Modeling drop size distributions

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Abstract

There is abundant literature discussing the prediction of a representative drop diameter in a spray. However, there are relatively few publications discussing prediction of a drop size distribution in sprays. In the present paper, we review the three available methods for modeling drop size distributions: the maximum entropy method, the discrete probability function method, and the empirical method. © 2002 Published by Elsevier Science Ltd.

Keywords: Atomization; Sprays; Drop size distribution; Modeling

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1. Introduction

Many processes use atomizers to deliver fluids. In certain applications, the drop size distribution must have a particular form (narrow, wide, few large drops, few small drops, etc.) for optimal operation. Knowledge of the effect of variations in fluid physical properties, atomizer geometry, and atomizer operating parameters on drop size distribution is crucial if control of the resulting distribution is to be realized. Three practical examples come to mind.

In automotive paint sprays transfer efficiency is known to depend on the width of the drop size distribution since small drops (diameters less than about 20 μm) are much more likely to follow air flow over a target surface than are big drops. Being able to predict the fraction of sub-20 μm drops would be very useful in this situation.

In waste incineration units burnout efficiency is known to depend on the width of the drop size distribution since large drops require considerably longer times to evaporate and burn to completion ($d^2$-law scaling). Being able to predict the fraction of large drops would certainly improve incinerator design.

In respirable sprays of pharmaceutical and/or medicinal products spray drug efficacy is known to depend on the width of the drop size distribution. Drops smaller than about 3 μm in diameter are ejected from the body during exhalation while those larger than about 10 μm in diameter are preferentially trapped in the respiratory system far
Nomenclature

\( a \) upper limit distribution parameter (dimensionless); Nukiyama–Tanasawa normalizing parameter (dimensionless); log–hyperbolic distribution normalizing parameter (dimensionless); three-parameter log–hyperbolic distribution parameter (dimensionless)

\( A_0 \) maximum amplitude of oscillation (dimensionless)

\( \text{ALR} \) air-to-liquid ratio by mass (dimensionless)

\( b \) Nukiyama–Tanasawa distribution parameter (dimensionless)

\( B \) modified Weber number (dimensionless)

\( C \) drop size constraint term (dimensionless)

\( d \) diameter (m)

\( D \) drop diameter (m)

\( \bar{D} \) representative diameter (m)

\( D_{10} \) arithmetic mean diameter (m)

\( D_{32} \) volume mean diameter (m)

\( D_{43} \) de Brouckere mean diameter (m)

\( E \) energy source term (dimensionless)

\( \text{erf} \) error function

\( f \) drop size distribution function (dimensionless); frequency (dimensionless)

\( F \) cumulative drop size distribution function (dimensionless)

\( F_n \) normalization factor (dimensionless)

\( g \) a priori drop size distribution function (dimensionless); function of state (dimensionless)

\( h \) height of liquid film (dimensionless)

\( I \) Kullback–Leibler number (dimensionless)

\( k \) Boltzmann’s constant (J/K); kinetic energy constraint (dimensionless)

\( K_1 \) modified Bessel function of the third kind and first order

\( K_p \) partition constraint (dimensionless)

\( m \) liquid mass flow rate (kg/s)

\( \dot{n} \) rate of drop production (number of drops/s)

\( M \) moment of a drop size distribution (dimensionless)

\( \dot{M} \) momentum source term (dimensionless)

\( p \) probability (dimensionless); Nukiyama–Tanasawa distribution parameter (dimensionless)

\( p_l \) fluctuating property (dimensionless)

\( q \) Rosin–Rammler distribution parameter (dimensionless); Nukiyama–Tanasawa distribution parameter (dimensionless)

\( r \) radius (m)

\( S \) Shannon entropy (dimensionless); source term (dimensionless)

\( \tilde{S} \) source term (dimensionless)

\( T_e \) unrelaxed axial tension (dimensionless)

\( \sigma_r \) interphase velocity slip ratio (dimensionless)

\( u_r \) drop velocity (dimensionless)

\( \bar{U}, \bar{u} \) average velocity (m/s)

\( U \) velocity (m/s)

\( v_r \) drop velocity (dimensionless)

\( \bar{v} \) average velocity (dimensionless)

\( \dot{V} \) average velocity at nozzle exit (m/s)

\( V \) velocity (m/s)

\( \nu \) drop volume (m³)

\( \text{We} \) modified Weber number (dimensionless)

Greek letters

\( \alpha \) modified wavenumber (dimensionless); log–hyperbolic distribution parameter (dimensionless)

\( \beta \) measure of inertial forces relative to surface tension forces (dimensionless); log–hyperbolic distribution parameter (dimensionless)

\( \gamma \) growth rate (dimensionless)

\( \Gamma \) gamma function

\( \delta_* \) upper limit distribution parameter (dimensionless); log–hyperbolic distribution parameter (dimensionless)

\( \Delta \) relative span factor (dimensionless)

\( \eta_0 \) zero shear viscosity (kg/m–s)

\( \eta_s \) solvent viscosity (kg/m–s)

\( \theta \) three-parameter log–hyperbolic distribution parameter (dimensionless)

\( \lambda \) Lagrange multiplier (dimensionless); characteristic time (s)

\( \lambda_1 \) relaxation time (s)

\( \lambda_2 \) retardation time (s)

\( \mu \) mean (dimensionless); log–hyperbolic distribution parameter (dimensionless); three-parameter log–hyperbolic distribution parameter (dimensionless)

\( \xi \) wavenumber (dimensionless)

\( \rho \) density (kg/m³)

\( \sigma \) standard deviation (dimensionless); surface tension (N/m)

\( \tau_x \) limit parameter of the log–normal distribution (dimensionless)

\( \tau_s \) sheet thickness (dimensionless)

\( \varphi_1 \) measure of viscoelasticity (dimensionless)

\( \varphi_2 \) measure of viscoelasticity (dimensionless)

\( \psi \) element of solution space (dimensionless)

Subscripts

\( 0 \) number distribution

\( 3 \) volume distribution

\( a \) air

\( \text{crit} \) critical

\( k \) kinetic

\( \text{ke} \) kinetic energy
upstream of their intended target (alveoli). Being able to determine a prior how many sub-3 and super-10 μm drops a particular device would produce would help reduce dosages.

The classical method of modeling drop size distributions is empirical: a curve is fit to data collected for a wide range of atomizer nozzles and operating conditions. Curves appearing frequently become the basis for ‘standard’ empirical distributions; given a sizable collection of such distributions (which would include Rosin–Rammler, Nukiyama–Tanasawa, log–normal, root–normal, log–hyperbolic, etc.) one can be fairly certain to locate a form that fits virtually any non-pathological data set. The problem with this approach is the difficulty of extrapolating the data to operating regimes outside the experimental range. Without additional experimentation, one can never be certain whether the extrapolated empirical correlation applies to the regime of interest; unfortunately, additional experimentation is often impractical, impossible, or prohibitively expensive.

As an alternative to the empirical approach, two analytical approaches to the problem of modeling drop size distribution have been developed in the past two decades: the maximum entropy (ME) method, and the discrete probability function (DPF) method. The ME method, pioneered by Sellens and Brzustowski [1] and Li and Tankin [2], views spray formation as a completely non-deterministic process than can be modeled using the principle of entropy maximization subject to a set of global constraints. The ME method assumes that the most likely drop size distribution is the one that maximizes an entropy function (either Shannon or the more general Bayes) under a set of physical constraints (e.g. conservation of spray mass, minimization of surface energy, etc.).

The most important part of applying the maximum entropy principle to predict drop size distribution is the correct formulation of the constraints. After approximately a decade of research, it appears that various formulations have converged to a commonly accepted form. The constraints contain ‘source terms’, which can be expressed in terms of representative diameters of the drop size distribution (e.g. the SMD, $D_{21}$, $D_{30}$, $D_{10}$, $D_{-10}$). It appears that at least two such diameters are needed to predict the drop size distribution. In principle, they can be obtained by other means (typically an instability analysis is used). However, an instability analysis can provide only one such representative diameter; at present, there are no means of obtaining two or more. This makes the maximum entropy method less useful for practical applications because at present experimental measurement is the only way of obtaining more than one representative diameter.

The DPF method, first applied to modeling drop size distributions in Newtonian sprays by Sovani et al. [3,4] and developed originally by Sivathanu and Gore [5], divides the spray formation process into deterministic and non-deterministic portions. It is assumed that spray formation involves a series of breakup stages of the initial fluid structure (flat sheet, annular sheet, jet, conical sheet, etc.). A fluid mechanic instability analysis (such as the ones of Rayleigh [6], Weber [7], Sterling and Sleicher [8], Goren and Gottlieb [9], or Panchagnula et al. [10]) can be used to describe the relevant breakup processes. It is important to emphasize that the DPF method is not tied to any particular instability analysis; any number of linear or non-linear analyses that acceptably describe the relevant breakup physics may be used.

Given a set of initial conditions (fluid physical properties and atomizer parameters) and a model of the breakup mechanism, it is postulated that the resulting drop size is uniquely determined. A drop size distribution is produced because the initial conditions fluctuate in a non-deterministic manner due to a variety of factors (such as turbulence, surface roughness, vortex shedding, mixture composition, etc.). The drop size distribution is computed by coupling a deterministic model that describes the formation of a single drop to the Sivathanu and Gore [5] DPF method.

2. Basics of spray characterization

Liquid atomization is the process of converting bulk fluid into a multitude of individual fragments (drops). Some means of describing the drops and obtaining quantitative information is necessary to evaluate and compare sprays.
There are two fundamental physical quantities associated with a given drop: its diameter and its velocity.

The set of drops produced by a given spray can be subdivided into classes, where each class consists of a drop whose diameter is within some range of a given diameter \( D \) (i.e. each class consists of drops whose diameters are in the range \([D - \Delta D, D + \Delta D]\)). By counting the number of drops in each class, it is possible to construct a histogram of the frequency of occurrence of a given class. The continuous version of the histogram is the probability density function (PDF) of the drop size, or the drop size distribution function.

The process above describes the number distribution; but it is also possible to construct area and volume distributions, where the area of the drop and the volume of the drop, respectively, are the variables of interest. From this point forward, the term ‘drop size distributions’ will be used to collectively refer to the family of possible distributions, and ‘a drop size distribution’ will be used to refer to any one of the possible distributions.

The velocity distribution function can be constructed in a similar manner. The drop size and velocity distribution functions are sufficient to completely characterize a given spray (we restrict our attention to isothermal, non-evaporating sprays where electrical charge is absent).

In general, the breakup of bulk fluid results in a spray where the sizes of drops are distributed between some non-zero minimum diameter and a finite maximum diameter. A finite maximum diameter exists because aerodynamic forces acting on a drop tend to break up a large drop into smaller ones. A non-zero minimum diameter exists because the cohesive surface tension forces increase as the drop size approaches zero, and the available aerodynamic forces cannot overcome the surface tension force. However, for convenience, most drop size distributions assume that the drop diameters range from zero to infinity. In this case, for the drop size distribution to be physically valid, it is necessary that the following conditions hold

\[
\lim_{D \to 0} \int_0^D f(D) \, dD = 0
\]

\[
\lim_{D \to \infty} \int_D^\infty f(D) \, dD = 0
\]

i.e. in a properly constructed PDF, the total number of drops below some minimum diameter and above some maximum diameter should be vanishingly small.

