Rate of Rise or Fall of Liquid Drops

ALBERT J. KLEE and ROBERT E. TREYBAL

New York University, New York

Measurements of the terminal velocity of liquid drops immersed in an insoluble liquid were made for eleven liquid systems, covering a wide range of physical properties: continuous-phase density 0.960 to 1.145 g./cc., viscosity 0.93 to 1.56 centipoises; dispersed-phase density 0.807 to 1.674 g./cc., viscosity 0.59 to 72.1 centipoises; interfacial tension 0.3 to 42.4 dynes/cm. A correlation of the data was developed for each of the two velocity–drop-diameter regions which permits easy computation of the drop velocities. The correlations are shown to reproduce the present data and much of the published data very satisfactorily. Photographs permitted measurement of the drop eccentricities, which are shown to be a function of the drop diameter, density difference, and interfacial tension.

Several recent papers (*3, 5, 6*) have presented data on the rate of rise or fall of liquid droplets through another insoluble liquid. The present paper offers new data covering a wider range of liquid physical properties and a new correlation permitting estimates of the velocity of rise or fall which appears to be more general than the correlations offered earlier. The literature has been thoroughly reviewed in the papers mentioned and need not be considered in detail here.

EXPERIMENTAL DETAILS

The drops were allowed to rise or fall in a glass-walled tank of square cross section 10 by 10 in., 40 in. tall, made from ¼-in. plate glass and brass angles, with brass end plates, the whole kept leaktight with Permanite cement. The tank was filled with the liquid through which the drops were to move (continuous liquid). Liquid to form the drops (dispersed liquid) flowed by gravity from a separatory-funnel reservoir through two pipettes in series, of 1- and 5-ml. capacity, through a needle valve to control the rate of flow, and thence to a brass or stainless steel sharp-edged nozzle, from which it issued into the continuous liquid. The nozzles entered the tank through the upper or lower end plates, depending upon whether drops were to fall or rise. Nine nozzles were used, varying in internal diameter from 0.018 to 0.516 in., in order to obtain the desired range of drop sizes. The temperature of the system was measured by a thermometer suspended in a corner of the tank.

In this work the dispersed liquid issued slowly from each nozzle to form a drop which then grew to sufficient size to break away from the nozzle owing to its buoyancy. This is emphasized because, as will be pointed out later, the method of forming the drops may influence their velocity characteristics. The method used here simulates, to some extent at least, the mode of drop formation obtaining in many applications of liquid extraction. The volume of the drops produced at the nozzles was determined by counting the number of drops produced by a given volume of dispersed liquid, as measured by the change in level within one of the pipettes. For a given valve setting and nozzle, drops of uniform volume could be obtained in any desired number.

Table 1. Systems Studied

System	Continuous liquid	Dispersed liquid	Densities ρ_C	Densities ρ_D	Viscosities μ_C	Viscosities μ_D	Interfacial tension, σ	Temp., °C.
1	Water	Benzene	0.9975	0.8870	0.0114	0.0068	30.0	23
2	Water	Kerosene	0.9986	0.8071	0.0108	0.0147	40.4	18
3	Water	S.A.E. 10W oil	0.9975	0.8650	0.0106	0.721	18.5	23
4	Water	Nonyl alcohol	0.9982	0.8242	0.0100	0.162	4.9	20
5	Water	Sec-butyl alcohol	0.9705	0.8660	0.0156	0.0278	0.6	28
6	Furfural	Water	1.1450	1.0110	0.0134	0.0096	1.5	26.5
7	Water	Methyl ethyl ketone	0.960	0.8370	0.0145	0.0060	0.3	24
8	20% Aqueous sucrose	Benzene	1.0600	0.8720	0.0139	0.0059	30.1	26
9	Water	Methyl isobutyl ketone	0.9947	0.8155	0.0093	0.0060	9.8	20
10	Water	Pentachloroethane	0.9978	1.6740	0.0095	0.0203	42.4	25
11	Water	*n*-Heptylic acid	0.9980	0.9200	0.0095	0.0427	6.9	24.5

The time, as measured by a stopwatch, for passage of the drops between two levels 50 cm. apart marked on both sides of the tank permitted calculation of drop velocities. For each drop size the time was also measured separately for the first and second 25-cm. distances, to be certain that terminal velocities were reached.

