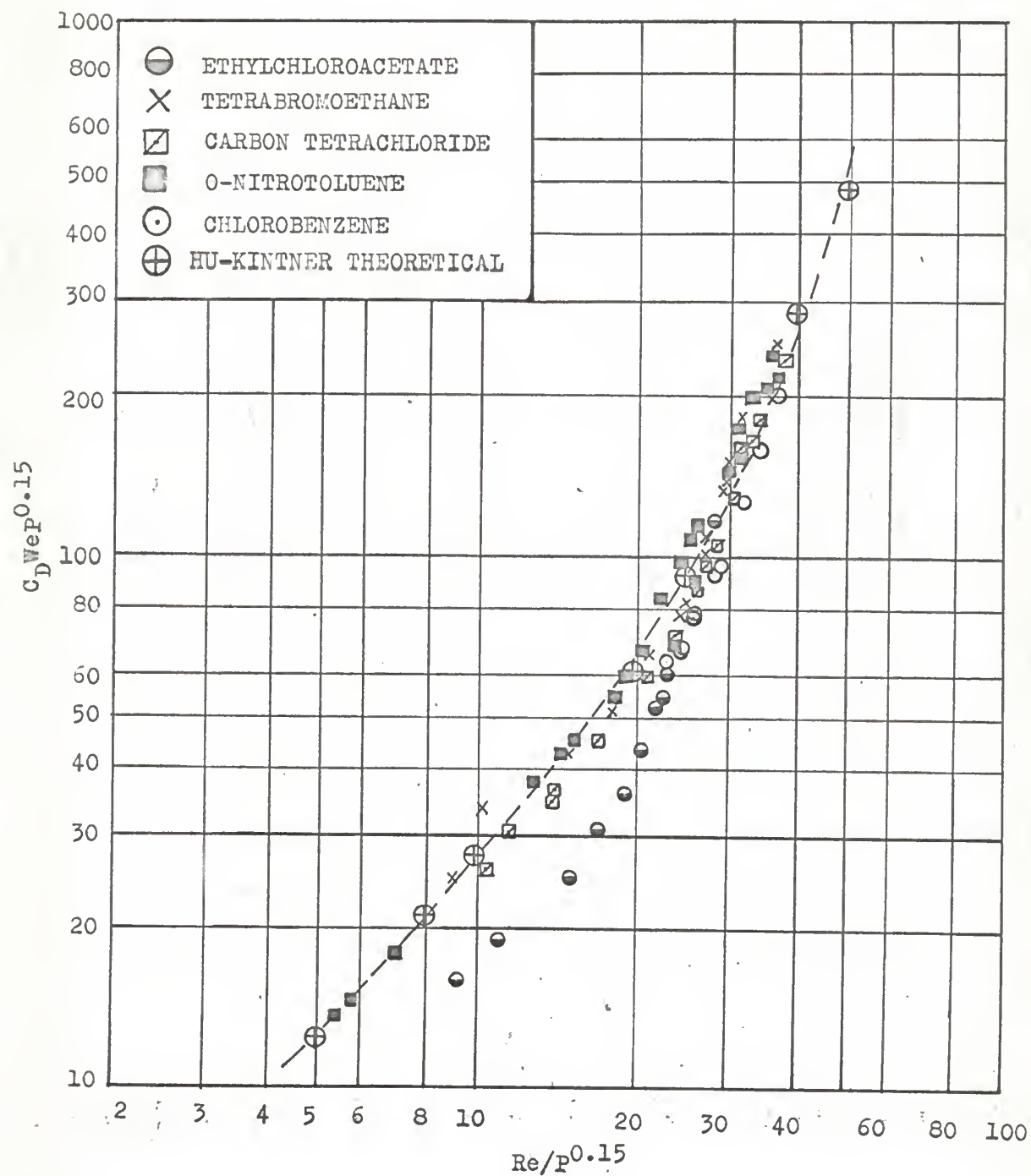


FIG. 3. PLOT OF DATA OF LICHT AND NARASIMHAMURTY USING HU-KINTNER CORRELATION.

unique curve for all systems except the aniline-water system. The failure of the latter to correlate may well be due to the fact that it has a very low interfacial tension as well as its susceptibility to oxidation in contact with air.

Data of Licht and Narasimhamurty (36) is shown graphically in Figure 2. Four of the liquids used by them were the same as those used by Hu and Kintner (33). The curves of Licht and Narasimhamurty do not exhibit sharp peaks as those exhibited by the curves of Hu and Kintner. Also a comparison of the two sets of data indicates that the velocities obtained by the former workers were higher than those obtained by the latter in each of these four cases. The differences amount to roughly 10 per cent. This indicates that their drops began to circulate internally at a smaller drop size than usual. This may well have been due to the relatively high purity of their materials.

