

## LITERATURE CITED

- (1) Bleakney, W., Weimer, D. K., and Fletcher, C. H., *Rev. Sci. Instruments*, **20**, 807 (1949).
- (2) Castleman, R. A., Jr., *Bur. Standards J., Research*, **6**, 396 (1931).
- (3) Hinze, J. O., *Applied Sci. Research, A*, **1**, 273 (1948).
- (4) Hochschwender, E., dissertation, Heidelberg, 1919.
- (5) Lane, W. R., and Edwards, J., Unpublished Ministry of Supply Reports.
- (6) Lenard, P., *Ann. Physik*, **65**, 629 (1921).
- (7) Merrington, A. C., and Richardson, E. G., *Proc. Phys. Soc.*, **59**, 1 (1947).
- (8) Nukiyama, S., and Tanasawa, Y., *Trans. Soc. Mech. Engrs. (Japan)*, **6**, No. 22, II-7 and II-15 (1939).
- (9) Payman, W., and Shepherd, W. C. F., *Proc. Roy. Soc. (London)*, **A186**, 293 (1946).
- (10) Schweitzer, P. H., *J. Applied Phys.*, **8**, 513 (1937).
- (11) Simons, A., and Goffe, G. R., Aero. Research Council R and M, No. 2343.
- (12) Taylor, Sir Geoffrey, Min. of Supply Paper AC 10647/Phys. C69 (1949).
- (13) Weber, C., *Z. angew. Math. u. Mech.*, **2**, 136 (1931).

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# Droplet Size Distribution in Sprays

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**A**N ACCURATE knowledge of drop size distribution as a function of the conditions of the system is a prerequisite for fundamental analysis of the transport of mass or heat or of the separation of phases in a dispersed system. For example, the "drop" size distribution in a fractionation column determines the rate of heat and mass transfer and also the amount of entrainment. The size range of droplets introduced into, or created within, a cyclone separator must be known in order to design the unit for the desired separation efficiency. In liquid-liquid systems the size distribution helps to determine settling rates (and thus holdup), mass and heat transfer rates, entrainment, and possibly coalescence rate. Again, in many combustion studies, rate of evaporation or burning of individual drops must be estimated; here also a knowledge of the drop sizes is important.

Despite the importance of drop size distribution in evaluating most separation processes, little is known about the actual size range of droplets in these units. This is due to difficulties inherent in measuring drop sizes in these types of apparatus. However, many data are available for size distribution in more simple atomizers such as spray nozzles of various types. It is believed that analysis of these data will make it possible to estimate drop size distribution in more complex apparatus with reasonable accuracy.

This paper presents a summary of the investigation of drop size distribution in sprays in the following order: fundamental definitions and concepts, analysis of previously developed distribution equations, development and application of a new distribution equation known as the "upper-limit" law, and comparison of the various distribution functions with experimental data.

For the case of a mechanical spray, a brief discussion is given concerning relation of the parameters required for the upper-limit law to the design conditions and physical properties of the spray.

## DEFINITION OF SPRAY

In the field discussed here, a spray is considered as a system of liquid droplets in a fluid continuous phase. By specifying "droplets" we eliminate macroscopic cases wherein large individual drops, slugs, or columns predominate, and microscopic cases wherein dispersion is of molecular order, or at least where drop-

General features of size distribution are reviewed for dispersed systems. The concepts of "mean diameter" and "distribution parameter" are clarified and generalized. Previously applied distribution equations (Rosin and Rammler, Nukiyama and Tanasawa, log-probability) are examined critically in regard to theoretical soundness, and application to spray data.

A new equation, called the upper-limit equation, is formulated and proposed as a standard for describing droplet size distributions in sprays. It is based on the differential equation of the "normal" or Gaussian distribution, the distributed quantity being  $y = \ln ax/(x_m - x)$  where  $a$  is a dimensionless parameter,  $x$  is droplet diameter, and  $x_m$  is maximum stable diameter.

The upper-limit equation is applied to a wide variety of experimental data on sprays and more limited results on other dispersoids. It is concluded that the new equation fits the available spray data accurately, calculates the mean diameters accurately, applies also to emulsions and aerosols when the mechanism of formation is not too different from that of sprays, and indicates the type of distribution function that may be derivable from the basic mechanism of dispersion, when this mechanism is better understood. For a mechanical spray, the relation of the parameters of the distribution equation to physical properties and design variables is indicated.

lets are sufficiently small to prevent detection of phase boundaries. By specifying a "system" we imply that the droplets have a common origin, usually a body of liquid.

Examples of natural sprays are rains, fogs, waterfall mist, ocean spray, and sneeze spray.

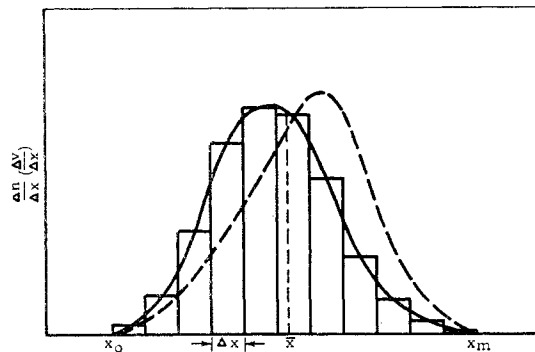


Figure 1. Droplet Size Distribution (Schematic)

Among artificial sprays are fountain sprays, atomizer sprays, entrained liquid in a fractionating column, and the disperse phase in a solvent extraction column.

Sprays considered in this paper all have a resultant motion

in a particular direction (above the random motions of droplets), but this restriction need not affect the applicability of the results to other types of systems—e.g., aerosols and fogs.

#### REPRESENTATION OF DISTRIBUTION

Suppose our droplets are counted by groups  $\Delta n$  (fraction of total count), limited by diameters  $x - \Delta x/2$  and  $x + \Delta x/2$ , while the over-all range of  $x$  is between  $x_0$  and  $x_m$ . Then a step diagram (as in Figure 1) represents the proportional count as made. The total area under the steps equals 1.00. But if the number of droplets is large and the  $\Delta x$  interval is small, we may represent the count by a smooth curve (also shown in Figure 1), the area under the curve being 1.00.

Again, we may represent group sizes by volume fractions  $\Delta v$  rather than number fractions. For this case, the step diagram is not shown in Figure 1, but the corresponding curve is shown (dashed line). The curve is skewed to the right, because each group is now weighted in proportion to  $x^3$ . Area under the volume distribution curve also equals 1.00.

#### FUNDAMENTAL SPRAY VARIABLES

The simplest type of statistical system may be defined by a distribution function, together with some sort of a mean and some measure of deviation from this mean. For sprays we take as our dependent variables a mean diameter,  $\bar{x}$ , and a distribution factor,  $\delta$ . Of course, this assumes that all droplets have the same shape (usually spheres), which is generally valid for fully developed sprays.