In general, a distribution must satisfy the following properties

\[
f(D) \geq 0
\]

\[
\int_0^\infty f(D) \, dD = 1
\]

i.e. the distributions must be positive and normalized.

Moments of a drop size distribution can be calculated in the following manner:

\[
M_n = \int_0^\infty D^n f(D) \, dD
\]

Moments of a valid drop size distribution should be non-zero and finite. They typically use positive integer values of \( n \), but there is no reason not to include negative and fractional values [11].

An empirical expression for a drop size distribution has a set of parameters that are adjusted to ‘fit’ the theoretical distribution to experimental data. It is desirable that the following two conditions hold:

1. The parameters of a given drop size distribution should be stable; that is, small variations in the input conditions of the spray should correspond to small variations in the values of the adjustable parameters.
2. The distribution should be unique; that is, there should exist one and only one set of parameter values to give a good fit to some particular experimental data.

It is often desirable to characterize a particular drop size distribution in terms of a single number, or a ‘representative drop diameter’. The polydisperse distribution is thus replaced with a monodisperse one, where the diameter of each drop is equal to the appropriate representative drop diameter. The representative size of the drops in a spray has been standardized by Mugele and Evans [12]. Various mean diameters can be calculated from the following equation:

\[
D_{pq} = \left( \frac{\int_0^\infty D^p f(D) \, dD}{\int_0^\infty D^q f(D) \, dD} \right)^{1/(p-q)}
\]

Again, \( p \) and \( q \) are typically positive integers, but fractional and negative mean diameters are not excluded from this general formulation. The following diameters are commonly used: \( D_{10} \), the arithmetic mean diameter, \( D_{30} \), the volume mean diameter, \( D_{32} \), the Sauter mean diameter, \( D_{43} \), the de Bruckere mean diameter. Many other mean diameters are possible using different values of \( p \) and \( q \).

As Sowa [11] points out, it is important to realize that a drop size distribution is a statistical entity and hence has a set of commonly accepted statistical moments. Four of the frequently used expressions that involve the statistical moments are the mean, the standard deviation, the coefficient of skewness, and the coefficient of kurtosis. Furthermore, the moments can be taken about the mean or about the origin. The Mugele and Evans [12] formula presented above produces expressions for mean diameters of a given distribution, but not all of them are the same as the statistical moments. For instance, the mean of the number distribution is \( D_{10} \), but the mean of the area distribution is \( D_{32} \), and the mean of the volume distribution is \( D_{43} \). These considerations will become important when we consider which representative
diameter should be used in deriving the drop size distribution using the maximum entropy principle.

There is a large body of work focused on modeling the mean drop size produced by a spray. The literature that focuses on modeling the primary drop size distribution, however, is somewhat limited. Modeling the primary drop size distribution is of paramount importance for two reasons:

1. In some industrial processes, primary atomization dominates and the effect of secondary atomization can be neglected. Then the primary drop size distribution is of direct interest.
2. The primary drop size distribution is needed as an input to models that predict the effect of secondary atomization. Then the primary drop size distribution is of secondary interest.

Presently, three methods have been used to model drop size distributions: the empirical method, the maximum entropy (ME) method, and the discrete probability function (DPF) method. These methods are discussed in detail in Sections 3–5, respectively. Their strengths and weaknesses will be presented, and recommendations made as to which approach is superior for various common applications.

3. The empirical method

The classical method of modeling drop size distributions is empirical: a curve is fit to data collected for a wide range of atomizer nozzles and operating conditions. Curves appearing frequently become the basis for ‘standard’ empirical distributions; given a sizeable collection of such distributions, one can be reasonably certain to locate a form that fits virtually any non-pathological data set. Some of the most commonly used distributions are listed below (the discussion here closely follows that of Palopouloski [13]). Masters [14] also provides a comprehensive list.

From this point forward, each symbol that represents a distribution function will have a subscript that indicates the type of the distribution the symbol represents. In particular, the subscript zero represents the number distribution and the subscript three represents the volume distribution. A lowercase symbol will be used to denote a marginal distribution, and an uppercase symbol will be used to denote a cumulative distribution.

The log–normal distribution is

$$f_0(D) = \frac{1}{D \ln \sigma_{LN} \sqrt{2\pi}} \exp \left\{ -\frac{1}{2} \left[ \frac{\ln(D/D_0)}{\ln \sigma_{LN}} \right]^2 \right\}$$  (7)

where $D_0$ represents the logarithmic mean size of the distribution and $\sigma_{LN}$ represents the width of the distribution ($\sigma_{LN} > 0$). Fig. 1 shows the typical shape of the distribution and illustrates the influence of changes in distribution parameters $D_0$ and $\sigma_{LN}$. It is interesting to note that the log–normal distribution arises when one considers the theoretical distribution that is produced by continuous random partitioning of a set of solid particles. This was first shown by Kolmogoroff [15], who was interested in the distribution produced by continuous grinding of solid particles.

The upper-limit distribution is a modification of the log–normal distribution in that a maximum drop size is introduced

$$f_3(D) = \frac{\delta D_{\text{max}}}{D} \exp \left\{ -\delta^2 \left[ \ln \left( \frac{aD}{D_{\text{max}} - D} \right) \right]^2 \right\}$$

$$a = \frac{D_{\text{max}}}{D}, \quad \delta = \frac{1}{\sqrt{2 \ln \sigma_{UL}}}$$  (8)

Here, $\sigma_{UL}$ represents the distribution width ($\sigma_{UL} > 0$), $D_{\text{max}}$ is the maximum drop diameter and $D$ is a representative diameter. Fig. 2 shows typical distributions and the effect of variation of distribution parameters $D$, $D_{\text{max}}$ and $\sigma_{UL}$.

The upper-limit distribution was first introduced by Mugele and Evans [12], who wanted to modify the log–normal distribution by specifying a maximum drop
diameter. The upper-limit distribution approaches the log-normal distribution as the maximum diameter tends to infinity.

The root-normal distribution is

\[ f_3(D) = \frac{1}{2\sigma_{RN}\sqrt{2\pi}D} \exp\left\{ -\frac{1}{2} \left[ \frac{\sqrt{D} - \sqrt{\bar{D}}}{\sigma_{RN}} \right]^2 \right\} \]  \hspace{1cm} (9)

where \( \sigma_{RN} > 0 \). \( \sigma_{RN} \) represents the width of the distribution, and \( \bar{D} \) is the mean diameter. Fig. 3 shows typical distributions and the effect of variation of distribution parameters \( \bar{D} \) and \( \sigma_{RN} \).

The root-normal distribution was proposed by Tate and Marshall [16] to express the volume distribution of drops in sprays. Note that the number distribution cannot be obtained from the volume distribution because of the resulting unphysical behavior at the lower end of the distribution (the PDF exhibits a gradient catastrophe near zero).

The Rosin–Rammler distribution is

\[ F_3(D) = 1 - \exp\left\{ -\left( \frac{D}{\bar{D}} \right)^p \right\} \]  \hspace{1cm} (10)

\[ f_3(D) = q\bar{D}^{-q}D^{q-1} \exp\left\{ -\left( \frac{D}{\bar{D}} \right)^p \right\} \]  \hspace{1cm} (11)

where \( \bar{D} \) represents the mean of the distribution, and \( q \)

Fig. 4. Typical Rosin–Rammler distribution. \( f_3(D) \) in \( \mu m^{-1} \), \( D \) in \( \mu m \).

Fig. 5. Typical Nukiyama–Tanasawa distribution. \( f_0(D) \) in \( \mu m^{-1} \), \( D \) in \( \mu m \).

indicates the value of the width of the distribution. Small values of \( q \) are associated with broad sprays, and large values of \( q \) are associated with narrow sprays. Fig. 4 shows typical distributions and the effect of variation of distribution parameters \( \bar{D} \) and \( q \). Note that if \( q < 3 \), the number distribution produces unphysical values (the probability becomes negative).

The Rosin–Rammler distribution was introduced by Rosin and Rammler [17] to describe the cumulative volume distribution of coal particles. Despite its shortcomings, the Rosin–Rammler distribution has been widely used in the spray literature, mainly because of its mathematical simplicity.

The Nukiyama–Tanasawa distribution is

\[ f_0(D) = aD^p \exp(-bD^q) \]  \hspace{1cm} (12)

where \( b, p, \) and \( q \) are adjustable parameters, and \( a \) is a normalizing constant. Sometimes \( p \) is taken to be fixed at two. The width of the distribution and the location of the mean is controlled by \( b, p, \) and \( q \). Fig. 5 shows typical distributions and the effect of variation of distribution parameters (\( p \) is taken to be fixed at 2). According to Palosposi [13], physically meaningful results are produced either if \( p > 1 \) and \( q > 0 \) or \( p < -4 \) and \( q < 0 \). The Nukiyama–Tanasawa distribution was introduced by Nukiyama and Tanasawa [18] to describe the number distribution of drops in sprays from a pneumatic atomizer.

The log–hyperbolic distribution, first applied to sprays by Bhatia et al. [19–21], is given by the following equation

\[ f_0(x; \alpha, \beta, \delta, \mu) = a(\alpha, \beta, \delta) \exp\left\{ -\alpha\sqrt{\delta^2 + (x - \mu)^2} + \beta(x - \mu) \right\} \]  \hspace{1cm} (13)

where \( a \) is a normalizing constant, given by

\[ a = \frac{\sqrt{\alpha^2 - \beta^2}}{2\alpha\delta K_1(\delta\sqrt{\alpha^2 - \beta^2})} \]  \hspace{1cm} (14)
and $A$ is given by

$$A = \frac{\sqrt{a^2 - (a^2 + 1)^2 \sin^2 \theta \cos^2 \theta}}{2a \sqrt{a^2 \cos^2 \theta - \sin^2 \theta} \sin \theta \cos \theta}$$

(17)

Recall that the logarithm of the log–hyperbolic distribution is a hyperbola. The geometrical meaning of the parameters is as follows: $a$ is the slope of the asymptotes, $\mu$ is the position of the vertex (the statistical mode), $\theta$ is the rotation angle of the asymptotes with respect to a fixed Cartesian coordinate system, and $K$ is a Bessel function of the third kind. The three-parameter log–hyperbolic distribution solves the stability problem at a cost of a slightly less accurate fit. Fig. 7 shows typical distributions and the effect of variation of distribution parameters ($a$, $\theta$, and $\mu$).

All of the empirical distributions outlined above were originally proposed to describe experimental data sets; hence, there is at least one experimental data set that is described well by a particular distribution. Moreover, some distributions give an excellent fit to a wide range of data.

The following question arises: which one of these distributions is best? Palosveski [13] used the $\chi^2$ test to analyze drop size distribution data from 22 sets provided by seven experimental studies; some of these data sets are widely used as benchmarks. Both number and volume distributions were considered.