A plot of the data of Licht and Narasimhamurty using the Hu-Kintner correlation, i.e., a plot of $C_D We_F^{0.15}$ versus $Re/\rho^{0.15}$ on log-log coordinates, is shown in Figure 3. In spite of a small scatter it appears that their data, with the exception of ethylchloroacetate-water system, can be well correlated by the Hu-Kintner correlation. The scatter might be due to the fact that on the whole the velocity-drop diameter data is not as smooth and shows more fluctuations than those of Hu and Kintner. The failure of the ethylchloroacetate-water system to correlate may very well be due to a faulty value of the interfacial tension. It may be noted that Warshay, Bogusz, Johnson and Kintner

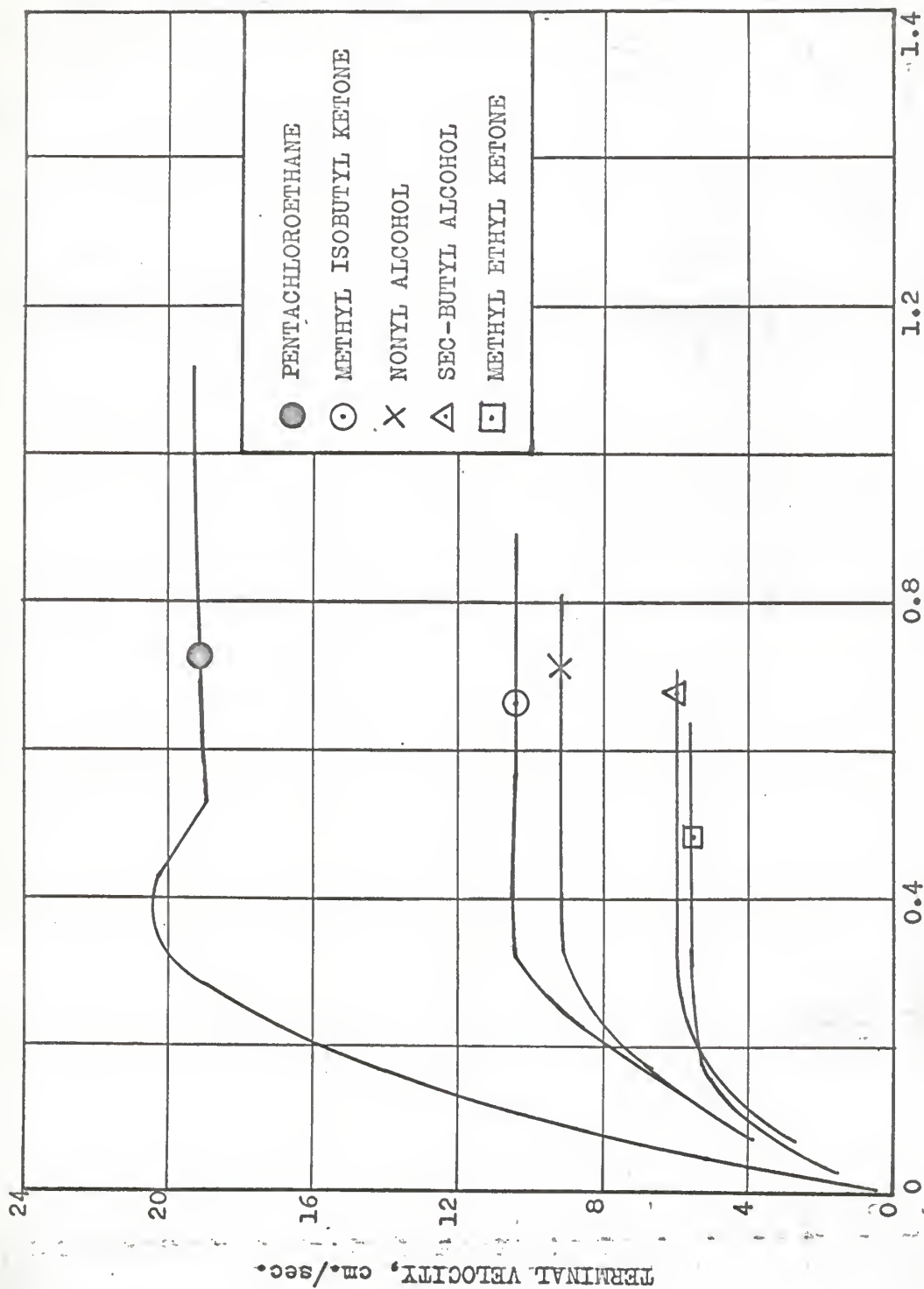


FIG. 4. VARIATION OF TERMINAL VELOCITY WITH DROP SIZE
(DATA OF KLEE AND TREYBAL)

(65), for the same system, reported a value of 16.43 dynes/cm. whereas the value used by Licht and Narasimhamurty was 14.6 dynes/cm.

The comparison of predicted values of peak terminal velocity and the diameter corresponding to this peak velocity with the observed values (Table 7) indicate rather poor agreement and this can be explained in view of the fact that the observed velocities of Licht and Narasimhamurty were approximately 10 per cent in excess of those reported by Hu and Kintner.

Licht and Narasimhamurty offered the following correlation:

$$C_r = \left(\frac{C_D}{C_D^S} \right)_{Re} = 0.87\beta + 0.425 \quad (8)$$

where β is defined as in equation (8). This correlation appears to be less useful and less general since it failed to bring all their data on one single line. When C_D/C_D^S was plotted as ordinate versus β as abscissae, a series of stright lines were obtained and equation (3) above represents the mean of all the six curves.