If, for physical reasons, maximum and minimum drop diameters are established, these will be designated by  $x_m$  and  $x_0$ , respectively.

#### MEAN DIAMETERS

In many mass transfer and flow processes it is desirable to work only with average diameters instead of the complete drop size distribution. Table I shows some of the mean diameters and some fields of application for each. In order to use these means effectively, they must be readily calculated from the drop size distribution equation.

Table I. Mean Diameters

(Cf. Equation 4.4)

$p$	$q$	$p + q$ (Order)	Name of Mean Diameter	Field of Application
0	1	1	Linear	Comparisons, evaporation
0	2	2	Surface	Surface area controlling—e.g., absorption
0	3	3	Volume	Volume controlling—e.g., hydrology
1	2	3	Surface diameter	Adsorption
1	3	4	Volume diameter	Evaporation, molecular diffusion
2	3	5	Sauter	Efficiency studies, mass transfer, reaction
3	4	7	De Brouckere	Combustion equilibrium

The actual application of these mean values forms too large a subject to treat here, but an example or two may be given to illustrate the method:

First, consider Langmuir evaporation of a droplet according to the law

$$\frac{dm}{dt} = -kx \quad (4.01)$$

Now for a number of droplets of different diameters,

$$\frac{d(\Sigma m)}{dt} = -k\Sigma x \quad (4.02)$$

Dividing by the total number of droplets, we then have

$$\frac{d\bar{m}}{dt} = -k\bar{x}_{10} \quad (4.03)$$

Thus it is seen that the law (Equations 4.01) holds for the av-

erage droplet as well as the individual droplet, provided the linear mean diameter is used.

Again, consider the efficiency of an atomizing operation, in which a fraction  $E$  of the energy lost in pressure drop  $\Delta p$  is used in forming new surface. This may be expressed by

$$E \left( \frac{\pi x^2}{6} \Delta p \right) = \pi x^2 \sigma \quad (4.04)$$

for a single droplet, or

$$E \Delta p \Sigma x^3 = 6\sigma \Sigma x^2 \quad (4.05)$$

for the entire spray. Then

$$E \Delta p \bar{x}_{32} = 6\sigma \quad (4.06)$$

indicating that atomizing efficiency may be calculated from the Sauter mean diameter.

The linear mean diameter can be calculated from the distribution data (as represented by Figure 1) according to the equation:

$$\bar{x}_{10} = \Sigma x \Delta n / \Sigma \Delta n = \Sigma x \Delta n \quad (4.1)$$

The linear mean diameter,  $\bar{x}_{10}$ , is a diameter which when multiplied by the total count will give the same result as adding the products of separate group counts by the representative diameters of the groups. For a smooth distribution curve we may write:

$$\bar{x}_{10} \int_{x_0}^{x_m} \frac{dn}{dx} dx = \int_{x_0}^{x_m} x \frac{dn}{dx} dx \quad (4.2)$$

The subscripts on this linear mean and other means (discussed below) indicate the order of the mean or the method of averaging. The first number of the subscript refers to the power of  $x$  appearing in the right-hand integral and the second to the power appearing in the left-hand integral. Alternatively, Equation 4.2 can be written as

$$\bar{x}_{10} = \int_{x_0}^{x_m} x \frac{dn}{dx} dx / \int_{x_0}^{x_m} \frac{dn}{dx} dx \quad (4.21)$$

Here it is apparent that the subscript 1 refers to the power of  $x$  in the numerator of the expression and the subscript 0 to the power of  $x$  in the denominator.

If the function of interest is proportional to droplet surface, we choose a mean  $\bar{x}_{20}$ , so that

$$\bar{x}_{20}^2 \int_{x_0}^{x_m} \frac{dn}{dx} dx = \int_{x_0}^{x_m} x^2 \frac{dn}{dx} dx \quad (4.3)$$

Extending this idea, let us consider two functions of droplet diameter which are compared, and let their orders be  $p$  and  $q$ . Then the relevant mean,  $\bar{x}_{qp}$ , is defined by

$$\bar{x}_{qp}^{q-p} \int_{x_0}^{x_m} x^p \frac{dn}{dx} dx = \int_{x_0}^{x_m} x^q \frac{dn}{dx} dx \quad (4.4)$$

This may also be written in terms of volume distribution as

$$\bar{x}_{qp}^{q-p} \int_{x_0}^{x_m} x^{p-3} \frac{dv}{dx} dx = \int_{x_0}^{x_m} x^{q-3} \frac{dv}{dx} dx \quad (4.5)$$

A limiting case for  $p = q$  is defined by

$$\bar{x}_{qq} = \lim_{c \rightarrow 0} \left[ \int_{x_0}^{x_m} x^{q+c} \frac{dn}{dx} dx / \int_{x_0}^{x_m} x^{q-c} \frac{dn}{dx} dx \right]^{1/2c} \quad (4.6)$$

Application of the theory of limits to Equation 4.6 gives

$$\ln \bar{x}_{qq} = \int_{x_0}^{x_m} x^q (\ln x) \frac{dn}{dx} dx / \int_{x_0}^{x_m} x^q \frac{dn}{dx} dx \quad (4.61)$$

This defines the family of "hypergeometric" mean values. The most familiar of these is the geometric mean,  $\bar{x}_{00}$ , given by

**Table II. Numerical Comparison of Mean Diameters**

x	Δn			
	I	II	III	IV
1	0.556	0.290	0.503	0.447
2	0.277	0.546	0.382	0.426
3	0.167	0.164	0.115	0.127
$\bar{x}_{10}$	1.61 <sup>a</sup>	1.87	1.61 <sup>a</sup>	1.68
$\bar{x}_{20}$	1.78	1.99	1.75	1.82
$\bar{x}_{30}$	1.94 <sup>a</sup>	2.09	1.88	1.94 <sup>a</sup>
$\bar{x}_{21}$	1.96	2.11	1.90	1.96
$\bar{x}_{32}$	2.30 <sup>a</sup>	2.30 <sup>a</sup>	2.17	2.21

<sup>a</sup> Examples are set up to give I and II the same value of  $\bar{x}_{32}$ , I and III the same value of  $\bar{x}_{10}$ , etc.