Photographs of the drops were obtained in order to measure the deviation from spherical shape. Two electronic flash lamps were placed about 1 ft. from opposite sides of the tank to illuminate the drops from the sides. To eliminate reflections, all reflective parts of the apparatus were painted flat black. Pictures were made with a 35-mm. Kini Exakta V camera at *f*11, with either one or two 3+ diopter auxiliary lenses and a Polascreen. The shutter was synchronized with the flash lamps, and 1/1,000-sec. exposures were made.

The liquid systems used are listed in Table 1. Some technical-grade materials used were without special purification, as physical properties rather than system identity were considered to be of importance. The two liquids of each system were mutually saturated with each other before use, and the properties of the saturated phases determined at the temperature of the drop measurements. Densities were determined by pycnometer; viscosities by Ostwald-Cannon-Fenske viscometers or, in the case of the oil of system 3, by a Brookfield Synchro-lectric viscometer; and interfacial tension was determined by the drop-volume method (*2, 8*).

DROP VELOCITIES*

The appearance of the drops, i.e., shape, oscillation, etc., seemed generally to agree with that described elsewhere (*3, 6*). The velocity of fall or rise, as measured experimentally, is plotted against drop diameter for each system in Figure 1. In this figure, and elsewhere in this paper, drop diameter d is the diameter of a sphere of the same volume as the drop. As observed elsewhere (*1, 3, 5, 6*), the velocities for some systems pass through a maximum with increasing drop diameter and for others merely level off at some ultimately high value. Evidently the appearance of a maximum in the velocity-diameter curves depends upon the development of oscillations of the

*Detailed numerical data are available as document 4963 from the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., for $1.75 for 35-mm. microfilm or $2.50 for photoprints.

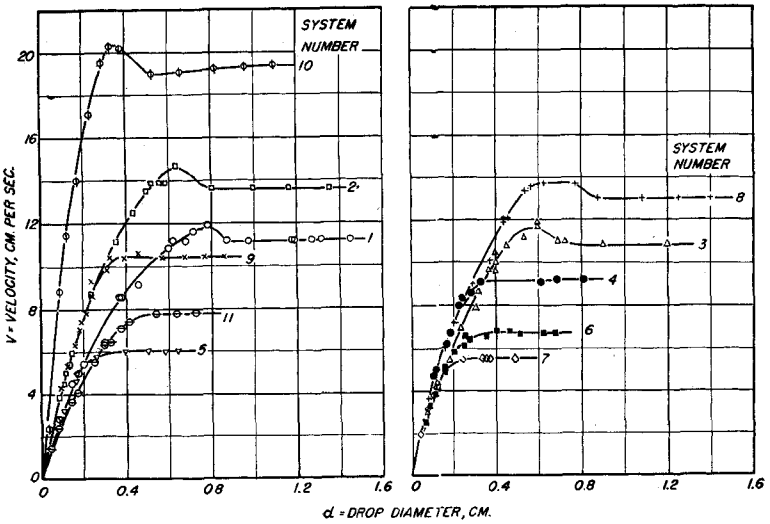

Fig. 1. Terminal velocities of liquid drops.

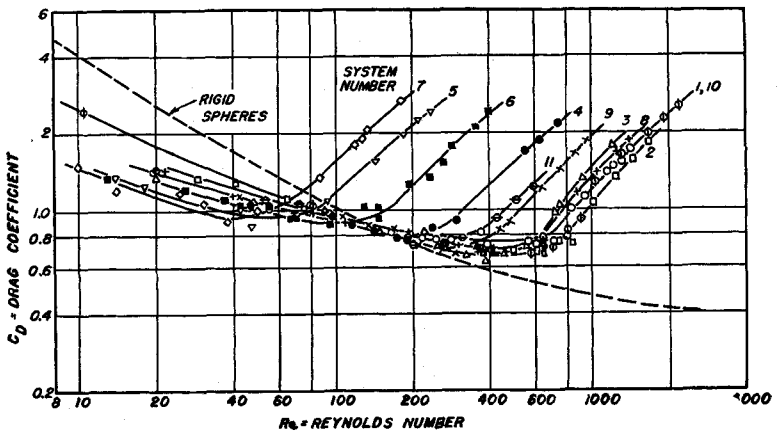

Fig. 2. Drag coefficients.