The results of the $\chi^2$ test show that the Nukiyama–Tanasawa and the log–hyperbolic distribution functions provided the best fit. Upper-limit and log–normal distributions were reasonably accurate, but the fit was clearly inferior to that obtained from the Nukiyama–Tanasawa and the log–hyperbolic distribution functions. The Rosin–Rammler and three-parameter log–hyperbolic distributions performed poorly. Generally, it was found that the greater the number of adjustable parameters in the equation of the distribution

Fig. 6. Typical log–hyperbolic distribution. $f_0(D)$ in $\mu m^{-1}$, $D$ in $\mu m$. and $K_1$ is the modified Bessel function of the third kind and first order. The constraints on the parameters are as follows: $-\infty < \chi < \infty$, $a > 0$, $|\beta| < a$, $\delta > 0$, and $-\infty < \mu < \infty$.

The distribution derives its name from the fact that the logarithm of the probability distribution function is a hyperbola. $\delta$ is the scale parameter, $\mu$ is the location parameter, and the remaining two parameters describe the shape of the PDF. In addition, the slope of the left asymptote of the parabola is given by $a + \beta$, and the slope of the right asymptote is given by $-(a - \beta)$, and $\beta$ is the abscissa of the point of intersection. Fig. 6 shows typical distributions and the effect of variation of selected parameters ($a$, $\beta$, $\delta$, and $\mu$).

The log–hyperbolic distribution has been one of the most successful empirical distributions because it can be fit to a wide range of experimental data. However, it also has a series of significant drawbacks. First, it is difficult to work with mathematically. A fair amount of computation is required to obtain the parameters needed to fit the distribution to a particular data set (the method of maximum likelihood is usually employed for this). Second, the parameters of the log–hyperbolic distribution are mathematically unstable, which is undesirable.

The three-parameter log–hyperbolic distribution was created by Xu et al. [22] to address the problem of stability of the log–hyperbolic distribution

$$f_0(x) = A \exp\left\{-\frac{a}{a^2 \cos^2 \theta - \sin^2 \theta} \times \sqrt{(a^2 \cos^2 \theta - \sin^2 \theta)(x + \mu_0 - \mu)^2} \right\}$$

(15)

where $a$, $\theta$, and $\mu$ are shape parameters and $A$ is the normalizing constant. $\mu_0$ is given by

$$\mu_0 = -\frac{(a^2 + 1) \sin \theta \cos \theta}{\sqrt{a^2 - (a^2 + 1)^2 \sin^2 \theta \cos^2 \theta}}$$

(16)

Fig. 7. Typical three-parameter log–hyperbolic distribution. $f_0(D)$ in $\mu m^{-1}$, $D$ in $\mu m$. and $A$ is given by

$$A = \frac{\sqrt{a^2 - (a^2 + 1)^2 \sin^2 \theta \cos^2 \theta}}{2a \sqrt{a^2 \cos^2 \theta - \sin^2 \theta} \sin \theta \cos \theta}$$

(17)
function, the better the fit to experimental data. This is to be expected, given the greater degree of ‘shape freedom’ provided by distributions with a large number of parameters. Paloposki [13] also performed a study to determine the mathematical stability of parameters of the distributions. The Nuki–Tanasawa and the log–hyperbolic distribution functions both had problems with the stability of parameters. The log–normal distribution was more stable.

Paloposki’s [13] work underscores one of the problems with using empirical distributions—it appears that no single empirical distribution can accurately fit a large fraction of the available data sets. This fact necessitates the trial-and-error procedure of trying to fit several available empirical distributions to the data and locating one that best fits the particular set.

A further problem with the empirical approach is the difficulty of extrapolating the data to operating regimes outside the experimental range. Without additional experimentation, one can never be certain whether the extrapolated empirical correlation applies to the regime of interest; unfortunately, additional experimentation is often impractical, impossible, or prohibitively expensive.

4. Maximum entropy method

4.1. Joint size–velocity distributions

The maximum entropy method is one of the two theoretical approaches that have been used to predict drop size distributions. The approach taken in the maximum entropy method is similar to that used in statistical thermodynamics. Given a system consisting of an extremely large number of particles, it is difficult to predict the state of the system by applying physical laws to each individual particle and all possible interactions between particles. Even if it were possible to precisely predict the behavior of a single particle, it would be prohibitively expensive to do so for the entire system. However, for practical applications, all that is needed is some measure of the global state of the system (the temperature, for example), and it is not necessary to perform a detailed analysis of each individual particle to predict the system macroscopic state.

The maximum entropy model treats the atomization process as a ‘black box’ that transforms the bulk liquid into a system of drops having a particular drop size distribution associated with it. The details of the transformation are irrelevant.

While there is an infinite number of possible drop size distributions produced by an atomization process, the transformation process is subject to a set of physical constraints, such as the conservation of mass, energy, etc. The maximum entropy principle states that the most likely (or the least biased) drop size distribution is the one that maximizes the entropy of the system subject to the constraint(s). This distribution contains the least amount of information; the only information that it carries is that embodied in the constraints. Any other distribution would be biased, because it would contain more information than the physics of the situation provides.

The maximum entropy method proceeds as follows: first, a set of appropriate physical constraints is formulated. Then the distribution that maximizes the entropy of the system subject to the constraints is found.

According to Paloposki [13], Griffith [23] applied the theory of probability to the grinding of solid materials. This was apparently the first attempt to develop a particle size distribution function based on the concept of maximum entropy. Probability considerations led Griffith [23] to propose a particle size distribution function, which is a special case of the Nuki–Tanasawa distribution function and has commonly been called the Griffith comminution function.

In the late 1980s and early 1990s, Sellens and Brzustowski [1] and Li and Tankin [2] produced a series of publications in which they used maximum entropy theory to predict drop size distributions in sprays. More recently, Cousin et al. [24] have also examined the use of the maximum entropy principle to predict drop size distributions. Several other investigators have produced papers on the subject in the past decade [25–29].

The development of the method of maximum entropy to predict drop size distributions has raised a number of intriguing issues relating to the proper way of applying the maximum entropy principle to spray formation. We examine these in detail as we review the available publications.

In the sections that follow, we refer to number distributions, $f_0$. However, the subscript will be dropped for the sake of brevity. We will still refer to volume distributions by the subscript (i.e. $f_0$).

Sellens and Brzustowski [1] appear to have been the first to apply the maximum entropy principle (MEP) to predict drop size distributions in sprays. Subsequent discussion is largely based on a modification of their original ideas, so we use their formulation to examine the use of the MEP.

Sellens and Brzustowski [1] state that the Jaynes–Tribus maximum entropy formalism deals with the probabilities of discrete states whose behavior is constrained by the expected values of several quantities, which are functions of the set of states $j$

$$\sum p_j g_{ij} = \langle g_1 \rangle$$

$$\sum p_j g_{2j} = \langle g_2 \rangle$$

...(18)

$$\sum p_j g_{nj} = \langle g_n \rangle$$

Another constraint arises from the normalization of the set of probabilities

$$\sum p_j = 1$$

(19)

The probability distribution is obtained by maximizing the
Shannon entropy

\[ S = -k \sum p_j \ln p_j \]  

(20)

where \( k \) is Boltzmann’s constant. The general result of the application of the above equations to a particular problem is

\[ p_j = \exp(-\lambda_0 - \lambda_1 g_1 - \cdots - \lambda_n g_n) \]  

(21)

where the set of \( \lambda s \) is a collection of arbitrary Lagrange multipliers which must be evaluated for each particular solution. \( p \) is the probability of state \( j \), \( g(j) \) is some function of state evaluated at state \( j \), and \( \langle \rangle \) is the known expectation or average value of the function \( g \) over the entire system. The summation is over some range of \( j \) so that all possible states of the system are evaluated.

An equivalent continuous expression for the maximum entropy formalism can be obtained by considering the space to be continuous and replacing the sums by integrals. A uniform discretization of the solution space is assumed, because a non-uniform one would introduce arbitrary bias into the formulation. The continuous formulation is

\[ \int f g_n \, d\psi = \langle g_n \rangle \]  

(22)

where \( f \) is a probability density function, and \( \psi \) is an element of the solution space. The solution then becomes

\[ f = \exp(-\lambda_0 - \lambda_1 g_1 - \cdots - \lambda_n g_n) \]  

(23)

To apply the above formalism to the problem of modeling drop size distributions, Sellens and Brzustowski [1] considered the breakup of a liquid sheet having velocity \( V \) and thickness \( \tau \) in the vicinity of the breakup. Parameters are non-dimensionalized by the mass mean diameter \( D_{20} \) for lengths and the sheet velocity \( V \) for velocities. It is important to note here that \( D_{20} \) is not actually the mean statistical diameter of any drop size distribution (according to Sowa [11], it happens to be the cube root of the third moment about the origin of the number distribution). We discuss this point later. Dimensionless parameters are denoted by an asterisk. An element of the solution space is then the product of drop velocity and size: \( d\psi = d\delta s \, d\nu s \).

Initially, one would expect the conservation of energy to provide a single constraint. However, according to Sellens, this would leave out important information concerning the irreversibility of certain energy transformations and the prior knowledge of the energy distribution between various modes before breakup. In particular, directed translational kinetic energy is readily transformed into surface energy, but the reverse transformation from surface to directed translational energy is generally not possible (it is interesting to point out that such a transformation can indeed happen after a collision that does not lead to coalescence). Hence, the conservation of surface energy and the conservation of kinetic energy each provide a constraint.

It is assumed that the drops are spherical. The following conservation equations are obtained.

Normalisation

\[ \int f \, d\delta s \, d\nu s = 1 \]  

(24)

Conservation of mass

\[ \int f \delta s^3 \, d\delta s \, d\nu s = 1 + S_m \]  

(25)

Conservation of momentum

\[ \int f \delta s^2 \nu s \, d\delta s \, d\nu s = \frac{1}{3 \tau s} + S_s \]  

(26)

Conservation of surface energy

\[ \int f \delta s^2 \, d\delta s \, d\nu s = 1 + S_{ke} \]  

(27)

where \( \delta s \) is the dimensionless drop diameter, \( \nu s \) is the dimensionless drop velocity, \( \tau s \) is the dimensionless sheet thickness, and \( S_s \) are source terms. For distributions where \( D_{20} \) and \( D_{30} \) are known, \( \tau s \) can be calculated using the following relation, which is derived by applying the definition of \( D_{20} \) to the surface energy constraint

\[ \tau s = \frac{1}{3((D_{20}/D_{30})^2 - S_s)} \]  

(29)

The source terms are present to allow the modeling of processes such as evaporation and condensation, aerodynamic drag, and the irreversible conversion of surface and kinetic energy into other forms of energy. The value of the source terms used is not specified.

The solution is obtained by a form of the Newton–Raphson root finding technique. Iteration produces quick convergence in the \( \lambda \) values with little sensitivity to the initial guesses, provided that normalization is enforced at each iteration.