The terminal velocity-drop diameter data of Klee and Treybal (29) is shown on Figure 4. They offered two different correlations for each of two drop diameter-terminal velocity regions:

$$U_{OI} = 38.3 \rho^{0.45} \Delta \rho^{0.58} \mu^{-0.11} d^{0.70} \quad (11)$$

$$U_{OII} = 17.6 \rho^{-0.55} \Delta \rho^{0.28} \mu^{0.10} d^{0.18} \quad (12)$$

Since they claimed that these two correlations were superior to the Hu-Kintner correlation for low interfacial tension systems, the data on five of their systems were plotted using the

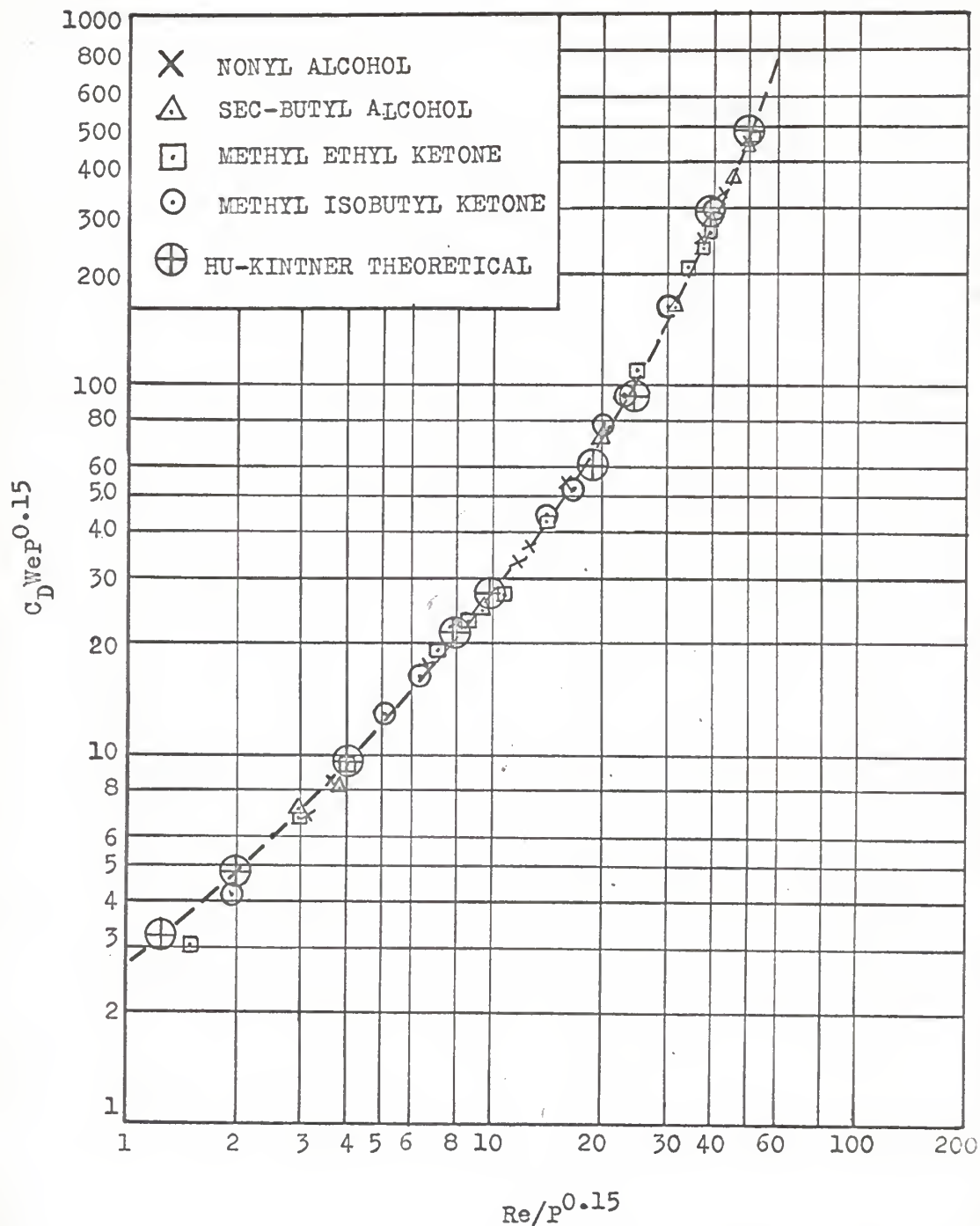


FIG. 5. PLOT OF DATA OF KLEE AND TREYBAL USING HU-KINTNER CORRELATION.

Hu-Kintner correlation as shown in Figure 5. Four of the systems chosen, nonyl alcohol, sec-butyl alcohol, methyl ethyl ketone and methyl isobutyl ketone had interfacial tensions of less than ten dynes/cm. Fentachloroethane-water system was chosen for comparison.

A Hu-Kintner plot of their data shows excellent agreement for all the systems studied. This clearly indicates that the Hu-Kintner correlation applies equally well to systems of extremely low interfacial tensions as well. The latter can be put, with slight modifications, in the same form as the equations offered by Klee and Treybal (equations 9 and 10), but with different constants. This indicates that their correlations are a special case of the Hu-Kintner correlation. Also the peak velocity between the two regions is not accounted for. According to their computations, using their own equations and the data of Hu and Kintner, resulted in rather poor agreement in Region I (15.2 per cent average deviation) where the velocity is increasing with drop size (29).