**Table III. Comparison of Mean Diameters via Rosin-Rammler Equation**

δ	$\bar{x}_{20}/\bar{x}$	$\bar{x}_{21}/\bar{x}$	$\bar{x}_{32}/\bar{x}$	$\bar{x}_{30}/\bar{x}_{32}$	$\bar{x}_{21}/\bar{x}_{32}$
3	0.000	0.506	0.738	0.000	0.686
4	0.651	0.692	0.816	0.798	0.848
6	0.826	0.833	0.887	0.931	0.939

$$\ln \bar{x}_{00} = \int_{x_0}^{x_m} (\ln x) \frac{dn}{dx} dx \quad (4.62)$$

The mean values defined by different *p*'s and *q*'s are not all independent. Thus

$$\bar{x}_{qp}^{q-p} = \bar{x}_{qc}^{q-c} / \bar{x}_{pc}^{p-c} \quad (4.7)$$

In particular,

$$\bar{x}_{qp}^{q-p} = \bar{x}_{q0}^q / \bar{x}_{p0}^p \quad (4.71)$$

Also

$$\bar{x}_{qp} = \bar{x}_{pq} \quad (4.8)$$

Fractional and negative values of *p* and *q* are not excluded by the general definition, and may in fact apply to certain transfer problems. However, the examples given in Table I are for integral pairs only, as these are the most generally useful cases.

Two different distributions of drop sizes may have the same value of one of the mean diameters and yet entirely different values of other mean diameters. In order to illustrate this point, the examples of Table II are presented.

These fictitious distributions I, II, III, and IV are assumed to have only three diameter classes (*x* = 1, 2, and 3). Distributions I and II have the same  $\bar{x}_{32}$ , but all other mean diameters are different. Again I and III have the same value of  $\bar{x}_{10}$  but somewhat different values of the other means. For this case a decrease in the proportion of smaller drops decreases  $\bar{x}_{32}$ . Because one large drop may obscure a number of small drops in counting, the experimental value of  $\bar{x}_{32}$  may be considerably in error, particularly when both *x* and Δ*n* may vary by factors of 100 or more. Finally, I and IV have the same  $\bar{x}_{30}$  but their other means differ.

In each example,  $\bar{x}_{30} \approx \bar{x}_{21}$ . This property is characteristic of experimental distribution data and is consistent with the log-probability law and its modifications. But distributions which approximate the Rosin-Rammler law (discussed below) will have this characteristic only at high values of the distribution factor, δ.

**ROSIN-RAMMLER EQUATION**

We now consider some of the functions which have been proposed to represent the distribution curves typified by Figure 1.

One of the first of these was presented by Rosin and Rammler (17) for application to powdered materials. It has been applied by a number of investigators with varying success to droplet systems.

This distribution function is generally given in the "cumulative volume" form:

$$1 - v = e^{-(x/\bar{x})^\delta} \quad (5.1)$$

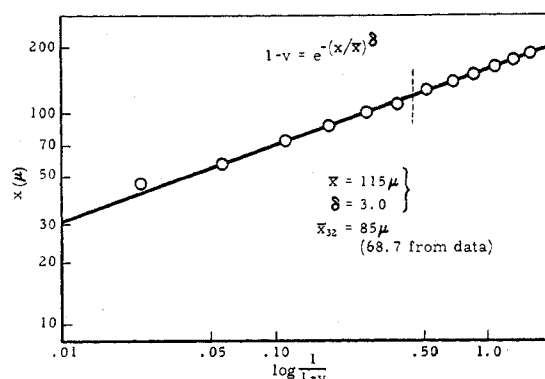
where 1 - *v* is the volume fraction of drop material occurring in drops of diameter greater than *x*,  $\bar{x}$  is a size parameter, and δ is a distribution parameter. From Equation 5.1 we can derive the "volume distribution" equation

$$\frac{dv}{dx} = \frac{\delta x^{\delta-1}}{\bar{x}^\delta} e^{-(x/\bar{x})^\delta} \quad (5.2)$$

Then the "numerical distribution" equation is obtained by dividing *dv/dx* by *x*<sup>3</sup> and inserting a factor to make

$$\int_0^\infty \frac{dn}{dx} dx = 1:$$

$$\frac{dn}{dx} = \frac{\delta x^{\delta-4}}{\bar{x}^{\delta-3} \Gamma\left(1 - \frac{3}{\delta}\right)} e^{-(x/\bar{x})^\delta} \quad (5.3)$$



**Figure 2. Rosin-Rammler Analysis of Lee Data (13)**

The gamma function appearing in this equation is the generalized factorial. Its defining equation is

$$\Gamma(c) = \int_0^\infty U^{c-1} e^{-U} dU \quad (5.31)$$

Tables of this function are given by Lange (12) and other handbooks.

If we required the "cumulative numerical" distribution, this could be obtained by integrating Equation 5.3. The result would involve the "incomplete gamma function," for which special tables are required.

The expression for mean diameters (cf. Equation 4.5) becomes

$$\bar{x}_{qp}^{q-p} = \bar{x}^{q-p} \Gamma\left(\frac{q-3}{\delta} + 1\right) / \Gamma\left(\frac{p-3}{\delta} + 1\right) \quad (5.4)$$

In particular, the Sauter mean diameter is

$$\bar{x}_{32} = \bar{x} / \Gamma\left(1 - \frac{1}{\delta}\right) \quad (5.41)$$

Table III summarizes some calculations of mean diameters via Equation 5.4.

Some interesting consequences of using the Rosin-Rammler equation are here illustrated:

Equivalence of  $\bar{x}_{30}$  and  $\bar{x}_{21}$  is predicted only at high values of δ. Unreasonable values of mean diameters may result. For δ = 3, we found  $\bar{x}_{30} = 0$ . For δ = 2, we would find  $\bar{x}_{30}$  negative.

For application to data we may put Equation 5.1 into the form:

$$\ln \ln \frac{1}{1-v} = \delta(\ln x - \ln \bar{x}) \quad (5.5)$$

This represents  $\ln \ln \frac{1}{1-v}$  as a linear function of  $\ln x$ . Figure 2 shows a set of data which gives a particularly good fit to a straight line when plotted on this basis. From this plot we readily find  $\delta$  as the slope of the line and  $\bar{x}$  as the value of  $x$  for which  $1-v = e^{-1}$ .

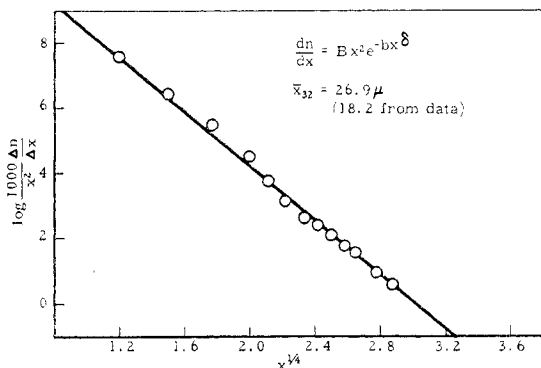


Figure 3. Nukiyama-Tanasawa Analysis of Houghton Data (8)

But even though a good linear fit is obtained, the value of  $\bar{x}_{32}$  calculated from the parameters is 85 microns, while that calculated by summation of data is 68.7 microns. This discrepancy is due partly to the deviation of two points from the line at small diameters, but mainly to the assumption of infinite range of  $x$  values. This again illustrates the unreliability of the Rosin-Rammler equation for calculating mean diameters, although it may be useful for comparisons over a small range of the parameters.