The solution for \( f \) as a PDF in size–velocity space yields an expression of the form

\[ f_0 = \exp\left(-\lambda_0 - \lambda_1 \delta s^2 - \lambda_2 \delta s^3 - \lambda_3 \delta s^3 \nu s - \lambda_4 \delta s^3 \nu s^2 - \lambda_5 \delta s^3 \nu s^3\right) \]  

(30)

which can be integrated over some maximum permitted velocity over the velocity dimension of the space to yield the size PDF. The maximum velocity does not have a substantial effect on the distribution, provided it is large enough. Sellens and Brzustowski [1] take the maximum velocity to be 2, meaning that the enforced ceiling on drop velocity is twice the initial velocity of the liquid sheet.
The resulting drop size distribution is

\[ f_0 = \left( \frac{\pi}{4 \lambda_4 \delta_4^4} \right)^{1/2} \left\{ \operatorname{erf} \left( \sqrt{\lambda_3 \delta_4^2 + \frac{\lambda_1}{2} \sqrt{\delta_4^2/\lambda_4}} \right) - \operatorname{erf} \left( \frac{\lambda_1}{2} \sqrt{\delta_4^2/\lambda_4} \right) \right\} \exp \left( -\lambda_0 - \lambda_1 \delta_4^2 - \left( \lambda_2 - \frac{\lambda_1^2}{4 \lambda_4} \right) \delta_4^2 \right) \]  

(31)

However, there is a problem with Eq. (31) in that it does not go to zero as the drop size goes to zero, as most experimental evidence seems to indicate. Fig. 8 shows the drop size distribution obtained using Sellens and Brzustowski’s [1] formulation.

Sellens and Brzustowski [1] conclude that the maximum entropy formulation has a fundamental analytical basis. The ME distribution derived in this work can be considered an ideal case. It represents the results of a loss-free atomization process in which liquid mass, momentum, surface energy, and kinetic energy are all conserved.

Sellens and Brzustowski’s [1] work provides insight to the issues that arise in attempting to use the MEP to predict drop size distributions in sprays. First, the process of formulating the constraints for the atomization process is not trivial. It is difficult to determine a priori which constraints should be included and which ones should be omitted. Sellens and Brzustowski’s [1] formulation also contains source terms, which are difficult to evaluate from first principles.

In their 1986 paper, Sellens and Brzustowski [30] clarify and extend the results of the 1985 [1] work. The constraints remain the same. The mass, kinetic energy, and surface energy source terms are set to zero, but the momentum source term is set to a small negative value. This produces an initial distribution in velocity. Sellens and Brzustowski [30] note that the momentum source does not arise from any physical argument. If the momentum source term is left equal to zero, the constraint equations for the momentum and kinetic energy taken together state that the variance of the velocity must be zero, which is clearly not the case for a real system. Hence, \( \tau_s \), the dimensionless sheet thickness, is used as the primary descriptive parameter of the distribution.

Sellens and Brzustowski [30] focus on the evolution of the velocity distribution as the spray proceeds downstream. They assume that the gas velocity is uniform and constant. Evaporation is neglected. The spray is assumed to be dilute, so that collisions between the droplets can be neglected. The effect of the gas on an individual droplet is modeled via a drag coefficient relation for solid spheres.

For any particular drop size, the velocity PDF is Gaussian in shape, and is in the following form

\[ f_0 = \exp \left( -\lambda_0 - \lambda_1 \nu - \lambda_2 \nu^2 \right) \]  

(32)

the \( \lambda \) values are initially determined from the solution for the size–velocity PDF, and are subsequently determined from the new downstream values of the mean and variance of the distribution. The downstream marching calculation indicates that the variance of the velocity distribution rapidly decreases in the downstream direction. This result suggests that, whatever the initial droplet velocity distribution, a substantial part of the identity of that initial distribution is lost over a distance, which may be short in comparison with the length of the spray.

In prior work [1,30], a problem of the negative momentum source term has arisen. To compensate, in the 1989 paper, Sellens [31] adds a new velocity constraint to the formulation; he now considers both the \( x \)-momentum and the \( y \)-momentum constraints. The new formulation is given below.

Normalizing

\[ \int \int f \, d\delta \, du \, dv = 1 \]  

(33)

Conservation of surface energy

\[ \int \int f \, d\delta \, du \, dv = \frac{1}{3 \tau} + S = \frac{D_{30}}{D_{32}} \]  

(34)

Conservation of mass

\[ \int \int f \, d\delta \, du \, dv = 1 \]  

(35)

Conservation of \( x \) momentum

\[ \int \int f \, d\delta \, du \, dv = \bar{u} + S_{uv} \]  

(36)

Conservation of \( y \) momentum

\[ \int \int f \, d\delta \, du \, dv = \bar{v} + S_{uv} \]  

(37)

Conservation of kinetic energy

\[ \int \int f \, d\delta \left( u_x^2 + v_y^2 \right) \, du \, dv = \frac{u_x^2}{\tau} + v_y^2 + S_{ke} \]  

(38)

where \( u_x \) is the \( x \) velocity, \( v_y \) is the \( y \) velocity, and the subscript \( s \) stands for the average value.

The maximum entropy formalism then yields the
following PDF

\[
f_0 = \exp \left( -\lambda_0 - \lambda_1 \delta^2 - \lambda_2 \delta^3 \right)
\]

\[
\times \left( -\lambda_4 \delta^4 \right) + \left( \lambda_5 \delta^5 \right) \left( u_x^2 + v_x^2 \right) \right)
\]

\[
(39)
\]

The addition of the \(y\)-momentum constraints has eliminated the need for the arbitrary small momentum source term that was needed in the 1D formulation to produce physically reasonable results.

When the conservation equations (Eqs. (33)–(38)) are applied, a solution may be obtained. However, the drop size PDF does not go to zero as the drop diameter approaches zero because the probability of small drop sizes is underestimated. In limiting the number of very small drops, the physical process at work is a limitation of the concentration of surface energy. With fixed values of surface tension, flow velocities, etc. it is unlikely that sufficient deformation energy will be expended on a given element of mass to reduce the drop size beyond a certain point. The critical quantity is the mean value of the surface–to–volume ratio of a drop. The additional constraint can be expressed as

\[
\text{Partition of surface energy}
\]

\[
\int \int f \delta^{-1} \, d \delta, \, du_x \, dv_x = K_p \]

(40)

where \(K_p\) expresses the strength of the partition constraint. The resulting PDF is then

\[
f_0 = \exp \left( -\lambda_0 - \lambda_1 \delta^2 - \lambda_2 \delta^3 \right)
\]

\[
- \lambda_4 \delta^4 \left( u_x^2 + v_x^2 \right) \right)
\]

\[
- \lambda_5 \delta^5 \]

To achieve a solution, it is necessary to estimate the parameters that appear in the constraint formulations. The sheet is modeled as having sinusoidal undulations, which are growing in amplitude as it moves downstream. The undulation amplitude is assumed to grow exponentially. An estimate of the mean velocities may be obtained by integrating over a complete undulation wavelength in the vicinity of the breakup length. The necessary sheet parameters were measured from high-speed photographs of the spray.

The mass source term is set to zero on the assumption of no evaporation. The source terms for momentum and kinetic energy were set to zero on the assumption that the acceleration due to environmental interactions is small within the breakup region.

A pressure-swirl atomizer was used for experiments, and a phase/Doppler system was used for measurements. The mean sheet velocity could not be measured, and was estimated from the measured large drop velocities and the angle of the spray cone. The value used in the surface energy constraint was taken directly from the spray measurements. A kinetic energy source term was used to obtain agreement with the data. Appropriate values of the partition coefficient were determined by trial and error. The following values were used:

\[
D_{3y}/D_{0y} = 0.827–0.885
\]

KE source: 3.5–5.5

\[
K_p = 1.25–1.40
\]

Sellens [31] compares the physical model predictions with the experimental data and reports that there is similarity among drop size distributions and that the measured drop velocity distribution is consistent with the predicted Gaussian profile.

In this paper [31], the problem with the arbitrary negative momentum source term has been fixed. However, the formulation has been complicated considerably, and new source terms have been introduced. Agreement with experimental data has been achieved by adjusting the values of the source terms. Hence, in this instance a case can be made that the ME approach is similar to curve fitting.

Unaware of Sellens and Brzustowski’s [1] work, Li and Tankin [2] independently used the MEP to predict the drop size distribution in sprays. Li and Tankin [2] did not consider the velocity of the drops. The constraints used are the following.

**Normalization**

\[
\sum_i P_i = 1
\]

(42)

**Conservation of mass**

\[
\sum_i P_i \rho_i \dot{n} = \dot{n}_i
\]

(43)

where \(P_i\) is the probability of finding a drop with volume \(y\) and density \(\rho\), \(\dot{n}\) is the total number of drops produced per unit time, and \(\dot{n}_i\) is the liquid mass flow rate.

In contrast to Sellens and Brzustowski [1], Li and Tankin [2] compute \(f_\rho\), the volume distribution function. They assume that the drops are spherical and convert the computed volume distribution

\[
f_\rho = \frac{3}{2} \left( \frac{\pi \rho \dot{n}}{m_i} \right)^2 \exp \left( -\frac{\pi \rho \dot{n}}{m_i} \right)
\]

(44)

into a number distribution function

\[
f_0 = \frac{3}{2} \left( \frac{\pi \rho \dot{n}}{m_i} \right)^2 \exp \left( -\frac{\pi \rho \dot{n}}{m_i} \right)
\]

(45)

Eq. (45) is a form of the Nukiyama–Tanasa function, where the distribution parameter is no longer variable, but equal to 3. The volume distribution is compared with experimental data from Lee et al. [32] and Tishkoff [33]. The agreement between the data and the predicted distribution is marginal (Fig. 9).

In this and many subsequent papers, Li and Tankin [2] formulate the constraints in terms of the drop volume, rather than drop size, yet they still maximize the Shannon entropy. As pointed out by van der Geld and Vermeer [34], and later
by Cousin et al. [24], this approach is inappropriate because it does not correctly formulate the maximum entropy principle.

In a subsequent paper, Li and Tankin [35] amended their prior work by adding momentum and energy constraints to the previous formulation. The volume formulation is again employed. The liquid velocity at the nozzle exit is taken as the reference velocity, and the drop volume is non-dimensionalized by \( D_{30} \)

\[
\bar{V} = \left( \frac{D}{D_{30}} \right)^3 = \bar{D}^3
\]  

(46)

Two additional parameters are defined in the formulation

\[
B = \frac{12}{\text{We}}, \quad \text{We} = \frac{\rho_l U_c^2 D_{30}}{\sigma}
\]  

(47)

Note that the Weber number is based on the liquid density, rather than the air density. The constraint equations are given below.