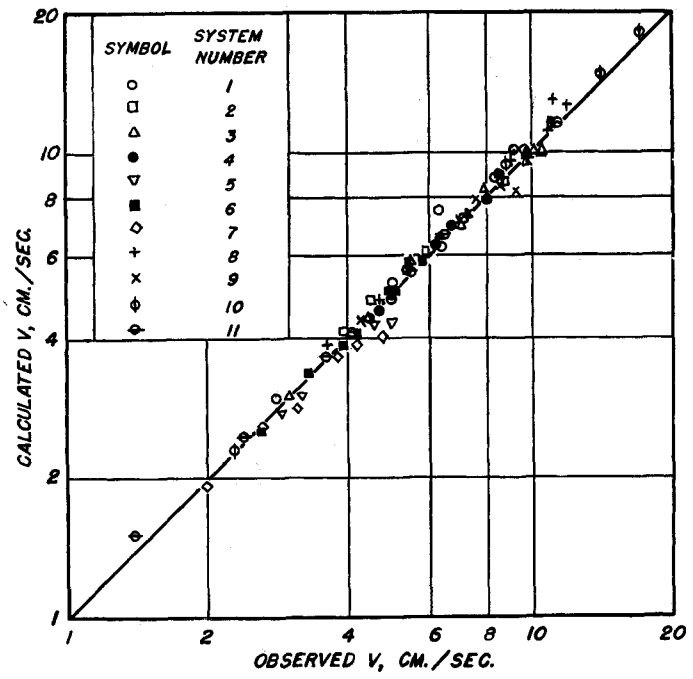

Fig. 3. Comparison of calculated and observed terminal velocities, region I.

drop (*1*), and for some systems this in turn can be dependent upon the manner in which the drops are formed as well as their physical properties.

When the velocity data are expressed in terms of drag coefficients, they give rise to the curves of Figure 2. At low Reynolds numbers the curves all fall below the curve for rigid spheres; whereas others (*3*, *6*) have shown data which lie both above and below. For a given system the position of the curves *evidently* will depend on the oscillations and internal circulation developed within the drop, since for nitrobenzene drops of various purities in water, Garner and Skelland (*1*) were able to develop drag coefficients varying by a factor of more than two at the same Reynolds number. From their work it would appear that interfacial tension is the property most likely to characterize this phenomenon.

The interfacial tensions of the systems corresponding to the curves at low Reynolds numbers, in order of decreasing departure from the curve for rigid spheres, are 42.4, 40.4, 9.8, 6.9, 30.1, 30.0, 18.5, 4.9, 1.5, 0.6, and 0.3 dynes/cm. These would appear roughly, at least, to confirm Garner and Skelland's observations that internal circulation, occurring in even small drops to an extent which decreases with increasing interfacial tension, increases the rate of rise or fall over that observed for rigid spheres. The interfacial tensions corresponding to the curves at high Reynolds numbers (reading from left to right, Figure 2) are 0.3, 0.6, 1.5, 4.9, 6.9, 9.8, 18.5, 30.0, 42.4, 40.4 dynes/cm. These Reynolds numbers correspond to large drop sizes, where the drop eccentricity, inversely proportional to interfacial tension as shown later, evidently has a greater influence on drop velocity, and in the reverse direction, than has internal circulation.

If it is assumed that

$$f(\mu_C, \mu_D, \rho_C, \Delta\rho, \sigma, d, V, g) = 0 \quad (1)$$

then dimensional analysis leads to

$$\left(\frac{\mu_C}{\mu_D}\right)^a \left(\frac{\Delta\rho}{\rho_c}\right)^b Re^c We^e C_D{}^h = m \quad (2)$$

No one set of values of the constants of Equation (2) was able to satisfy all the data. But if the velocity-diameter curves were considered 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, then suitable values of the constants could be obtained. By the method of least squares, with the data for all the measurements in each region, the equations obtained were for region I:

$$Re = 22.2 C_D{}^{-5.18} We^{-0.169} \quad (3)$$

and for region II:

$$Re = 0.00418 C_D{}^{2.91} We^{-1.81} \quad (4)$$

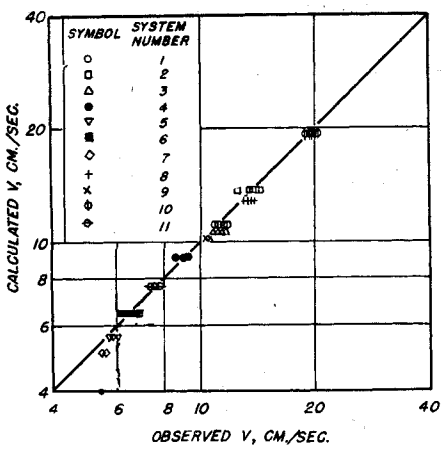

Fig. 4. Comparison of calculated and observed terminal velocities, region II.

mum velocities shown by some of the systems as indicated in Figure 1, but the errors thus introduced are small. Figures 3 and 4 compare the observed data with those calculated for the present systems by Equations (5), (6), and (7). The average deviation, including that incurred by use of Equation (7), is 4.15% in the case of region I and 2.82% in the case of region II for eighty-two measurements in each region.