#### NUKIYAMA-TANASAWA EQUATION

This completely empirical equation is generally given in the numerical distribution form (15)

$$\frac{dn}{dx} = Bx^2 e^{-bx^\delta} \quad (6.0)$$

where the size parameter,  $b$ , has the dimension of  $x^{-\delta}$ .

When the constant  $B$  is properly evaluated, this becomes:

$$\frac{dn}{dx} = \frac{\delta b^{\delta/\delta}}{\Gamma(3/\delta)} x^2 e^{-bx^\delta} \quad (6.1)$$

Then the volume distribution is

$$\frac{dv}{dx} = \frac{b^{\delta/\delta}}{\Gamma(6/\delta)} x^5 e^{-bx^\delta} \quad (6.2)$$

The cumulative volume fraction is then

$$v = \int_0^{bx^\delta} U^{\frac{6}{\delta}-1} e^{-U} dU / \int_0^\infty U^{\frac{6}{\delta}-1} e^{-U} dU \quad (6.3)$$

This may be expressed as a ratio of incomplete and complete gamma functions:

$$v = \Gamma_{bx^\delta} \left( \frac{6}{\delta} \right) / \Gamma \left( \frac{6}{\delta} \right) \quad (6.31)$$

The equation for mean diameters becomes:

$$\bar{x}_{qp} q^{-p} = b^{-\left(\frac{q-p}{\delta}\right)} \Gamma \left( \frac{q+3}{\delta} \right) / \Gamma \left( \frac{p+3}{\delta} \right) \quad (6.4)$$

Comparison of some mean diameters for some representative values of  $\delta$  is illustrated in Table IV.

This shows better agreement between  $\bar{x}_{30}$  and  $\bar{x}_{21}$  than that indicated by the Rosin-Rammler equation (cf. Table III).

For application to data, we put Equation 6.1 in the form

$$\ln \left( \frac{1}{x^2} \frac{dn}{dx} \right) = \ln \frac{\delta b^{\delta/\delta}}{\Gamma(3/\delta)} - bx^\delta \quad (6.5)$$

which expresses  $\ln \left( \frac{1}{x^2} \frac{dn}{dx} \right)$  as a linear function of  $x^\delta$ .

Figure 3 shows a satisfactory fit of the linear form to a set of data. This is obtained by trying different values of  $\delta$  to see which gives the best line (usually 1, 1/2, 1/3, ...). The case illustrated has been equally well fitted by Lewis *et al.* (14), using  $\delta = 1/3$  instead of 1/4. The uncertainty of  $\delta$  leads to some uncertainty in  $\bar{x}_{32}$ , which is calculated as 20.3 microns for  $\delta = 1/3$ , 26.9 microns for  $\delta = 1/4$ , and 18.2 microns from data.

In some cases, application of this equation has led to values of  $\bar{x}_{32}$  larger than any experimentally observed drop diameter. Lewis *et al.* (14) have tried to explain this by assuming that some large droplets existed but were not counted. But there are so many cases of this kind that if we are to maintain the validity of the Nukiyama-Tanasawa equation, we must discredit the counting techniques used in obtaining the data.

#### LOG-PROBABILITY EQUATION

When we deal with single-valued—i.e., nondistributed—variables in a physical problem where the fundamental mechanism is not clearly understood, we may apply dimensional analysis to limit the grouping in which the variables can appear. Analogously, when we deal with multivalued—i.e., distributed—variables, we may apply statistical analysis to limit the distributions which the variables may take.

Suppose, then, we have a distribution of drop diameters such as that represented in Figure 1. As a first assumption, let us take  $x_0 = 0$  and  $x_m = \infty$  as end points of the curve, saying that if there actually are "smallest" and "largest" droplets in the spray,  $dv/dx$  may be taken as infinitesimal or zero beyond actual  $x_0$  and  $x_m$ . Furthermore, let us assume that the "classes" established by the fundamental mechanism are graded exponentially rather than linearly—e.g., say the proper grading series for representative diameters is not 0.5, 1.5, 2.5, 3.5, ... microns, but 0.01, 0.1, 1.0, 10, ... microns. Then a suitable revision of Figure 1 will represent the distribution by

$$\frac{dv}{dy} = \phi(y) \quad (7.1)$$

where

$$y = \ln x/\bar{x} \quad (7.2)$$

When sets of spray data are plotted up on this basis, we find that they ordinarily give a somewhat symmetrical distribution about some value of  $y$  which is close to a single maximum in the curve. However, some sets of measurements do not include sufficiently small diameters to make the maximum apparent.

Table IV. Comparison of Mean Diameters via Nukiyama-Tanasawa Equation

$\delta$	$\bar{x}_{30} b^{1/\delta}$	$\bar{x}_{21} b^{1/\delta}$	$\bar{x}_{32} b^{1/\delta}$	$\bar{x}_{30}/\bar{x}_{32}$	$\bar{x}_{21}/\bar{x}_{32}$
1/4	2068	2184	4080	0.507	0.535
1/3	69.3	72.0	110.0	0.630	0.655
1	3.913	4.000	5.000	0.783	0.800

Now, if our knowledge of the spray-forming mechanism is so sketchy as to provide nothing better than a random distribution of  $y$  values about some mean value, we can still make a good guess at the nature of  $\phi(y)$ . This will be simply the "normal distribution" function

$$\frac{\delta}{\sqrt{\pi}} e^{-\delta^2 y^2}$$

(cf. Coolidge, 2, or any standard treatise on statistical method). Here  $\delta$  is related to the standard deviation of  $y$ , and hence to the standard deviation of  $x$ .

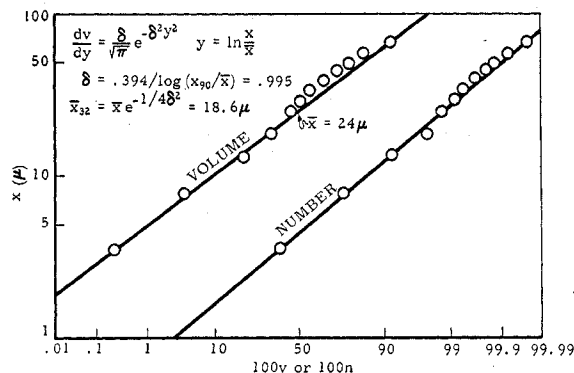


Figure 4. Log-Probability Analysis of Houghton Data (8)

From these considerations we arrive at the volume distribution equation

$$\frac{dv}{dy} = \frac{\delta}{\sqrt{\pi}} e^{-\delta^2 y^2} \quad (7.3)$$

$y$  being defined by Equation 7.2, in which  $\bar{x}$  now corresponds to the volume median diameter.