Normalization

\[
\int C d\bar{U} d\bar{D} = 1
\]  

(48)

Conservation of mass

\[
\int C d\bar{D}^3 d\bar{U} d\bar{D} = 1 + \bar{S}_m
\]  

(49)

Conservation of momentum

\[
\int C \bar{D}^3 \bar{U} d\bar{U} d\bar{D} = 1 + \bar{S}_{mv}
\]  

(50)

Conservation of energy

\[
\int C (\bar{D}^3 \bar{U}^2 + B \bar{D}^2) d\bar{U} d\bar{D} = 1 + \bar{S}_e
\]  

(51)

Note that there is a single energy conservation constraint, as

Fig. 9. Comparison of Li and Tankin’s [2] ME predictions with experimental data. Vertical bars indicate scatter in the data.
opposed to Sellens and Brzustowski’s [1] separate constraints for surface energy and kinetic energy.

Initially Li and Tankin [35] intended to set all of the source terms appearing in the constraint formulation to be zero. However, it was not possible to obtain a solution, so the momentum source term was set to $-0.05$. It is noted that the selected values for the minimum and the maximum velocity have little effect on the resulting PDF, provided that they span the velocity variations.

The resulting drop size–velocity joint distribution is

$$f = 3\bar{D}^2 \exp\left\{-\lambda_0 - \lambda_1 \bar{D}^3 - \lambda_2 \bar{D}^3 \bar{U} - \lambda_3 (\bar{D}^3 \bar{U}^2 + B\bar{D}^2)\right\}$$

and the drop size distributions obtained are

$$f_0 = \frac{3}{2} \left(\frac{\pi \bar{D}}{\lambda_3}\right)^{1/2} \{\text{erf}(x_{\text{max}}) - \text{erf}(x_{\text{min}})\} \exp\left\{-\lambda_0 - \lambda_3 B\bar{D}^2 - \left(\lambda_1 - \frac{\lambda_2}{4 \lambda_3}\right)\bar{D}^3\right\}$$

$$x_{\text{max}} = \left(\bar{U}_{\text{max}} + \frac{2 \lambda_2}{\lambda_3}\right)^{1/2}$$

$$x_{\text{min}} = \left(\bar{U}_{\text{min}} + \frac{2 \lambda_2}{\lambda_3}\right)^{1/2}$$

$$f_3 = \frac{3}{2} \left(\frac{\pi}{\lambda_3}\right)^{1/2} \bar{D}^{7/2} \{\text{erf}(x_{\text{max}}) - \text{erf}(x_{\text{min}})\} \exp\left\{-\lambda_0 - \lambda_3 B\bar{D}^2 - \left(\lambda_1 - \frac{\lambda_2}{4 \lambda_3}\right)\bar{D}^3\right\}$$

The authors note that the calculated number distribution goes to zero as the drop size goes to zero, and that there is a Gaussian distribution in velocity for any specific droplet size.

In their 1989 paper, Li and Tankin [36] made minor adjustments to the previous formulation. Several values of the momentum source term were used, namely $-0.005$, $-0.01$, $-0.025$ and $-0.05$. As the momentum loss increased, the probability density in the velocity space became broader, while the probability density in the drop size space became narrower. They noted that the selection of the limits of integration were inadequate and a better choice is $U_{\text{min}} = 0$, and $U_{\text{max}} = 3.5$.

Li et al. [37] continue their prior work [36] in a 1990 paper. The equations are the same as the ones in the prior paper [36]. Li et al. emphasize that the volume formulation results in a number drop size distribution that goes to zero as the drop size approaches zero; this is consistent with physical intuition and experimental observation.

Li et al. [37] also note that the velocity distribution is very narrow for large drops, and that the average value is slightly less than unity. For small drops, the distribution becomes increasingly broader in the velocity space. The drop size distributions for different momentum and energy source terms ($S_{\text{env}}$ and $S_3$) are virtually identical. The calculated values of $D_{25}/D_{30}$ are very close to unity and agree with experimental observations.

In their 1991 paper, Li et al. [38] conducted experiments to compare their ME predictions with experimental data. A hollow cone nozzle was used, spraying distilled water into a quiescent, saturated environment. Drops were sized using a two-color, four-beam phase Doppler particle analyzer (PDPA). Symmetry measurements were performed, which indicated that the spray had good symmetry about its axis. Hence, the size and velocity measurements, obtained from point measurements, can be integrated over the ring area associated with each point to yield a total spray measurement.

The formulation of the constraints is the same as that in the prior papers [35–37]. The control volume is assumed to extend from the nozzle exit to the breakup region, whereas Sellens’ [31] control volume extends from the start of the breakup region to the droplet region. The mass source is assumed to be zero. The momentum source term is estimated by ‘unfolding’ the conical liquid sheet and using a flat sheet drag correlation and computed to be $-0.017$. The energy source term is taken to be zero.

The agreement between the measured and predicted size distributions is reasonable. The experimental and predicted velocity distributions are not in agreement. This situation is improved somewhat when droplet drag is also accounted for.

The authors also noted that there is an inconsistency in the experimental portion of this study: the metered mass flow of water is an order of magnitude greater than the mass flow computed from the optical measurements. It is postulated that this could be due to (1) defining the optical probe measurement cross-section and (2) high rates of data rejection. This discrepancy could be responsible for the lack of agreement between theory and experiment.

In the 1991 paper, Chin et al. [39] used the ME formalism to predict the drop size distributions resulting from the breakup of a cylindrical liquid jet. A plain pressure orifice nozzle was used. Drops were sized using a PDPA.

Two modifications are made to the prior formulation of Li and Tankin [37]: (1) the surface and kinetic energy constraints are separated and (2) the control volume chosen is the region of liquid jet between the orifice plane and the location downstream where the drops are measured. The constraint set is given below.

Normalization

$$\int \int f \ d\bar{D} \ d\bar{U} = 1 \quad (56)$$

Conservation of mass

$$\int \int f \bar{D}^2 \ d\bar{D} \ d\bar{U} = 1 \quad (57)$$
Conservation of momentum
\[ \int \int f\bar{D}^3 \bar{U} \, d\bar{D} \, d\bar{U} = 1 + \bar{S}_{nv} \]  

(58)

Conservation of kinetic energy
\[ \int \int f\bar{D}^3 \bar{U}^2 \, d\bar{D} \, d\bar{U} = 1 + \bar{S}_{ke} \]  

(59)

Conservation of surface energy
\[ B \int \int f\bar{D}^2 \, d\bar{D} \, d\bar{U} = \frac{2B}{3D_{jet}} + \bar{S}_{se} \]  

(60)

where
\[ B = \frac{12}{We}, \quad We = \frac{\rho U_{jet}^2 D_{30}}{\sigma} \]  

(61)

The joint probability density function is of the following form
\[ f = 3\bar{D}^2 \exp\left\{ -\lambda_0 - \lambda_1 \bar{D}^3 - \lambda_2 \bar{D}^3 \bar{U} - \lambda_3 \bar{D}^3 \bar{D}^2 - \lambda_4 B \bar{D}^2 \right\} \]  

(62)

Chin et al. [39] noted that the shortcoming of Li and Tankin’s previous [37] formulation is that no information is provided as to how the total energy source is distributed between kinetic energy and surface energy (this echoes Sellsens and Brzustowski’s [1] earlier sentiment). Any combination of constant total energy source will result in the same probability density function. However, the kinetic energy term primarily affects the drop velocity distribution, while the surface energy source term primarily affects the drop size distribution, and hence should be considered separately. The need for separate constraints for surface and kinetic energy is illustrated in Fig. 10.

The distribution of drops in the present predictions is extremely sensitive to the assumed source term. A minor variation (less than 1%) changes the distribution dramatically. The source terms were estimated by measuring the relevant quantities directly from the drop size distribution
\[ \bar{S}_{nv} = \bar{U}_m - 1, \quad \bar{S}_{ke} = \left( \bar{U}_{rms}^2 + \bar{U}_{rms}^2 \right) - 1, \]  

(63)

where \( \bar{U}_m \) is the mean velocity of the jet and \( \bar{U}_{rms} \) is the root mean square velocity of the jet. These quantities, as well as \( D_{32} \) and \( D_{jet} \), are obtained from the measured drop size distributions. In making these source term estimates, it was assumed that all the drops have the same velocity, and that the velocity and size are uncoupled. It is generally accepted that drop size—velocity correlations exist near the spray. Chin et al. [39] do not mention whether such correlations are observed in the experimental data.

The agreement between the calculated and experimental data is acceptable, but there is a discrepancy between the peak of the predicted and measured distributions. A better fit was obtained by adjusting the source terms. However, since the distributions are extremely sensitive to the source terms, the adjustment needed was minor (less than 0.0005%). The result is shown in Fig. 11.

The flow rate measured by the PDPA and the actual flow rate agreed. When the previous formulation (combined kinetic energy terms) was used, the numerical calculations did not converge.

In their 1992 paper, Li and Tankin [40] raised the point that the experimental measurements should be integrated across the whole spray cross-section to yield the true distribution for all the drops produced by the spray, as opposed to making measurements in just one location, as was done by Sellsens [31].

In 1995, Chin et al. [41] used the ME formalism to predict bi-modal size distributions. Three components of velocity are considered. The maximum entropy model was simplified by postulating that the drop velocity at the peak probability is not a function of droplet size. This allows the simplification of the constraint set in that the momentum constraint is no longer required (however, the momentum source term is still needed to complete the calculation). The constraint set is given below.

Normalization
\[ \int \int f \, d\bar{D} \, d\bar{U} = 1 \]  

(64)

Conservation of mass
\[ \int \int f \bar{D}^3 \, d\bar{D} \, d\bar{U} = \bar{m} \]  

(65)

Conservation of surface energy
\[ \int \int Bf \bar{D}^2 \, d\bar{D} \, d\bar{U} = \bar{E}_s \]  

(66)

Conservation of kinetic energy
\[ \int \int f \bar{D}^3 \left( \sum \bar{U}_{rms}^2 \right) \, d\bar{D} \, d\bar{U} = \bar{E}_k \]  

(67)
where $B$ is $12/\text{We}$, the modified Weber number, and $U$ is the velocity vector. The resulting distribution is

$$f = 3D^2 \exp \left\{ -\lambda_0 - \lambda_1 D^3 - \lambda_2 B D^2 - \lambda_6 \sum_{p=1}^{3} \left( -2\tilde{M}_p \tilde{U}_p + \tilde{U}^2_p \right) D^3 \right\}$$

and the source terms are estimated as follows

$$\tilde{m} = 1, \quad \tilde{E}_s = \frac{B}{D^{3/2}}, \quad \tilde{M}_p = \tilde{U}_{m,p},$$

$$\tilde{E}_k = \sum (\tilde{U}^2_{m,p} + \tilde{U}^2_{var,p})$$

where $\tilde{U}_{m,p}$ is the mean velocity in each of the coordinate directions and $\tilde{U}_{var,p}$ is the variance of the mean velocity in each of the coordinate directions.

A hollow cone, non-swirl spray nozzle was used. Water was sprayed into quiescent, saturated air and drops sized using a PDP.

The source term estimates were: $E_s = 0.11, M_r = 0.61, M_g = 0.04, E_k = 1.1$. After adjusting the estimated values to $E_k = 0.87$ and $E_r = 0.12$, the computed drop size distribution curves agree reasonably well with the experiments, and exhibit a bimodal distribution.