Equations (5), (6), and (7) were used to predict the drop velocities measured by Hu and Kintner (*3*), who formed their drops in a manner different from that used here. The comparison for both regions is shown in Figure 5. Deviations are greatest in region I (15.2% average deviation) but are satisfactory for region

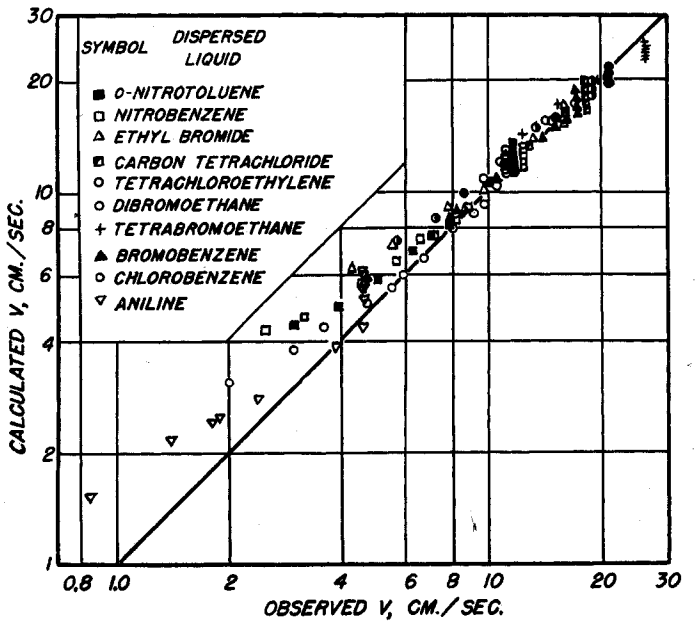

Fig. 5. Comparison of calculated and observed terminal velocities, data of (*3*), regions I and II.

The constants a and b, as determined by this method, were substantially zero. The equations are more useful when solved for velocity of the drops. If the value 0.015 as the exponent on σ developed by Equation (3) and 0.01 as that on d developed by Equation (4) are neglected, the equations become the dimensional equations

$$V_{\mathrm{I}} = 38.3\rho_c^{-0.45}\Delta\rho^{0.58}\mu_c^{-0.11}d^{0.70} \quad (5)$$

and

$$V_{\mathrm{II}} = 17.6\rho_c^{-0.55}\Delta\rho^{0.28}\mu_c^{0.10}\sigma^{0.18} \quad (6)$$

The critical diameter d_c which determines when the transition from region I to II is to be made is most easily established by solving Equations (5) and (6) simultaneously, eliminating V:

$$d_c = 0.33\rho_c^{-0.14}\Delta\rho^{-0.43}\mu_c^{0.30}\sigma^{0.24} \quad (7)$$

Use of Equation (7) will, however, fail to indicate the development of the maximum II (5.5% average deviation). Hu and Kintner presented an empirical correlation of their own data which, with slight modification, can be put in the same form as Equations (3) and (4) but with different constants. Their correlation is satisfactory for both their own and the present data only for systems of fairly high interfacial tension; whereas the present correlation is superior for systems of low interfacial tension.

Licht and Narasimhamurty (*6*) also present drop-velocity data and a correlation of their data permitting a trial-and-error computation of the velocity in region II only, suitable for systems of fairly high interfacial tension. Figure 6 shows a comparison of their observed data and those calculated by Equations (5), (6), and (7) for both regions. The manner of formation of the drops in this case was somewhat similar to that used in the present work; yet the deviation between observed and computed

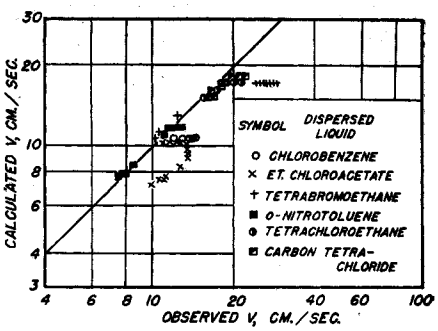

Fig. 6. Comparison of calculated and observed terminal velocities, data of (*6*), regions I and II.