Cumulative volume fraction is determined by integrating Equation 7.3:

$$v = \frac{1}{\sqrt{\pi}} \int_{-\infty}^y \delta e^{-U^2} dU \quad (7.4)$$

This may be evaluated if desired by use of the "probability integral"  $\Theta$  [cf. Coolidge (2) or Lange (12) for suitable tables]. However, this evaluation is unnecessary if we use log-probability paper which has one coordinate scaled by  $\Theta(y)$  and the other by  $\log x$ . This is illustrated in Figure 4. Also shown in the figure is a line for the cumulative numerical fraction. The equation for this can be obtained from the numerical distribution equation:

$$\frac{dn}{dy} = \frac{\delta}{\sqrt{\pi}} e^{-\left(\delta y + \frac{3}{2\delta}\right)^2} \quad (7.5)$$

by integrating:

$$n = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\delta y + \frac{3}{2\delta}} e^{-U^2} dU \quad (7.6)$$

The method of calculating the distribution factor,  $\delta$ , from the line is discussed below (in connection with the upper-limit equation).

The general expression for mean diameters for this distribution is

$$\bar{x}_{pq} = \bar{x} e^{\frac{p+q-6}{4\delta^2}} \quad (7.7)$$

In particular, the Sauter mean diameter is

$$\bar{x}_{32} = \bar{x} e^{-\frac{1}{4\delta^2}} \quad (7.71)$$

For the example of Figure 4, the Sauter mean diameter has been calculated as 18.6 microns. This is somewhat closer to the value of 18.2 given directly by the data than is the best value of 20.3 given by the Nukiyama equation. Similar results for other data indicate that the log-probability equation is so far the best one for calculating mean diameters.

Also we note that Equation 7.7 predicts equality of means of the same order—e.g.,  $\bar{x}_{20} = \bar{x}_{21}$ ,  $\bar{x}_{40} = \bar{x}_{41}$ . This equality is approximated by most of the distribution data and is also illustrated by the fictitious data of Table II.

Table V summarizes certain mean diameters calculated for particular values of  $\delta$ .

Additional remarks concerning the log-probability equation may be found in Bevans (1). Meteorological applications and references are provided by Howell (9).

#### MODIFIED PROBABILITY EQUATIONS

It has been found that the log-probability equation predicts the general distribution trend correctly for most data and gives good results for mean-diameter calculations. But many data show a deviation from the straight line (Figure 4) at high values of  $x$ . Also, the data indicate that it may be desirable to emphasize the existence of a maximum drop diameter  $x_m$ , rather than just considering extremely large drops to have extremely low frequencies. These considerations lead us to seek a modification of the relation of  $y$  to  $x$  (Equation 7.2).

In determining the new relation, we seek to fulfill the following requirements:

- As  $y$  goes from  $-\infty$  to  $+\infty$ ,  $x$  shall go from  $x_0$  (minimum sized drop in the distribution) to  $x_m$  (maximum sized drop)
- Qualitatively, the function shall be capable of ultimate interpretation in terms of fundamental mechanisms—i.e., it shall not predict trends contrary to those reasonably established from physical principles
- Quantitatively, the resulting equations shall satisfy existing data within the accuracy of method and measurement

In addition to the above, we should like to find simplicity of mathematical form in our function. This will permit ready evaluation of the distribution parameters from the data, and of the means, quartiles, etc., from the parameters, without the aid of involved charts and tables.

Now several functions have been found which satisfy these requirements. The most satisfactory class of such functions appears to be:

$$y = \ln \frac{ax^s}{x_m^s - x^s} \quad (8.1)$$

where  $a$  is a dimensionless constant and  $s$  a positive number. Here the fulfillment of condition (a) above (with the assumption  $x_0 = 0$ ) is evident.

Table V. Comparison of Mean Diameters via Log-Probability Equation

$\delta$	$\bar{x}_{30}/\bar{x}$	$\bar{x}_{21}/\bar{x}$	$\bar{x}_{32}/\bar{x}$	$\bar{x}_{30}/\bar{x}_{32}$	$\bar{x}_{21}/\bar{x}_{32}$
0.5	0.050	0.050	0.368	0.136	0.136
1.0	0.475	0.475	0.780	0.609	0.609
1.5	0.636	0.636	0.860	0.740	0.740

In regard to (b), we may think of  $x^s$  as representing a simple function of drop size—e.g., diameter, surface, or volume. Then  $x_m^s$  is the same function of maximum drop size; so that  $x_m^s - x^s$  is a measure of "size deficiency" or reduction from maximum drop size.

Now if we consider a mechanism wherein "maximum" drops originally exist but subsequently break into two fragments each, we can represent the ratio of measures (whether by volume, surface, or diameter) of these fragments (after they assume uniform shape) by  $x^s/(x_m^s - x^s)$ . Then we can reproduce the argument which led to the log-probability equation, applying it this time to the above ratio, so that the parameter  $y$  (the quantity which distributes randomly) assumes the form given in Equation 8.1.

In regard to condition (c), we find that for the data which have been analyzed, Equation 7.1 gives good fits for  $s = 1, 2, \text{ or } 3$ .

#### SPECIAL UPPER-LIMIT FUNCTION

As indicated above, application of Equation 8.1 to the data has proved equally satisfactory for  $s = 1, 2, \text{ or } 3$ . To fit the mechanism which was suggested, we might favor  $s = 3$  as being

most easily interpreted in terms of volume redistribution. But unless evidence can be found substantiating this choice, we shall do just as well to employ  $s = 1$ , for which the calculations are somewhat simpler.

On this basis,

$$y = \ln \frac{ax}{x_m - x} \quad (9.1)$$

Equations 7.3, 7.4, 7.5, and 7.6 are still valid for the present case, even though  $y$  has been redefined.

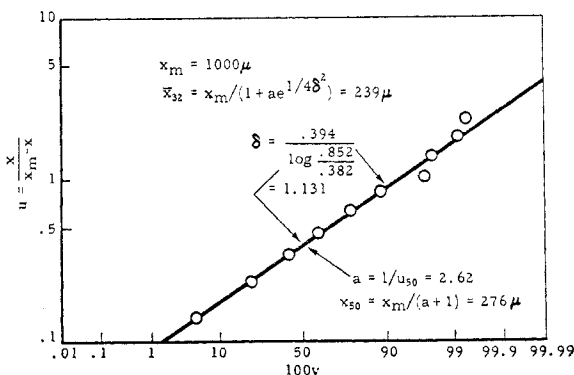


Figure 5. Upper-Limit Analysis of Kolupaev Data (11)

Figure 5 illustrates the application of the upper-limit equation to data. Here  $x_m$  is determined by trial and error to give the best alignment of data points when  $x/(x_m - x)$  is plotted against  $100v$ . Usually three or two trial values of  $x_m$  will suffice, and frequently one. However, if we wish to get a good approximation for  $x_m$  directly, we may proceed as follows:

Plot the data as  $x$  against  $100v$ , drawing a smooth curve to fit these points. From these curves read the 10th, 50th, and 90th percentiles,  $x_{10}$ ,  $x_{50}$ , and  $x_{90}$ . Then calculate  $x_m$  from the formula

$$\frac{x_m}{x_{50}} = \frac{x_{90}(x_{90} + x_{10}) - 2x_{90}x_{10}}{x_{50}^2 - x_{90}x_{10}} \quad (9.2)$$

This equation is easily derived from the geometry of Figure 5.