The materials used by Klee and Treybal were of commercial grade, and no attempt was made to purify them. Also, their equations predict a velocity fully independent of drop size while the data of Hu and Kintner (23) and Licht and Narasimhamurti (36) indicate otherwise.

Comparison of calculated values of peak velocity and the drop diameter corresponding to this peak velocity using relations offered by Hu and Kintner, with those observed (Table 13) show poor agreement particularly for low interfacial systems. This

Table 6. Physical properties: Data of Klee and Trevbal.

Drop liquid	ρ_o	ρ	$\Delta\rho$	μ_o	μ	σ_i	F	$T^{\circ}\text{C.}$
Nonyl alcohol	0.8242	0.9982	0.1740	0.162	0.0100	4.9	6.87×10^7	20
Sec-butyl alcohol	0.8660	0.9705	0.1045	0.0278	0.0156	0.6	3.34×10^4	28
Methyl ethyl ketone	0.8370	0.960	0.1230	0.0060	0.0145	0.3	4.67×10^3	24
Methyl iso-butyl ketone	0.8155	0.9947	0.1792	0.0060	0.0093	9.8	7.24×10^8	20
Penta-chloro-ethane	1.6740	0.9978	0.9978	0.0203	0.0095	42.4	1.355×10^{10}	25

ρ = density, gm/cc.

μ = viscosity, poises

σ_i = interfacial tension, dynes/cm.

Sub o = organic phase

others = continuous phase

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

Table 9. Acetyl alcohol--water

No.	Drop : Diameter : d : (cm.)	Drop : Velocity : U_0 : (cm/sec)	Re	C_D	We	$Re/F^{0.15}$	$C_{DWe} F^{0.15}$
1	0.10	4.7	47	1.03	0.45	3.14	6.93
2	0.11	5.0	55	1.003	0.56	3.63	8.4
3	0.16	6.2	99	0.95	1.25	6.62	17.73
4	0.18	6.7	120	0.915	1.65	8.02	22.6
5	0.22	8.0	176	0.784	2.87	11.77	33.6
6	0.23	8.3	190	0.761	3.26	12.7	37.1
7	0.28	9.1	240	0.864	4.22	16.05	54.5
8	0.33	9.1	300	0.906	5.57	20.05	75.5
9	0.61	9.1	554	1.68	10.1	37.0	254.0
10	0.69	9.2	634	1.86	11.9	42.4	331.0
11	0.81	9.2	743	2.18	14.0	49.6	456.0

Table 10. Sec-butyl alcohol--water

No.	Drop : Diameter : d : (cm.)	Drop : Velocity : U_0 : (cm/sec)	Re	C_D	We	$Re/F^{0.15}$	$C_{DWe} F^{0.15}$
1	0.08	2.9	14.4	1.31	1.09	3.02	6.96
2	0.09	3.2	19.0	1.24	1.49	3.73	8.21
3	0.15	4.6	45.8	1.0	5.14	9.52	24.5
4	0.15	5.0	46.7	0.846	6.08	9.8	24.6
5	0.26	5.8	94	1.09	14.2	19.7	73.0
6	0.39	6.0	146	1.53	22.7	30.6	166.0
7	0.50	6.0	187	1.96	29.2	39.2	273.0
8	0.58	5.9	213	2.35	32.7	44.7	363.0
9	0.64	6.0	240	2.51	37.3	50.3	446.0

Table 11. Methyl ethyl ketone--water

No.	Drop : Diameter : d : (cm)	Drop : Velocity : U_0 : (cm/sec)	Re	C_D	We	$Re/F^{0.15}$	$C_{DWe}F^{0.15}$
1	0.04	2.0	5.3	1.67	0.51	1.49	3.03
2	0.06	2.6	10.3	1.49	1.3	2.9	6.9
3	0.07	3.1	14.4	1.22	2.15	4.05	9.35
4	0.10	3.3	25.2	1.16	4.62	7.08	19.1
5	0.11	4.2	30.6	1.04	6.2	8.6	23.0
6	0.12	4.8	33.2	0.87	8.35	10.7	27.4
7	0.15	5.0	49.7	1.00	12.0	13.95	42.7
8	0.24	5.5	87.5	1.33	23.2	24.6	110.0
9	0.33	5.6	122	1.76	33.1	34.2	207.0
10	0.35	5.6	130	1.37	35.1	36.6	234.0
11	0.37	5.5	135	2.05	35.3	37.9	261.0
12	0.49	5.6	182	2.62	49.1	51.2	458.0

Table 12. Methyl isobutyl ketone--water

No.	Drop : Diameter : d : (cm)	Drop : Velocity : U_0 : (cm/sec)	Re	C_D	We	$Re/F^{0.15}$	$C_{DWe}F^{0.15}$
1	0.09	4.3	42	1.15	0.17	1.97	4.16
2	0.12	5.4	69	0.97	0.36	3.24	7.44
3	0.16	6.3	108	0.95	0.64	5.06	12.96
4	0.18	7.0	135	0.87	0.89	6.34	16.5
5	0.22	9.3	219	0.60	1.93	10.3	24.6
6	0.23	9.8	294	0.69	3.0	13.3	44.1
7	0.32	10.4	356	0.70	3.52	16.7	52.5
8	0.39	10.4	434	0.85	4.36	20.4	79.0
9	0.43	10.6	488	0.90	4.90	22.9	94.0
10	0.57	10.4	645	1.24	6.25	30.3	164
11	0.67	10.4	745	1.46	7.36	35.0	229
12	0.77	10.4	856	1.68	8.45	40.2	302
13	0.86	10.4	955	1.88	9.45	44.8	378

Table 13. Comparison of observed and calculated peak velocities and corresponding drop diameters.