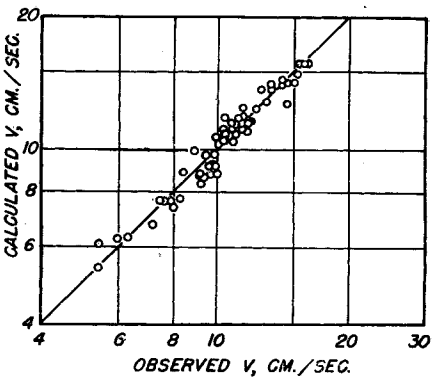

Fig. 7. Comparison of calculated and observed terminal velocities, data of (*5*), regions I and II, twenty systems.

velocities is fairly large for some systems. On the other hand, the observed data for some of the systems common to their study and that of Hu and Kintner are not entirely reconcilable, possibly because of the circulation and oscillation effects which can so easily be introduced.

Figure 7 compares the data of Keith and Hixson (*5*) with the velocities predicted by Equations (5), (6), and (7). As there were so many experimental points, comparisons were made from their smoothed curves at intervals of $d = 0.1$ cm. Even so, there were 114 calculations, and most of the points fall on top of each other in the figure. The average deviation for both velocity regions, for twenty systems of great range of physical properties, is 4.5%. This excellent check is considered particularly significant, as the drops were formed in a manner coinciding most closely with that found in liquid-extraction equipment and the drops flowed through a countercurrently moving continuous liquid. The drop velocities of Figure 7 are values relative to the velocity of the continuous liquid, and there seemed to be no segregation of the data, regardless of whether surfactants or other solutes were used to influence the interfacial tension.

The drop velocities of Smirnov and Ruban (*7*) were also compared with those predicted by Equations (5), (6), and (7). Most of the data were obtained with drops rising or falling against a counterflow of the continuous liquid in very

narrow tubes, some as small as 1.8 cm. diameter. Study of the data showed a marked influence of the velocity of the continuous liquid on the relative velocities of the drops, possibly as a result of the small tube diameter. For twenty-five measurements made with a stagnant continuous liquid, involving nine systems and both regions, the average deviation of predicted and observed velocities was 8.8%. When the continuous liquid moved, the agreement was generally poor.

DROP ECCENTRICITY

Photographs of the drops for all but two of the systems, which did not photograph well, provided data on the distortion of the drops from spherical shape. The shapes varied from almost perfect spheres for the smallest drops to almost perfect spheroids for the large drops of some systems. In other cases the drops were considerably flattened and irregular. Expressing the eccentricity simply as the ratio of the largest horizontal diameter to the largest vertical diameter, the data are shown in Figure 8. The drop shape for most of the systems varied somewhat rhythmically with time as the drops passed through the continuous liquid, and the data plotted in the figure represent rough averages of the maximum and minimum distortions. The eccentricity–spherical-diameter relation is substantially linear for all but the smallest drop sizes, which coincides with the findings of Keith and Hixson (5). An unsuccessful attempt was made to correlate these data through the distortion coefficient of Hughes and Gilliland (4). It was found, however, that the slopes of the lines of Figure 8 are evidently related to the quantity $(\Delta\rho)^{0.5}/\sigma$, as shown in Figure 9. The data of Keith and Hixson, except for the system toluene-water, the eccentricity-diameter curve of which was not straight, seem to conform roughly to this generalization.

SUMMARY

Measurements of the terminal velocity of rise or fall of liquid drops immersed in another insoluble liquid have led to two correlations of velocities with drop diameters and physical properties, one for each of the two velocity-diameter regions. The correlations are easily used and cover a wide range of physical properties. The eccentricity of the drops varies substantially linearly with drop diameter and is related to the quantity $(\Delta\rho)^{0.5}/\sigma$.