The parameter  $a$  is readily determined from the line representing the distribution. Since  $y = 0$  at the 50th percentile (where  $x = x_{50}$ , the volume median), we have here

$$0 = \ln \frac{ax_{50}}{x_m - x_{50}} \quad (9.3)$$

or

$$a = \frac{x_m - x_{50}}{x_{50}} \quad (9.4)$$

The parameter  $\delta$  is determined by the slope of the line, hence by any two points on it. Let us designate the coordinate on the log scale by  $u$ :

$$u = \frac{x}{x_m - x} \quad (9.5)$$

Then if we read the values  $u_{90}$  and  $u_{50}$  at the 90th and 50th percentiles ( $100v = 90$  and  $50$ , respectively), we find:

$$\delta = \frac{0.394}{\log(u_{90}/u_{50})} \quad (9.6)$$

This results from consideration of a table of  $\Theta(y)$  together with Equations 9.1 and 7.3. Application of Equation 9.6 is illustrated in Figure 5.

#### MEAN DIAMETERS FROM UPPER-LIMIT EQUATION

From Equations 9.1, 7.3, and 4.5 we may calculate expressions or the mean diameters according to the upper-limit distribution.

Table VI. Comparison of Mean Diameters via Upper-Limit Equation

$a$	$\delta$	$\bar{x}_{31}/x_{30}$	$\bar{x}_{30}/x_{30}$	$\bar{x}_{21}/x_{30}$	$\bar{x}_{32}/x_{30}$	$\bar{x}_{31}/\bar{x}_{32}$	$\bar{x}_{30}/\bar{x}_{32}$	$\bar{x}_{21}/\bar{x}_{32}$
1.0	0.5	0.256	0.099	0.122	0.538	0.475	0.184	0.227
1.0	1.0	0.798	0.708	0.727	0.876	0.911	0.809	0.830
1.0	1.5	0.880	0.831	0.839	0.924	0.952	0.899	0.907

Those most readily derived are those having  $q = 3$ . The results are

$$\bar{x}_{32} = x_m / (1 + ae^{1/4\delta^2}) \quad (10.1)$$

$$\bar{x}_{31} = x_m / (1 + 2ae^{1/4\delta^2} + a^2e^{1/\delta^2})^{1/2} \quad (10.2)$$

$$\bar{x}_{21} = x_m / (1 + 3ae^{1/4\delta^2} + 3a^2e^{1/\delta^2} + a^3e^{9/4\delta^2})^{1/3} \quad (10.3)$$

From these, values of  $\bar{x}_{21}$ ,  $\bar{x}_{30}$ , and  $\bar{x}_{10}$  may be calculated via Equation 4.7.

Table VI summarizes certain mean diameters calculated for particular values of  $\delta$ .

The last two columns of the table indicate that the upper-limit distribution predicts  $\bar{x}_{21}$  slightly higher than  $\bar{x}_{30}$ . This prediction is borne out by the data, giving the upper-limit law a slight advantage in this regard over the log-probability law, which predicts  $\bar{x}_{21}$  exactly equal to  $\bar{x}_{30}$  (cf. Table V).

#### COMPARATIVE FIT OF DISTRIBUTION EQUATIONS

The various distribution equations discussed may be compared directly in terms of (a) numerical distribution, (b) volume distribution, (c) cumulative droplet count, or (d) cumulative volume. Comparisons (b) and (d) have been made of several representative sets of data. Those of type (b) are illustrated in Figures 6, 7, and 8.

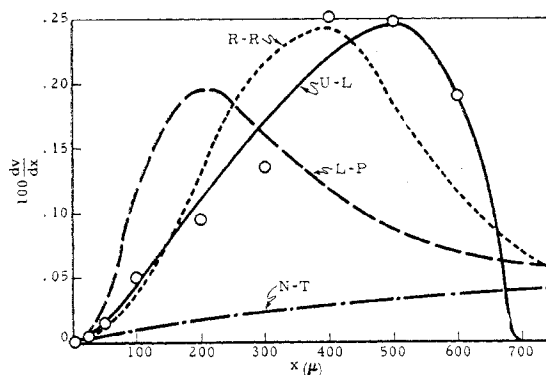


Figure 6. Comparison of Distribution Equations with Houghton Data (8)

Spraco-T nozzle

These comparisons show that the upper-limit equation gives the proper trend in all cases, as well as a good quantitative fit. On the other hand, the equations of Rosin-Rammler, Nukiyama-Tanasawa, and the log-probability are found to fit well in some cases and poorly in others.

In particular, the Nukiyama-Tanasawa equation gives a completely wrong trend for volume distribution in some cases when the parameters are calculated from numerical distribution data. This is a basic weakness of this equation. Typical examples of the poor volume distribution predicted by the Nukiyama-Tanasawa equation using parameters obtained from numerical distribution data are shown in Figures 6 and 8. For these cases the Nukiyama-Tanasawa equation predicts too high a proportion of large drops and actually calculates a Sauter mean diameter beyond the largest observed drop. This discrepancy between numerical and volume distribution does not occur in the

case of distributions based on the Gaussian equation, as is indicated in Figure 4.

Figure 9 summarizes the results of the cumulative distributions of the types shown in Figures 6 to 8.

#### STANDARDIZED DESCRIPTION OF DROPLET SIZE DISTRIBUTION

From such comparisons as those of Figures 6 to 9, it is concluded that the upper-limit equation fits the data better than equations previously used. And from previous considerations, we believe that it may be derived from theoretical considerations (when the fundamental mechanisms of spray formation are better understood). Therefore the upper-limit equation has been adopted as a standard for describing distributions.