System	$(U_c)_p$	$(d)_p$	F	$(U_c)_p$	$(d)_p$	Dev.	Dev.
	: cbs.:	: cbs.:		: calc.:	: calc.:	: d %	: U_c
	: (cm/sec):	: (cm/sec):		: (cm/sec):	: (cm):		: %
Methyl alcohol	9.2	0.33	6.87×10^7	8.2	0.26	-20.2	-10.9
Sec-butyl alcohol	6.0	0.34	3.34×10^4	4.0	0.14	-53.8	-33.3
Methyl ethyl ketone	5.6	0.24	4.67×10^3	3.4	0.13	-45.8	-39.3
Methyl isobutyl ketone	10.5	0.37	7.24×10^8	10.1	0.35	-5.4	-3.8
Penta-chloro-ethane	20.3	0.36	1.355×10^{10}	21.4	0.33	-8.3	0.5

$$(U_c)_p = 1.23 \left(\frac{\sigma_i}{\mu} \right) F^{-0.238}$$

$$(d)_p = 3.58 \left(\frac{\sigma_i}{\rho} \right) \frac{1}{(U_c)_p^2}$$

Table 14. Physical properties: data of Johnson and Braida.
 $\rho = 1.0685$ gms/cc.; $\mu = 0.01974$ poises; $\mu_w = 0.003927$ poises; $T = 25^\circ\text{C}$.

liquid	ρ_c	$\Delta\rho$	σ_i	F
Carbon tetra- chloride	1.5857	0.5172	44.26	1.286×10^9
Bromobenzene	1.4900	0.4215	34.60	7.54×10^8
Ethyl bromide	0.4445	0.3760	27.36	4.18×10^8
Nitrobenzene	1.1980	0.1295	21.61	5.98×10^8
O-Nitrotoluene	1.1583	0.0898	24.39	1.24×10^9

ρ = density, gm./cc.

μ = viscosity, poises

σ_i = interfacial tension, dynes/cm.

Sub c = organic phase

Sub w = water phase

others = continuous phase

$$F = \frac{613 \rho^2}{g \mu^4 \Delta\rho}$$

Table 15. Carbon tetrachloride--23.5% glycerol

No.	: Drop	: Drop	: Re	: C_D	: We	: $\frac{Re}{F^{0.15}}$: $C_D We$: $C_D We F^{0.15}$
	: Diam.	: Vel.	:	:	:	: $F^{0.15}$: $F^{0.15}$: $(\mu/\mu_w)^{0.14}$
	: d	: U_0	:	:	:	:	:	:
	: (cm)	: (cm/sec)	:	:	:	:	:	:
1	0.125	6.3	42.5	1.99	0.12	1.825	5.56	4.93
2	0.151	8.1	66	1.45	0.198	2.84	6.69	5.99
3	0.175	9.2	87	1.31	0.357	3.74	10.9	9.75
4	0.19	10.0	103	1.2	0.457	4.42	12.3	11.45
5	0.27	15.3	230	0.68	1.62	9.86	25.6	22.9
6	0.28	16.3	246	0.67	1.79	10.55	27.9	25.0
7	0.32	16.8	290	0.76	2.13	12.45	33.6	34.6
8	0.35	17.5	331	0.72	2.58	14.2	43.3	38.8
9	0.38	18.3	376	0.72	3.07	16.3	51.5	46.1
10	0.43	18.8	436	0.76	3.47	18.7	61.5	55.0
11	0.475	18.5	475	0.79	3.92	20.4	72.1	64.6
12	0.52	17.8	500	1.04	3.97	21.5	96.2	86.1

Table 16. Brombenzene--23.5% glycerol

No.	: Drop	: Drop	: Re	: C_D	: We	: $\frac{Re}{F^{0.15}}$: $C_D We$: $C_D We F^{0.15}$
	: Diam.	: Vel.	:	:	:	: $F^{0.15}$: $F^{0.15}$: $(\mu/\mu_w)^{0.14}$
	: d	: U_0	:	:	:	:	:	:
	: (cm)	: (cm/sec)	:	:	:	:	:	:
1	0.125	5.5	37	2.13	0.144	1.73	6.55	5.86
2	0.16	7.2	62	1.59	0.315	2.9	10.7	9.59
3	0.175	8.1	76	1.37	0.436	3.56	12.8	11.46
4	0.20	9.0	97	1.27	0.615	4.53	16.7	14.96
5	0.30	14.0	227	1.1	2.24	10.6	52.7	47.2
6	0.375	15.7	318	0.78	3.52	14.3	58.3	52.7
7	0.41	16.3	360	0.79	4.15	16.8	70.2	62.9
8	0.44	17.1	407	0.77	4.89	19.1	80.5	72.0
9	0.50	17.0	460	0.89	5.49	21.5	104.4	93.5
10	0.53	16.5	472	1.0	5.48	22.5	117	105
11	0.6	15.5	502	1.29	5.48	23.5	151	135
12	0.75	15.5	627	1.61	6.35	29.3	236	212