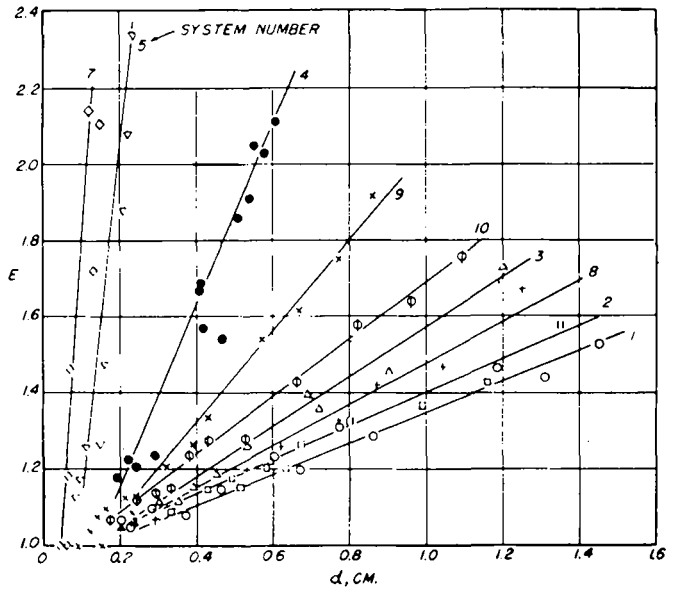

Fig. 8. Drop eccentricities.

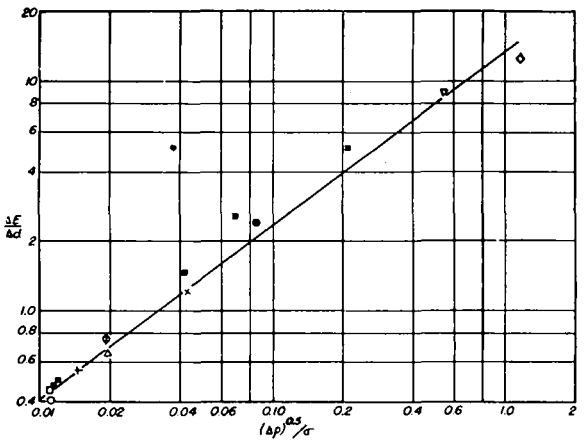

Fig. 9. Correlation of eccentricity data. (Data of Keith and Hixson are shown by the darkened squares.)

ACKNOWLEDGMENT

The authors are indebted to R. C. Kintner, who kindly supplied an English translation of the Smirnov-Ruban paper, and to F. W. Keith for his suggestions.

NOTATION

a, b, c, e, h, m = constants
C_D = drag coefficient $4\Delta\rho\ dg/3\rho_C V^2$, dimensionless
d = diameter of a sphere of the same volume as a drop, cm.
d_c = critical diameter of a drop, at the transition from region I to region II, cm.
d_H = maximum horizontal diameter of a drop, cm.
d_V = maximum vertical diameter of a drop, cm.
E = drop eccentricity = d_H/d_V
f = function
g = acceleration due to gravity, cm./sec.^{2}
g_c = conversion factor, 980 (g.-mass)(cm.)/(g.-force)(sec.)^2
Re = Reynolds number = $dV\rho_C/\mu_C$, dimensionless
V = terminal velocity, cm./sec.
We = Weber number = $V^2\ d\rho_C/\sigma g_c$
$\Delta\rho$ = difference in densities of dispersed and continuous liquids, g./cc.
μ = viscosity, poises
ρ = density, g./cc.
σ = interfacial tension, dynes/cm.

Subscripts

C = continuous liquid
D = dispersed liquid
I, II = regions I and II

LITERATURE CITED

1. Garner, F. H., and A. H. P. Skelland, *Chem. Eng. Sci.*, **4**, 149 (1955).
2. Harkins, W. D., and F. E. Brown, *J. Am. Chem. Soc.*, **41**, 499 (1919).
3. Hu, Shengen, and R. C. Kintner, *A.I.Ch.E. Journal*, **1**, 42 (1955).
4. Hughes, R. R., and E. R. Gilliland, *Chem. Eng. Progr.*, **48**, 497 (1952).
5. Keith, F. W., and A. N. Hixson, *Ind. Eng. Chem.*, **47**, 258 (1955).
6. Licht, William, and G. S. R. Narasimhamurty, *A.I.Ch.E. Journal*, **1**, 366 (1955).
7. Smirnov, N. I., and V. L. Ruban, *J. Appl. Chem. (U.S.S.R.)*, **22**, 1068 (1949).
8. Weisberger, Arnold, ed., "Physical Methods of Organic Chemistry," Interscience Publishers, Inc., New York (1945).