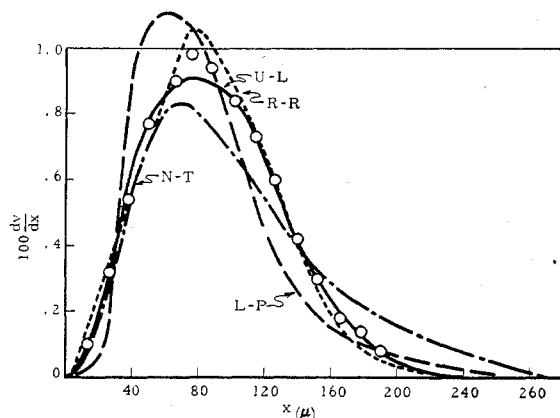


Figure 7. Comparison of Distribution Equations with Lee Data (13)

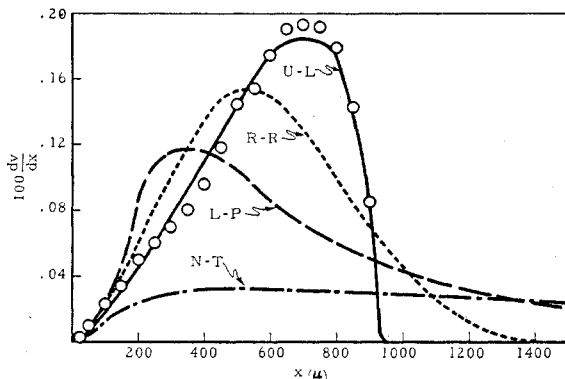


Figure 8. Comparison of Distribution Equations with Johnstone-Kleinschmidt Data (10)

On this basis a distribution is completely characterized by (1) the maximum diameter  $x_m$ , (2) the size parameter  $a$ , and (3) the distribution parameter  $\delta$ . From Equations 10.1 to 10.3 it is evident that these three parameters can be calculated from any three independent mean diameters which are known. Or if  $x_m$  is known,  $a$  and  $\delta$  may be calculated from  $x_{50}$  and  $\bar{x}_{32}$ .

Data of fifty different runs made by nine different investigators (7, 8, 10, 11, 13-16, 18) have been analyzed as in Figure 5. These data are all believed to fit the straight lines within experimental error of the individual runs. However, if each run were repeated a number of times, the averages might show slight trends away from the lines.

Figure 10 shows a similar analysis of some emulsion data. Considering the extremely small size of emulsion droplets and the difficulty of measuring them, the fit is satisfactory. This indicates that the formation mechanism of emulsion droplets may be closely similar to that of spray droplets. The data are those of Cooper (3) and of Harkins and Beeman (5).

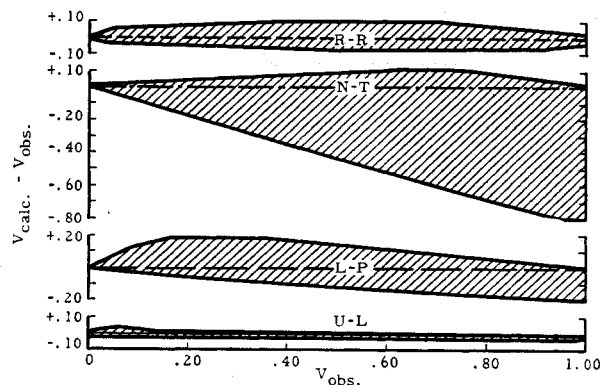


Figure 9. Deviations of Distribution Equations from Data

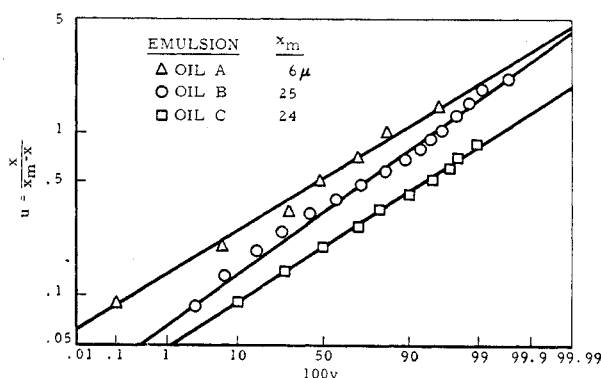


Figure 10. Upper-Limit Analysis of Emulsion Data (3, 5)

#### RELATION OF DISTRIBUTION PARAMETERS TO PHYSICAL VARIABLES

The remaining problem in the drop-size distribution study consists in correlating  $x_m$ ,  $a$ , and  $\delta$  (or the equivalent set  $x_m$ ,  $x_{50}$ , and  $\bar{x}_{32}$ ) with the physical characteristics of the spray-generating system.

These characteristics, of course, include physical properties of the droplet phase (subscript  $d$ ) and of the continuous medium (subscript  $c$ ). Among these properties are densities  $\rho_d$  and  $\rho_c$ , viscosities  $\mu_d$  and  $\mu_c$ , and interfacial tension,  $\sigma$ .

Certain linear dimensions,  $D$  and  $l$ , associated with the origin of the spray must also be included. In atomizers, these may be the orifice diameter and channel length; in a packed column, the nominal packing diameter or hydraulic radius, and the volume-surface ratio; in an ocean spray, the wave length and height.

Finally, must be considered an initial velocity,  $V$ , or velocity distribution  $f(\bar{V}, \delta_v)$ —e.g., the ejection velocity of a nozzle, or the Maxwellian distribution of velocities for colloid droplets.

Among the physical variables, various dimensionless groups may be constructed which represent various aspects of the behavior of the system. Hinze (6) suggests the following three independent groups:  $\rho_c/\rho_d$ ;  $\mu_d\sqrt{\sigma\rho_d D}$ ; and  $\rho_c V^2 D/\sigma$ —i.e., density ratio,  $Z$  number, and Weber number—to characterize breakup of a jet, hence composition of a spray. Hopkins (7) suggests the independent groups  $DV\rho_d/\mu_d$ ;  $\sigma/\mu_d V$ ;  $\alpha$ —i.e., Reynolds number, surface-tension group, and cone angle—and the dependent group  $\bar{x}_{32}\rho_d\sigma/\mu_d^2$ . Neither of these sets will be found adequate for a general correlation. But such a correlation may be reached by selecting groups characteristic of assumed mechanisms, then relating these to groups containing the three respective diameters which determine the distribution.

## CONCLUSIONS

The foregoing discussion has analyzed several size-distribution formulas for dispersed materials, and their applications to spray data. A modification of the log-probability equation, called the upper-limit equation, has been developed which has the following properties: It introduces a new and physically significant parameter,  $x_m$ , the maximum stable drop diameter. It fits available data accurately, probably within the experimental error in each case. It calculates the mean diameters accurately. It indicates the type of distribution function that may be derivable from mechanical principles.