Table 17. Nitrobenzene--23.5% glycerol

No.	: Drop	: Drop	: Re	: C_D	: We	: $\frac{Re}{F^{0.15}}$: $\frac{C_D We}{F^{0.15}}$: $\frac{C_D We F^{0.15}}{(\mu/\mu_w)^{0.14}}$
	: Diam.	: Vel.						
	: d	: U_c						
	: (cm)	: (cm/sec)						
1	0.41	9.8	217	0.676	1.95	10.5	27.2	24.4
2	0.52	10.6	298	0.734	2.89	14.5	43.7	39.2
3	0.55	11.0	327	0.72	3.3	15.9	49.0	43.9
4	0.60	12.0	389	0.66	4.23	18.9	53.2	52.1
5	0.67	12.5	452	0.68	5.13	21.9	72.5	65.0
6	0.725	12.3	482	0.76	5.43	23.4	85.0	76.1
7	0.825	11.5	512	0.99	5.4	24.3	110.0	93.5
8	0.86	11.5	534	1.03	5.63	25.9	119.5	107
9	0.90	11.6	564	1.06	6.0	27.4	131.0	117.3

Table 18. C-Nitrotoluene--23.5% glycerol

No.	: Drop	: Drop	: Re	: C_D	: We	: $\frac{Re}{F^{0.15}}$: $\frac{C_D We}{F^{0.15}}$: $\frac{C_D We F^{0.15}}{(\mu/\mu_w)^{0.14}}$
	: Diam.	: Vel.						
	: d	: U_c						
	: (cm)	: (cm/sec)						
1	0.49	9.4	248	0.61	1.89	10.7	26.7	23.0
2	0.58	9.8	307	0.665	2.44	13.25	37.5	33.6
3	0.65	11.0	386	0.59	3.44	16.65	47.0	42.1
4	0.68	12.0	440	0.52	4.29	19.0	51.7	45.3
5	0.74	12.0	430	0.565	4.66	20.7	61.0	54.6
6	0.82	11.9	527	0.64	5.08	22.7	75.3	67.5
7	0.90	11.0	535	0.82	4.77	23.0	90.5	81.0