In 1995, Chin et al. [42] conducted experiments to compare ME predictions for the breakup of a cylindrical liquid jet to experimental data. A plain pressure nozzle with a 50 $\mu$m orifice was used. A Kodak image analyzer was used to take pictures of the water drops. The apparatus operated in a regime where both primary and satellite drops were produced.

The satellite drops were nearly spherical, while the primary drops were not. An equivalent diameter was defined for the non-spherical drops (using mass conservation).

Two new constraints are added to the ME formulation: one related to the surface/volume ratio of small drops and the other related to the non-sphericity of large ones.

It was assumed that (1) the mean area/volume ratio for the group of small satellite drops varied inversely with $D/D_{30}$ and (2) the area/volume ratio for the group of large drops varied linearly with $D/D_{30}$. These assumptions were based on the observations of the experimental drop size distributions. The constraint set is given below.

Normalization

$$\int \int f \, d\tilde{D} \, d\tilde{U} = 1$$

Conservation of mass

$$\int \int \tilde{f} \tilde{D}^3 \, d\tilde{D} \, d\tilde{U} = 1$$

Conservation of momentum

$$\int \int \tilde{f} \tilde{D}^3 \tilde{U} \, d\tilde{D} \, d\tilde{U} = 1 + \tilde{S}_{mv}$$

Conservation of kinetic energy

$$\int \int \tilde{f} \tilde{D}^3 \tilde{U}^2 \, d\tilde{D} \, d\tilde{U} = 1 + \tilde{S}_{ke}$$

Conservation of surface energy

$$B \int \int \tilde{f} \tilde{D}^2 \, d\tilde{D} \, d\tilde{U} = \frac{2B}{3D_{jet}} + \tilde{S}_{se}$$

Small drop constraint

$$\int \int \frac{f}{D} \, d\tilde{D} \, d\tilde{U} = C_{-1}$$

Large drop constraint

$$\int \int f \tilde{D} \, d\tilde{D} \, d\tilde{U} = C_{+1}$$

The joint probability density function is of the following
form
\[ f = 3\tilde{D}^2 \exp\left\{ -\lambda_0 - \lambda_1 \tilde{D}^3 - \lambda_2 \tilde{D}^2 \tilde{U} - \lambda_3 \tilde{D} \tilde{U}^2 - \lambda_4 B \tilde{D}^2 \\
- \lambda_5 \tilde{D}^{+1} - \lambda_6 \tilde{D}^{-1} \right\} \quad (77) \]

Chin et al. [42] responded to points raised about the sensitivity of the drop size distribution function to the domain of integration of the joint function. They stated that the integration limits do not affect the drop size distribution.

The source terms used were: \( S_{mw} = 0.037 \), \( S_{ke} = 0.041 \), \( C_{-1} = 2.23 \) and \( C_{+1} = 0.76 \), and were obtained from measurements of the jet velocity as in prior work. However, \( C_s \)s were determined from direct measurement of the distribution. The agreement is reasonable, and a bimodal distribution resulted (Fig. 12).

The model was also used to predict the distribution produced by a pressure-swirl atomizer. Theory fit reasonably well with experiment after an appropriate adjustment of the source terms; in addition, the distribution was not bimodal. Hence, the introduction of the \( C \) terms does not necessarily lead to a bimodal distribution.

Finally, in 1999, Mitra and Li [43] combined a linear instability model to predict the breakup length and a nonlinear model to enhance the predictions. Boundary layer theory was used to evaluate the momentum and energy source terms. Prior models of Li et al. [37] were used to predict the drop size distribution. A parametric study was then carried out to demonstrate the effect of various parameters on the resulting drop size distribution. This is a follow-up to Cousin et al.’s [24] work that coupled a linear instability analysis with the ME method to predict drop size distributions. No experiments have been carried out by Mitra and Li [43], so no new information is produced.

### 4.2. Simplified models

The experience in using the MEP to predict joint drop size–velocity distributions has raised a number of interesting issues that should be examined. Some of these are related to the ability of the ME method to predict drop size distributions from first principles (without relying on experimental input or adjusted source terms), others are related to the proper application of the method.

Let us begin with the observation that the constraint equations always contain source terms. While some of them (mass, momentum, energy) can be justified on physical grounds, others (energy partition coefficient) are more difficult to defend. In either case, attempts to accurately estimate them a priori by appealing to first principles (using drag correlations, boundary layer theory, etc.) have been unsuccessful. The source terms always had to be adjusted a posteriori to ensure that predictions agree with the experimental data. Typically, the adjustment is made by referring to the experimental distribution. As such, the ME approach cannot be considered predictive in its current form. It is important to note that after an appropriate adjustment to the source terms has been made, the predicted drop size distributions agree with the data reasonably well.

This raises questions about the general utility of the ME method in modeling drop size distributions—if the source terms must be ‘tweaked’ by referring to the experimental distribution, why not just fit one of the empirical distribution functions to the data? The latter approach is considerably simpler and can produce a better fit. The situation is further exacerbated by the progressive complication of the constraint equations and the concomitant increase in the number of source terms (none of which can be adequately estimated from first principles).

A number of issues related to the proper application of the method have also been raised. Some of these are addressed in detail by the papers in this issue, others have never been conclusively resolved. The most important issues are listed below:

1. Is it appropriate to use the Shannon entropy (as opposed to the more general Bayesian entropy) if the drop volume, rather than size, is used as the primary variable in formulating the constraint equations?
2. Given that ME has its roots in thermodynamics, it is necessary to choose a control volume for the analysis. This raises the question “What is the appropriate control volume?” Is it necessary to consider the entire spray or can a representative portion be used instead? Where should the control volume be placed along the axis of the spray? (Note that if ME is based on some mean drop size the method essentially becomes a tool of statistical inference and no control volume is needed.)
3. Given a joint drop size–velocity distribution, are the limits of integration on the velocity space important if one wishes to obtain only the drop size distribution?
4. Should there be two separate energy constraints, one for surface energy, and one for kinetic energy, or a single energy constraint? Including conservation of surface energy as a constraint in a spray formation process is both unrealistic and unphysical since sprays are formed with the sole purpose of increasing liquid surface area. Therefore, with the exception of Rayleigh breakup of cylindrical jets (where the liquid velocity does not contribute to droplet formation) a separate surface energy constraint should not be included.

5. What is the proper way of formulating the constraints?

Faced with the ever-increasing complexity of the early formulations, Ahmadi and Sellens [44] were the first ones to consider using the ME method to predict the drop size distribution independently of the velocity distribution. In this paper, they reiterated the importance of conserving kinetic and surface energy separately to account for the lack of energy transfer paths between different modes of energy. The authors also stated that any ensemble of drops can be used for the purposes of the ME method; it is not necessary to apply the method to the entire spray.

When the velocity of the drops is not considered, a simplified set of constraint equations results.

Normalization

\[ \int_{\delta_{\text{min}}}^{\delta_{\text{max}}} \delta_s \, d\delta_s = 1 \]  

(78)

Conservation of surface energy

\[ \int_{\delta_{\text{min}}}^{\delta_{\text{max}}} \delta_s \, d\delta_s = \frac{D_{30}}{D_{32}} \]  

(79)

Conservation of mass

\[ \int_{\delta_{\text{min}}}^{\delta_{\text{max}}} \frac{\delta_s}{\delta_s} \, d\delta_s = 1 \]  

(80)

Partition of surface energy

\[ \int_{\delta_{\text{min}}}^{\delta_{\text{max}}} \delta_s^{-1} \, d\delta_s = K_p \]  

(81)

The limits of integration are normally taken to be zero and infinity, but are set to finite values outside the expected range of drop sizes in order to avoid difficulties in the numerical solution. The partition coefficient is necessary to ensure that the drop size distribution approaches zero as the drop size approaches zero.

The resulting PDF is

\[ f_0 = \exp\left( -\lambda_0 - \lambda_1 \delta_s^2 - \lambda_2 \delta_s^3 - \lambda_3 \delta_s^{-1} \right) \]  

(82)

The source terms used in previous work that employed the 2D formulation were used in the current formulation to test its validity. The resulting drop size distributions agreed closely with experiment (Fig. 13). This leads to the conclusion that the constraints on momentum and kinetic energy carry only velocity information.

The primary inputs to this model are values of \( D_{10}, D_{32}, \) ...

---

**Fig. 13.** Comparison of Ahmadi and Sellens' [44] simplified ME model to experimental data, the log–hyperbolic distribution, and Li and Tankin’s model.
and $K_p$. They were obtained from the experimental distribution, and a reasonable fit resulted.

Note that it is possible to cast $K_p$ in terms of a mean diameter

$$K_p = \int_0^\infty f \delta^{-1} \, d\delta = D_{30} \int_0^\infty f' \delta^{-1} \, d\delta = D_{30} D_{10} \tag{83}$$

The only parameters appearing in the model are then the mean diameters $D_{1-10}$, $D_{2-2}$, and $D_{3-0}$. This development suggests that the constraints can be expressed solely in terms of some set of representative diameters of a distribution. This is a vast improvement over the quasi-physical nature of the source terms appearing in prior formulations, since it is possible to use other methods to predict a representative diameter of a drop size distribution produced by an atomizer.

van der Geld and Vermeer [34] used a simplified formulation to investigate the effect of satellite drop formation. The model of Dombrowski and Johns [45] was used to describe the formation of the ligaments, which subsequently broke up into drops. The drops were characterized by primary drops and secondary, or satellite, drops. It was assumed that the primary drop distribution was Gaussian. The free parameters in the model were the primary drop distribution, parameters describing the satellite number density, and the ligament diameter. The constraint set included conservation of mass, conservation of surface energy, and normalization. The constraints were applied to unit cells (from which a single primary and a number of satellite drops form), rather than to the liquid sheet as a whole. Slightly bimodal distributions resulted (Fig. 14).

The authors were the first to address the applicability of using the drop volume rather than the drop diameter in the formulation. They point out that the Shannon entropy is not the proper measure if drop volume is used. This point is considered in detail in Cousin et al.’s work [24].

Cousin et al.’s work [24] is monumental in that it rigorously addresses the proper application of the Maximum entropy formalism. They point out that most of the studies carried out on the prediction of drop size distribution in sprays concentrate on modeling one type of distribution only (either number of volume). The ME approach should, in principle, be able to predict either distribution. That is, the volume-based formulation can be used, provided that the information that the drops are spherical is added.