## NOMENCLATURE

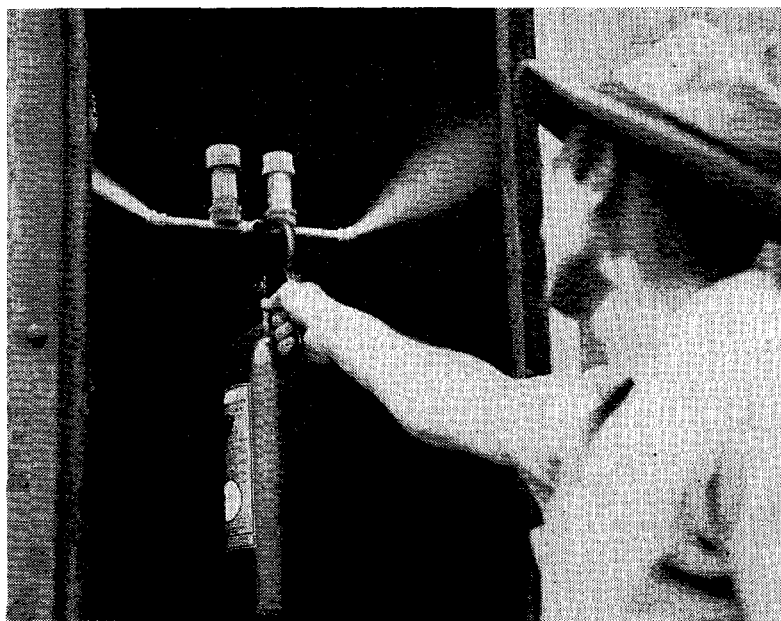
$a$	= parameter in upper-limit Equations 8.1, 9.03
$b$	= parameter in Equation 6.0
$B$	= constant in Equation 6.0
$c$	= a number (Equations 4.6, 5.31)
$d$	= derivative
$D$	= orifice diameter
$e$	= natural constant (2.718...)
$E$	= atomizing efficiency
$f$	= function
$k$	= proportionality factor (Equation 4.01)
$l$	= channel length
$m$	= mass of droplet (Equation 4.01)
$\bar{m}$	= mass of average droplet (Equation 4.03)
$n$	= numerical fraction of droplets having diameter $< x$
$p$	= subscript for mean diameter (Equation 4.4)
$\Delta p$	= pressure drop (Equation 4.04)
$q$	= subscript for mean diameter (Equation 4.4)
$s$	= exponent in Equation 8.1
$t$	= time
$u$	= dimensionless function of $x$ (Equation 9.5)
$U$	= variable of integration (Equations 5.31, 6.3, 7.4, 7.6)
$v$	= volume fraction of droplets having diameter $< x$
$V$	= initial or jet velocity
$\bar{V}$	= average velocity
$x$	= droplet diameter
$\bar{x}$	= size parameter in Equations 5.1, 7.2
$x_m$	= maximum droplet diameter
$x_0$	= minimum droplet diameter
$x_{10}$	= droplet diameter at $v = 0.10$
$x_{50}$	= droplet diameter at $v = 0.50$
$x_{90}$	= droplet diameter at $v = 0.90$
$\bar{x}_{qp}$	= mean diameter (Equation 4.4)
$y$	= dimensionless function of $x$ (Equations 7.2, 8.1, 9.1)
$\alpha$	= spray cone angle
$\Delta$	= increment (as in $\Delta n$ or $\Delta v$ )
$\Gamma$	= gamma function (Equation 5.31)

$\delta$	= size distribution parameter
$\delta_v$	= velocity distribution parameter
$\Theta$	= probability integral
$\mu_c$	= viscosity of continuous phase
$\mu_d$	= viscosity of droplet phase
$\pi$	= natural constant (3.141...)
$\rho_c$	= density of continuous phase
$\rho_d$	= density of droplet phase
$\sigma$	= interfacial tension
$\phi$	= a function (Equation 7.1)

## BIBLIOGRAPHY

- (1) Bevans, R. S., "Mathematical Expressions for Drop Size Distributions in Sprays," Conference on Fuel Sprays, University of Michigan, March 1949.
- (2) Coolidge, J. L., "Introduction to Mathematical Probability," London, Oxford University Press, 1925.
- (3) Cooper, F. A., *J. Soc. Chem. Ind.*, **56**, 447-53 T (1937).
- (4) De Juhasz, K. J., "Bibliography on Sprays" (Supplement issued in 1949), New York, Texas Co., 1948. Recommended to the attention of those further interested in the subject. Contains many references not given in the present list.
- (5) Harkins, W. D., and Beeman, N., *J. Am. Chem. Soc.*, **51**, 1674 (1929).
- (6) Hinze, J. O., "Mechanism of Disintegration of High-Speed Liquid Jets," paper presented at 6th International Congress for Applied Mechanics, Paris 1946.
- (7) Hopkins, J. L., "Size Distribution of Droplets in a Fuel Spray," Shell Petroleum Co. (London), *Rept. ICT/6* (1946).
- (8) Houghton, H. G., "Spray Nozzles," "Chemical Engineers' Handbook," J. H. Perry, 3rd ed., p. 1170, New York, McGraw-Hill Book Co., 1950.
- (9) Howell, W. E., *J. Meteorol.*, **6**, 134-49 (1949).
- (10) Johnstone, H. F., and Kleinschmidt, R. V., *Trans. Am. Inst. Chem. Engrs.*, **34**, 181-98 (1938).
- (11) Kolupaev, P. G., "Atomization of Heavy Fuel Oil," Sc.D. thesis in chemical engineering, Massachusetts Institute of Technology, 1941.
- (12) Lange, N. A., "Handbook of Chemistry," 7th ed., Sandusky, Ohio, Handbook Publishers, 1949.
- (13) Lee, D. W., "Effect of Nozzle Design and Operating Conditions on Atomization of Fuel Sprays," Natl. Advisory Comm. Aeronaut., *Tech. Rept.* **425** (1932).
- (14) Lewis, H. C., *et al.*, *IND. ENG. CHEM.*, **40**, 67-74 (1948).
- (15) Nukiyama, S., and Tanasawa, Y., *Trans. Soc. Mech. Engrs. (Japan)*, **4**, No. 14, 86 (1938); **5**, No. 15, 138 (1939); **6**, No. 18, 63, 68, No. 22, II-7, No. 23, II-8 (1940).
- (16) Pierce, N. C., "Efficiency of Hydraulic Nozzles for Atomization," M.S. thesis in chemical engineering, University of Illinois, 1937.
- (17) Rosin, P., and Rammler, E., *Z. Ver. deut. Ing.*, **71**, 1 (1927); *J. Inst. Fuel*, **7**, 29 (1933).
- (18) Rupe, J. H., "Technique for Investigation of Spray Characteristics of Constant Flow Nozzles," Third Symposium on Combustion, University of Wisconsin, 1948.

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### DDT Dust Is Propelled with Carbon Dioxide Gas Inside a Refrigerator Car Loaded with Potatoes

A new treatment involves the application of DDT with a special device in loaded cars or trucks and removes previous requirements that hatches of cars must be screened while they move through Japanese beetle infested areas. Two shots, one before and one after loading, give quick uniform distribution of the DDT.

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