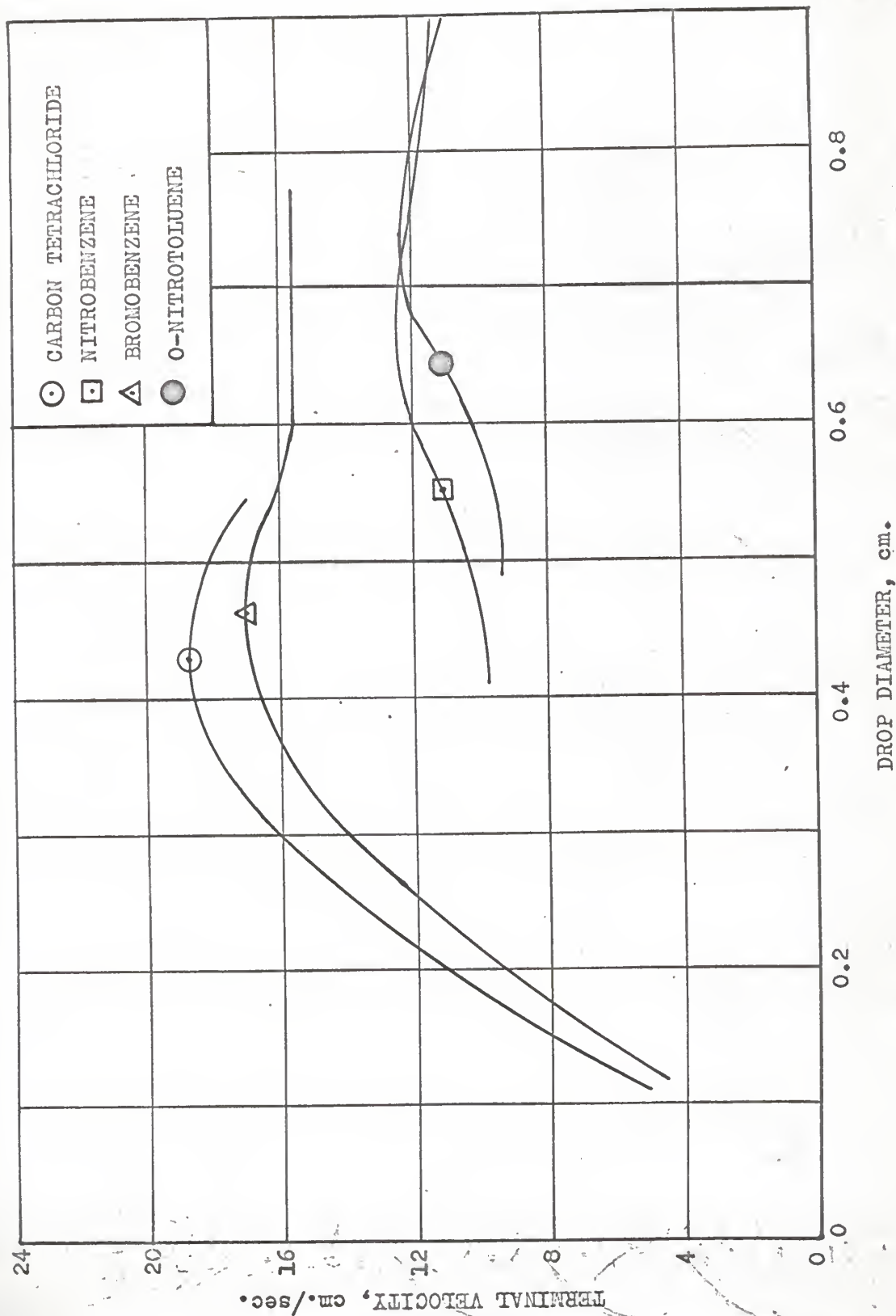


FIG. 6. VARIATION OF TERMINAL VELOCITY WITH DROP SIZE IN AQUEOUS 28.5% GLYCEROL SOLUTION (DATA OF JOHNSON AND BRAIDA)

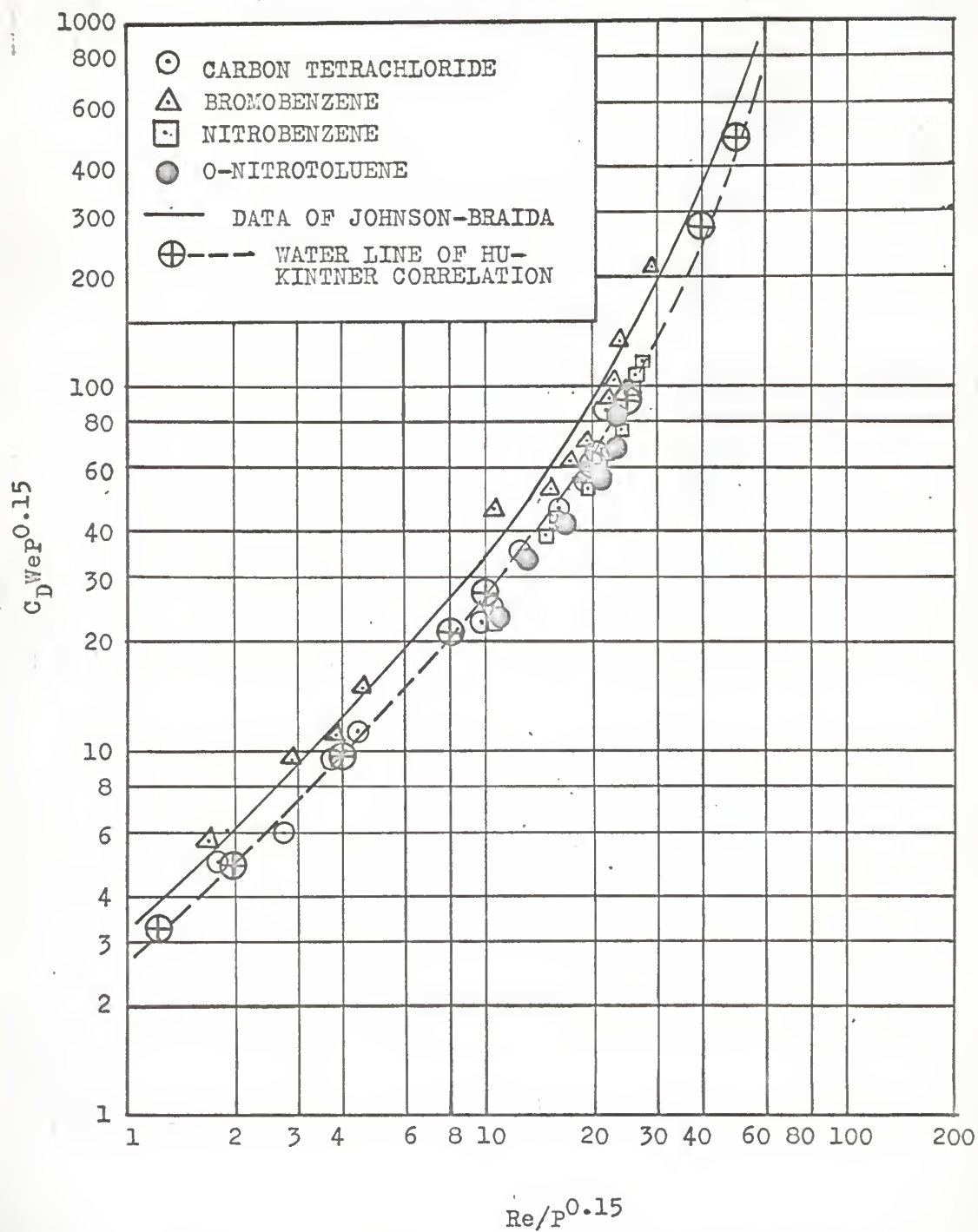


FIG. 7. PLOT OF DATA OF JOHNSON AND BRAIDA USING HU-KINTNER CORRELATION.

is not surprising in view of the fact that expressions for predicting peak velocity and the corresponding diameter were developed from data obtained on high interfacial tension systems. Agreement is good for methyl isobutyl ketone-water ($\sigma_i = 9.8$ dynes/cm) and pentachloroethane-water ($\sigma_i = 42.4$ dynes/cm) systems, as would be expected. Two of the other three systems had interfacial tensions of less than one dyne per cm.