The constraints that have been utilized in prior ME work typically involve the conservation of some quantity (mass, momentum, energy, etc.) that characterizes the process of atomization. Cousin et al. [24] advocate a new approach, in which the constraint is based on some representative diameter of the resulting distribution. The first approach is to calculate the number based distribution of an ensemble of spherical drops from one constraint only (other than normalization) that is based on the definition of some mean drop diameter

$$\int_0^\infty f_0 \, dD = 1, \quad \int_0^\infty f_0 D_0 \, dD = D_{\phi_0} \tag{84}$$

where $D_{\phi_0}$ is some mean diameter, but not necessarily the actual mean diameter $D_{\text{md}}$. This produces the following distribution

$$f_0 = \exp(-\lambda_0 - \lambda_1 D_0) \tag{85}$$

The mean diameter $D_{\text{md}}$ can be expressed in the following

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{drop_size_distribution.png}
\caption{Typical drop size distribution obtained by van der Geld and Vermeer [34].}
\end{figure}
manner using the standard definition

\[ D_{m0}^m = \int_0^\infty f_0 D^n \, dD \]  

(86)

Using this relation the volume distribution is given by

\[ f_3 = q^{(q-4)/q} \frac{D^3}{D_{q0}^3} \frac{\Gamma(4/q)}{\Gamma(4/(q-3))} \exp \left( -\frac{D^q}{qD_{q0}^q} \right) \]  

(87)

The parameter \( q \), the order of the constraint, is equivalent to the distribution parameter of a Rosin–Rammler distribution, and is therefore related to the width of the distribution. \( D_{q0} \) contains the same information as \( D_{m0} \), but in a form that is suitable for the maximum entropy formalism.

The second approach is to consider an ensemble of particles and apply the maximum entropy formalism to predict the volume distribution directly. The relation between the diameter of the particles and their volume must be introduced within the formalism to ensure that a meaningful volume distribution is produced. That is, if no information is specified besides the sphericity of the drops, the formalism will provide a volume distribution for an ensemble of spherical particles whose entropy is maximum, i.e. whose number-based distribution is uniform \( f_0(D) = \text{const.} \).

The information related to the sphericity of the particles is a characteristic of the variable of the distribution and not of the process. Therefore, it may not be expressed as a constraint. In such a situation, the maximum entropy principle requires us to define an a priori distribution \( g_3 \) and to minimize the directed divergence of the distribution sought, \( f_3 \), from the a priori distribution, such that the number distribution is the same as the a priori distribution in the absence of any other information.

A priori number distribution is uniform, so that the corresponding volume distribution for spherical drops is given by

\[ g_3 = 4D^3 \frac{4}{D_{\text{max}}^3} \]  

(88)

the directed divergence is then provided by the Kullback–Leibler number, given by

\[ I(f_3 : g_3) = \int_{D_{\text{min}}}^{D_{\text{max}}} f_3 \ln \left( \frac{f_3}{g_3} \right) \, dD \]  

(89)

which is also equivalent to a statistical measure of the nearness of two probability density functions. The Kullback–Leibler number should be minimized subject to a set of constraints characterizing additional information on the distribution sought. This formalism is also identical to maximizing the Bayesian entropy, which is a measure that takes the a priori distribution into account. Shannon’s entropy is a special case of the Bayesian entropy when the a priori probability distribution is the uniform one.

The constraint equations for the case where the formalism is applied to determine the volume distribution of a set of spherical drops are

\[ \int_0^\infty f_3 \, dD = 1, \quad \int_0^\infty f_3 D^{p-3} \, dD = D_{p3}^{p-3} \]  

(90)

and the volume distribution is

\[ f_3 = g_3 \exp \left( -\lambda_0 - \lambda_1 D_{p3}^{p-3} \right) \]  

(91)

The final form of the distribution is

\[ f_3 = \frac{p - 3}{\Gamma(4/(p-3))} \left( \frac{D^3}{D_{p-3,0}^3} \right) \exp \left( -\frac{D^{p-3}}{p - 3|D_{p-3,0}^{p-3}|} \right) \]  

(92)

where the order of the constraint, \( p - 3 \), is similar to the distribution parameter \( q \). If both approaches are applied to the same spray, both solutions should have at least the same distribution parameter, i.e. \( p - 3 = q \).

Substituting this relation into Eq. (92) leads to exactly the same distribution as the one obtained from the first approach. This shows, first, that a volume distribution cannot be predicted correctly if it does not contain the information related to the shape of the particles studied. It also shows that Shannon’s entropy cannot be used to predict volume distributions. This is related to the fact that a volume distribution is not exactly equivalent to a probability density function, since it also contains information on the nature of its elements.

To use the developments above to predict drop size distributions, it is first assumed that the \( D_{32} \) can be obtained in some other manner. It can be shown that the relation between the \( D_{32} \) and \( D_{q0} \) needed for the distribution is given by

\[ D_{32} = D_{q0} \sqrt[4]{\frac{\Gamma(4/q)}{\Gamma(3/q)}} \]  

(93)

Hence, the value of the mean diameter \( D_{q0} \) needed for the constraint can be determined, given the order of the constraint, \( q \). The resulting volume distribution is

\[ f_3 = \frac{D^3}{D_{30}^3} \exp(-\lambda_0 - \lambda_1 D) \]  

(94)

The experimental and theoretical distributions can be made to be reasonably close once the order \( q \) of the constraint is correctly estimated (Fig. 15). Cousin et al. [24] indicate that the order of the constraint may be constant for a given disintegration scheme. That is, the order of the constraint would be some constant value for pressure atomizers, another for pressure-swirl atomizers, and so on.

Cousin et al.’s [24] developments are a major step forward in using the ME approach to predict drop size distributions. However, while the source terms have been eliminated, the order of the constraint, \( q \), still remains a parameter that must be determined to predict the distribution. At this time, it is not clear whether it is possible to predict the parameter a priori.

Fig. 15. Comparison of Cousin et al.’s [24] ME predictions with experimental data.

work by noting that the parameter \( q \) is related to the relative span factor of a volume distribution, where the relative span factor is

\[
\Delta_3 = \frac{D_{0.9} - D_{0.1}}{D_{0.5}}
\]  

(95)

However, there is a discontinuity in \( q(\Delta_3) \) as the RSF approaches zero, so that the RSF approach would not work if delta-like distribution functions are considered. It also appears that the de Brouckere mean diameter \( D_{43} \), rather than \( D_{23} \), produces the best results. Mathematically, \( D_{43} \) is the mean diameter of the volume distribution (as pointed out by Sowa [11]), and hence the most appropriate statistical parameter. The results are illustrated in Fig. 16.

Malot and Dumouchel [27] extend Cousins et al.’s [24] work further. They point out that there are two limitations to the Cousin et al. procedure: nearly monodisperse sprays cannot be modeled, and the equation describes a symmetric distribution only for a certain value of \( q \). Experimental data is compared to the ME prediction in Fig. 17.

One of the features of the Cousin et al. [24] distribution is that the minimum diameter is always equal to zero. It is possible to shift the coordinate axis so that the Cousin et al. distribution can describe a symmetric one. The result is

\[
f_3 = q \frac{\Gamma^4(5/q)}{\Gamma^3(4/q)} \frac{(D - D_0)^3}{(D_{43} - D_0)\exp}
\]

\[
\times \left\{ \frac{\Gamma(5/q)}{\Gamma(4/q)} \left( \frac{D - D_0}{D_{43} - D_0} \right) \right\}
\]

(96)

This function belongs to the family of the generalized Gamma function. In the transformation of variables \( D_0 \) is

Fig. 16. Comparison of Boyaval and Dumouchel’s [25] ME predictions with experimental data. Prior work refers to Cousin et al. [24].

Fig. 17. Comparison of Malot and Dumouchel’s [27] ME predictions with experimental data.
the shift parameter—the distribution is shifted \( D_0 \) units to the right. A fit to experimental data occurs when \( D_0 \) is set to the minimum drop size that gives reasonable agreement.

It is interesting to note that the previous Cousin et al. [24] formulation provided for a number distribution that did not go to zero as the diameter went to zero. This behavior demonstrates the necessity of introducing information related to the small drop population in order to have a reliable number-based drop size distribution, as was done by Ahmadi and Sellens [44].

The maximum entropy principle was used to predict the drop size distribution in ultrasonic sprays by Dobry and Bolle [26]. Initially, only two constraints were used—normalization and mass conservation. The fit was not acceptable, so an energy constraint was added. The system of constraints then became

\[
\sum_{i=1}^{n} p_i = 1, \quad \sum_{i=1}^{n} p_i d_i^3 = 1, \quad \sum_{i=1}^{n} p_i d_i^2 = k, \quad (97)
\]

where \( f \) is the frequency of oscillation, \( A_0 \) is the maximum amplitude of oscillation, \( h \) is the height of the liquid film, and \( V \) is the average velocity of the drops. \( k \) can be estimated, but it turns out that the estimate is extremely poor (the actual values are twenty times greater than the estimated ones). Reasonable agreement is achieved once the values of \( k \) are adjusted accordingly (Fig. 18).

In summary, great strides were made in using the ME method to predict size distributions produced by sprays. The following guidelines for using the ME method were formulated:

1. It is important to use the proper measure when computing the drop size distribution. Shannon entropy can only be used if one is interested in the number distribution, while the Kullback–Leibler number is the appropriate measure if the volume distribution is desired.

2. The constraints should be formulated in terms of some representative diameters of the resulting distribution. The diameters must be obtained by some other means.

3. The representative diameters used should be some statistical measure of the distribution sought, i.e. the mean, variance, etc.

4. It appears that a single constraint that involves only one representative diameter results in unrealistic number distributions. A second constraint (the partition of surface energy, as used by Ahmadi and Sellens [44]), should be used to ensure that the number distribution approaches zero as the size of the drops approaches zero.

It appears that at least two representative diameters are required as inputs to the ME method to produce a realistic number distribution. It is generally possible to predict one representative diameter by using a stability analysis (either linear or non-linear). However, at present, it appears that it is not possible to predict more than one representative diameter, which seriously hampers the utility of ME as a method to predict drop size distributions from first principles.

5. The discrete probability function (DPF) approach

The maximum entropy method, discussed in Section 4, is a completely non-deterministic approach in that it is concerned only with the state of the system prior to atomization and the state of the system immediately after atomization. The details of the breakup mechanism are ignored. The DPF method, first used to predict drop size distributions by Sovani et al. [3], and pioneered by Sivathanu and Gore [5], divides the spray formation process into deterministic and non-deterministic portions. The deterministic portion of the model describes the breakup of the gross fluid structure, while the non-deterministic portion describes the influence of fluctuating initial condition on the resulting drop size distribution.

The DPF method assumes that spray formation involves a series of breakup stages of the initial fluid structure (which may be a flat sheet, an annular sheet, a conical sheet, etc.). The initial fluid structure separates into ligaments, which in turn break up into ligament fragments, which eventually collapse into drops. A fluid mechanic instability analysis is used to describe the relevant breakup processes. Current analyses are 1D, so a geometrical model is used to describe the separation of the sheet into ligaments. An instability analysis is used to describe the breakup of ligaments, and mass conservation is used to treat the collapse of ligament fragments into drops. It is important to emphasize that the DPF method is not tied to any particular instability analysis, linear or non-linear, 1D or 2D; any number analyses that model the relevant breakup problem may be used.

The breakup model described above is deterministic. For a given set of initial conditions the model predicts the
production of a drop of some specific diameter. A direct application of the model predicts a monodisperse spray. Since most sprays are not monodisperse, it is necessary to introduce a non-deterministic component to the overall model.