The velocity-drop diameter data obtained by Johnson and Braida (26) on four liquids, using a 23.5 per cent glycerol solution in water as the continuous phase, is shown graphically in Figure 6. These curves also do not exhibit sharp peaks and are similar in this respect to those of Licht and Narasimhamurti (36) and Klee and Treybal (29). A peakless curve is formed in systems which have a surface active agent present. However, because a peakless curve is formed it does not necessarily follow that a surface-active agent is involved.

A Hu-Kintner plot of the data is shown in Figure 7. The solid line indicates the mean line through all the data points. The water line, which is the theoretical Hu-Kintner line for drops falling in water is shown dashed. A correlation can be obtained by shifting the former curve to the water line. This can be done by dividing the ordinates by $(\mu/\mu_w)^{0.14}$ and plotting against $\log Re/F^{0.15}$ as usual. The entire data points are thus shifted down and can be represented by the dashed theoretical line, thus giving a correlation of the data. This indicates applicability of the Hu-Kintner correlation to systems that have viscosities higher than that of water, though not excessively

high.

CONCLUSION

The terminal velocities for the data of Licht and Narasimhamurty (36) were found to be higher than those reported by Hu and Kintner (33) for the same systems. This indicates the presence of internal circulation in the drops studied by the former workers at a drop size smaller than usual. For the data of all the three workers (26, 29, 36) studied, the Hu-Kintner correlation was found to give good correlation of the data; in the case of the data of Johnson and Braida (26), the ordinate has to be divided by a viscosity correction factor in order to make the data fall on the water line. The correlations developed by Klee and Treybal (29) were found to be special cases of the Hu-Kintner correlation. The latter gave a good correlation even for systems of interfacial tensions less than one dyne per cm.

The comparison of predicted values of peak terminal velocities and the corresponding drop diameters showed poor agreement for the data of Licht and Narasimhamurty (36). This is due to the fact that their reported values for the fall velocities were higher than those reported by other workers. In the case of the data of Klee and Treybal (29) agreement between predicted and observed values was found to be poor for low interfacial tension systems, and good for systems of higher interfacial tensions.

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LIST OF SYMBOLS

A	Total cross section of tower
a	Constant
C_D	Drag coefficient, $\frac{4}{3} \cdot \frac{\Delta f}{f} \cdot \frac{dg}{U_0^2}$
C_D^S	Drag coefficient of a rigid sphere
d	Droplet equivalent diameter, cm.
D	Column internal diameter, cm.
e	Base of natural logarithm
F	Drag force, dynes
Fr	Froude number, U_0^2/dg
g	Gravitational acceleration, cm./sec ²
K_1	Correction factor for fluidity of moving body
K_w	Wall correction factor, U_0/u
N	N group, $g\mu^4/\rho\sigma i^3$
Q_d	Superficial volumetric dispersed phase flow rate
Q_f	Superficial volumetric continuous phase flow rate
Re	Reynolds number, dU_0^2/μ
S	Distance of fall required for reaching terminal velocity when starting from rest, cm.
t	Time, sec.
T	Temperature, °C
u	Point velocity, cm./sec.
U_0	Terminal velocity with no wall effect, cm./sec.
V_s	Slip velocity, cm./sec.
V_t	Droplet terminal velocity, cm./sec.
We	Weber number, $U_0^2 d\rho/\sigma i$

LIST OF SYMBOLS
(Continued)

- Γ Physical property group, $\frac{\rho \epsilon_i^3}{g \mu^4} \frac{\rho}{\Delta \rho}$
- β Drop property group, $g d^2 \Delta \rho / 4 \epsilon_i$
- ϵ Void fraction
- $1-\epsilon$ Holdup of dispersed phase
- μ Viscosity of field fluid, poises
- μ_o Viscosity of organic phase, poises
- μ_w Viscosity of water, poises
- π 3.1416
- ρ Density of field fluid, gm./cc.
- ρ_o Density of organic phase, gm./cc.
- $\Delta \rho$ Density difference of a system, $(\rho_o - \rho)$
- ϵ_i Interfacial tension, dynes/cm.
- Subscripts:
- c Property at critical drop size
- p Property at peak velocity
- I Region where drop velocity increases with increasing drop diameter
- II Region where drop velocity remains constant with increasing drop diameter

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MECHANICS OF DROP MOTION IN STATIONARY
LIQUID MEDIA

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AN ABSTRACT OF MASTER'S REPORT

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Literature pertinent to the subject has been reviewed at some length and also various aspects of practical importance have been considered. Theoretically developed mathematical expressions for drop motion in liquid media are presented.

Correlations developed by four different authors have been compared and their relative merits discussed.

The data of Licht and Karasimhamurty (36), Klee and Treybal (29) and Johnson and Braida (26) have been recalculated using the Hu-Kintner correlation. Plots of $\log C_D We F^{0.15}$ versus $\log Re/F^{0.15}$ have been obtained in order to check whether different systems can be represented by one single curve.

Calculated and observed values of peak terminal velocities and the corresponding diameters have been compared so as to check the consistency of the empirical correlations relating the above variables with the physical properties of the systems.

By using a viscosity correction factor the data of Johnson and Braida (26), for systems with higher field fluid viscosity, have been made to coincide with the theoretical curve for systems with water as field fluid.