The DPF formalism rests on two postulates: (1) a set of initial conditions and a gross fluid structure breakup model predicts the production of a unique drop of some diameter; (2) a drop size distribution is produced because the initial conditions fluctuate. In practical atomizers, the fluctuations can be due to a number of factors, some of which are vibration of the atomizer, fluctuation in fluid delivery rate, fluctuation in exit velocity (due to turbulence, vortex shedding, etc.), fluctuation in fluid physical properties (if the liquid is non-homogeneous), etc. The list above is by no means exhaustive.

Experimental support for the second postulate of the dynamical method is provided by comparisons of drop size distributions produced by two identical atomizers using the same working fluid. In the first atomizer, the mass flow rate of the working fluid is nearly constant. In the second atomizer, the mass flow rate is irregular. The second atomizer produces a drop size distribution that is much wider than that produced by the first atomizer, which indicates that the fluctuations in initial conditions are responsible (at least to some extent) for the variation in the resulting drop sizes.

The DPF method requires the probability density function (PDF) of the fluctuating parameter as an input. The initial conditions fluctuate in a non-deterministic manner. The DPF method provides a computationally efficient means of coupling the deterministic breakup model with the non-deterministic fluctuations of initial conditions and enables one to predict the resulting drop size distribution.

The fluctuating initial conditions can be described by a continuous probability density function (the input PDF). The input PDF is then discretized into an integral number of bins (where the number of subdivisions depends on the desired resolution). Each bin is characterized by two values: the value of the initial condition at the midpoint of the bin (bin value) and the associated probability (bin probability). Hence, each bin contains a set of unique initial conditions.

The initial conditions of each bin are used in the drop formation model to predict the drop size corresponding to a given bin. Since the breakup model is deterministic, the probability of occurrence of the drop size given a set of initial conditions is the same as the probability of occurrence of the initial conditions. Essentially, the DPF method defines a transform from the input PDF to the drop size distribution PDF, where the bin values are transformed while the bin probabilities are kept fixed. The breakup model serves as the transform function (Fig. 19).

The technique outlined above uses a 1D PDF, that is, only one initial condition fluctuates while the others are held fixed. The DPF method can be readily extended into higher dimensional PDFs, where several initial conditions fluctuate simultaneously. The procedure is virtually identical to that used in the 1D PDF. The only difference is that the probability of occurrence of a given drop size bin is equal to either the product of the probabilities of each input PDF (if the initial conditions are independent) or to the product of probabilities and correlation factors (if the initial conditions are not independent).

Work using the DPF method has been performed by Sovani et al. [3] for Newtonian fluids and Babinsky and Sojka [46] for non-Newtonian fluids. Recall that the input PDF of the fluctuating property is required in order to
predict a drop size distribution a priori. Unfortunately, no experimentally measured PDFs of the relevant fluctuating properties are available at this time. Hence, it is impossible to compare the theoretical predictions and experimental measurements in a meaningful manner. Therefore, both papers used a parametric approach to investigate the influence of fluctuating properties on the drop size distribution.

In their paper, Sovani et al. [3] applied the DPF method to effervescent atomization of Newtonian fluids. The geometrical sub-model of Lund et al. [48] was used to predict the ligament diameter. The Sterling and Steicher [8] linear stability analysis was used to predict the size of a typical ligament fragment upon breakup. The normal distribution was used as the input PDF for liquid exit velocity and liquid physical properties. Sovani et al. [3] performed a series of parametric studies to determine the effect various fluctuating input parameters have on the resulting drop size distribution. They determined that fluctuations in the relative velocity have the greatest effect on the resulting drop size distribution, while fluctuations in fluid physical properties have a negligible effect. A typical drop size distribution is shown in Fig. 20.

In a subsequent paper, Sovani et al. [4] used the DPF method to study the effect of several input parameters fluctuating simultaneously. A 2D normal distribution was used as the input PDF, and the effect of both velocity and physical property fluctuations was considered. The results indicate that small (1%) fluctuations in fluid physical properties have no effect on the drop size distribution. Larger fluctuations in fluid physical properties widen the drop size distribution slightly, but only at low velocities. As the velocities increase, the effect of fluctuation in fluid physical properties diminishes.

Babinsky and Sojka [46,47] applied the DPF method to effervescent atomization of non-Newtonian (viscoelastic) fluids. The geometrical model of Lund et al. [48,49] was again used to predict the ligament diameter. Studies [50,51], indicate that the breakup of a viscoelastic liquid jet is markedly different from that of a Newtonian jet because of the unrelaxed axial tension that is present as the fluid is exiting the orifice. Hence, the Goren and Gottlieb [9] linear instability analysis of a viscoelastic liquid jet with simplifications provided by Bousfield [52] was used to predict the size of a typical ligament fragment. One fluctuating property and two simultaneously fluctuating properties were considered. The three-parameter log-normal distribution was used as the input PDF. A typical drop size distribution is shown in Fig. 21.

Results indicate the fluctuations in ALR and interphase velocity slip ratio have the most effect on the drop size distribution, while fluctuations in fluid physical properties have a much smaller effect.

The results for two simultaneously fluctuating parameters have two distinct regions. When the level of fluctuation of both parameters is low, the influence on the distribution can be deduced simply by adding the influence of each fluctuation considered separately. For example, if fluctuations in ALR alone widen the distribution by 5% and fluctuations in relaxation time alone widen the distribution by 3%, then the combined effect of simultaneous fluctuations in both ALR and relaxation time produce an 8% overall widening. On the other hand, when the fluctuation level of one parameter is

![Fig. 20. Typical drop size distributions obtained by Sovani et al. [3].](image-url)
large, the effect of the second fluctuating parameter on the overall distribution is negligible.

The DPF method has a number of limitations, which are outlined below:

1. It is limited to primary atomization.
2. To date, the DPF method has not been compared to experimental data because of the difficulties involved in obtaining experimental measurements of the quantities of interest.
3. The method requires a ligament formation sub-model. The computational complexity of the method greatly increases if a non-linear multi-dimensional breakup model is required to adequately model physics of the breakup process. It is crucial to emphasize again, that the DPF method is not tied to any particular ligament formation sub-model; any number of instability analyses, linear or non-linear, single or multi-dimensional, can be used.
4. The DPF method requires PDFs of fluctuating initial conditions. Since these are likely to depend on atomizer geometry and operating conditions, the PDFs must be

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Table 1

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<th>ME</th>
<th>DPF</th>
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<tr>
<td>Inputs needed (in addition to</td>
<td>At least two representative drop diameters</td>
<td>Probability density function of the fluctuating parameter</td>
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<td>fluid physical properties and</td>
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<td>atomizer parameters)</td>
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<tr>
<td>Inputs that can be currently</td>
<td>One representative diameter using an instability analysis</td>
<td>No. No experimental PDFs exist. Comparison with experiments is meaningless without a correct input PDF</td>
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<td>computed</td>
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<td>Can the lack of necessary</td>
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<td>inputs be rectified?</td>
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<tr>
<td>Produced a priori</td>
<td>No. Agreement achieved only after an adjustment of source terms or obtaining the representative diameter by direct measurement of the experimental distribution</td>
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<td>predictions that agree with</td>
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<td>experiments?</td>
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<tr>
<td>Computational algorithm</td>
<td>Reasonable. Requires the minimization of a multi-dimensional potential function. The number of dimensions is proportional to the number of constraints</td>
<td>Simple if CFD is not involved and 1D fluid breakup model is used. Requires the minimization of a one dimensional function. The complexity of the overall method is directly proportional to the complexity of the CFD code and the complexity of the fluid breakup model (which will increase if 2D breakup models are necessary)</td>
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<td>complexity</td>
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<tr>
<td>Can be applied to fluids with</td>
<td>Unknown. Unclear whether it would be possible to formulate the appropriate constraint</td>
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<td>complex rheology or different atomization modes?</td>
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determined for each particular situation. Unless a theoretical method is found to predict the appropriate PDFs, they will have to be experimentally determined, which would limit the predictive utility of the DPF method.

6. Summary

Three methods of modeling drop size distributions, empirical, maximum entropy (ME) and discrete probability function (DPF), were reviewed.

The empirical method is extremely flexible; it can be used to model virtually any non-pathological data set. However, its predictive power is severely limited. If it is found that an atomizer operating under certain operating conditions produces a drop size distribution that is described by a given empirical distribution, it is possible to establish an empirical relationship between atomizer geometry and operating conditions and the parameters of the distribution. This can then be applied to predict the drop size distribution produced by a different atomizer operating in a different regime. However, there is no guarantee that the empirical relationships (or the empirical distribution function) will remain the same.

It is difficult to select an empirical distribution function that fits a wide range of actual drop size distributions. Those that do work well (such as the log–hyperbolic distribution) suffer from numerical stability problems and are difficult to handle mathematically. The proliferation of parameters that invariably accompanies flexible distribution functions is concomitant with relatively complex computer algorithms required to estimate the value of the parameters. The difficulties with the empirical method have prompted the development of other techniques for predicting drop size distributions.

The maximum entropy method has worked well in a number of situations; it appears to be a reasonable approach to modeling drop size distributions, and, in particular, would be useful for processes dominated by secondary atomization where the breakup physics are highly stochastic in nature. The ME method focuses only on the initial and final stages of a process, and the exact mechanism is ignored. In the concrete case of predicting drop size distributions, only the initial conditions and the resulting drop distributions are considered; the details of the breakup process are irrelevant. Experience with using the ME technique has shown that the method requires more information about the resulting drop size distribution than is possible to predict using other means. Specifically, it appears that at least two representative diameters are required to accurately predict the drop size distribution, while currently only one representative diameter can be obtained by using an instability analysis. Attempts to remedy this situation have required modeling the details of the breakup process, which should be ignored in a proper application of the ME method.

The DPF method attempts to resolve difficulties encoun-
tered by the ME method by mathematically describing the details of the breakup process and introducing probabilistic effects to account for the randomness inherent in breakup. The method can be adapted to a variety of atomizer configurations and operating conditions simply by selecting an appropriate instability model for breakup. However, the method is limited to primary atomization. In atomization processes in which secondary atomization dominates (pressure, airblast, air-assist, twin-fluid, Diesel, paint spray, etc.) the DPF method is inappropriate. In these cases, the ME method, which does not model the breakup process, is more appropriate.

The DPF method has not been validated to date because of the difficulties involved in obtaining experimental data. In addition, the DPF method requires the PDF of the fluctuating initial conditions as input. If the PDF is obtained experimentally, the DPF method loses its predictive power, since PDFs are likely to vary under different conditions.

Future work on the DPF method will involve the use of CFD to predict the input PDFs. While the state of the art in CFD has not yet reached a point where this can be accomplished, it will probably be possible in the future.

Out of the three available methods for predicting drop size distributions, only the ME and the DPF methods can be considered ‘predictive’. Table 1 summarizes the advantages and disadvantages of these two methods and represents the authors’ opinions.

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