ABSTRACT

In sprays theoretical characterization, a difficult step is to find the most likely drops size probability distribution with respect to the known mean diameter and the type of atomization. The classical procedure consists of fitting an analytical function (for ex. Rosin-Rammler) on experimental data. Another more logical and more physical meaningful approach is to apply the maximum entropy formalism (M.E.F.), a widely used method which allows to choose among all the possible probability distributions the most suitable to the available knowledge about the phenomena. The classical M.E.F., states that, when given some information about a stochastic process (for ex., given the mean diameter of the spray), the probability distribution that agrees the best with that information is obtained by maximizing Shannon’s entropy \( S(p) = k \sum_i p_i \ln p_i \) subject to the given constraints (written in form of conservation laws). Shannon's entropy, besides other required properties, is additive. Additivity is not a necessary condition for a function to be a measure of uncertainty of a probability distribution. Several authors suggested some other non-additive (non-extensive) forms of entropy. Tsallis introduced a generalized pseudo-additive function \( F(p) = k(1 - \sum_i p_i^{q-1}) \). Tsallis entropy is applied to an increasing number of physical problems. It has been showed that generalized entropy predicts well physical systems involving long-range interactions (anomalous diffusion, granular systems,…). Violent break-up in sprays, fragmentation with high energies, leads to long-range interactions between liquid elements. Spraying regimes where high energies are involved (high pressure or high velocity) may be well depicted by power law distributions as obtained from the Tsallis entropy. An atomizer working at low pressure will follow an exponential Shannon law but when increasing pressure the distribution tends to power Tsallis law, revealing scaling. Tsallis nonextensivity parameter \( q \) may also be used in order to classify break-up regimes in sprays.

INTRODUCTION

The common approach used to determine droplets size distribution in spray research is based on extensive measurements followed by a best-fit procedure in order to find an analytical probability density function (pdf). The choice of the suitable pdf (Rosin-Rammler, log-normal, log-Boltzmann, etc…) is only based on best fit: it cannot be founded upon neither physical nor logical basis. This common approach provides no physical link between the atomization parameters (type of atomizer, liquid properties) and the resulting spray characteristics.

Provide a probability distribution function closely related to the physics of atomization can be achieved by means of the maximum entropy formalism (MEF) proposed by Jaynes [1]. This statistical widely used method allows to choose among all the possible probability distributions the most suitable one with respect to to the available knowledge about the phenomena. The MEF is applied to sprays since 1986 (precursors are Sellens and Bruztowski [2] and Li and Tankin [3]) in order to predict droplets size and, eventually, velocity probability distributions. The MEF states that, when given some information about a stochastic process (for ex., given the mean diameter of the spray), the probability distribution that agrees the best with that information is obtained by maximizing Shannon’s information entropy (\( S(p) = k \sum_i p_i \ln p_i \)) subject to constraints describing the available knowledge. The constraints are usually physical conservation laws (mass, momentum, energy). The same formalism is widely used in other fields such as astronomy, biological systems, financial data and always yields exponential type distributions. As many of the empirical laws already used in atomization are exponential laws, the MEF based on Shannon entropy suits well spray prediction. But nature does not exhibit only exponential laws and power laws may occur in phenomena like anomalous diffusion, fragmentation, granular matter, self-gravitating systems, etc.. As an attempt to theoretically solve such "anomalies", Tsallis [4] proposed in 1988 a new form of entropy which fulfills all the requirements of an entropy function but is not additive as the Shannon entropy. Additivity is not a necessary condition for a function to be an entropy and many types of pseudo or non-additive entropy expressions were proposed in past years [5]. Tsallis entropy (\( T \)) is a generalized entropy; Shannon entropy is a limiting case of it.
Tsallis entropy has found many applications in past years mainly in phenomena exhibiting long-range interactions, fractallity or high-energy levels. A review of successful applications is presented in reference [6]. As MEF based on Tsallis entropy has been shown to predict well fragmentation processes, Sotolongo-Costa & al. [7] suggested that it could be also useful in droplets high-energy break-up (as power law rather than exponential distribution are reported in application such as blasting oil drops).

Other types of atomization might exhibit power laws, such as airblast atomizers as they involve much higher energy inputs per unit atomized liquid mass than other sprays. Rizk & Lefebvre [8] observed that the classical Rosin-Rammelr exponential distribution does not fit airblast sprays droplet distribution. They suggested a modified Rosin-Rammelr where original variables are replaced by their logarithms. In terms of the original variables this will give a power law distribution.

The log-Boltzman distribution which fits successfully air-assisted sprays, as reported by Jang & al. [9] and Lee & al. [10] is also a power law.

Furthermore, in their exhaustive study of airblast atomizers, Harari and Sher [11] identified two distinct atomization regimes yielding a bimodal distribution. Droplets form both by a classical hydrodynamic instability mechanism and by direct interaction of a toroidal liquid vortex with the orthogonal high-velocity air stream. If a maximum entropy approach is applied to airblast atomizers, this latter high-energy violent break-up mechanism requires the Tsallis entropy formulation, the exponent from MEF Tsallis distribution reflecting the rate of input energy to the liquid mass.

Maximum entropy formalism based on Shannon entropy suits well ultrasonic spray prediction [12] because low-power atomizers produce an uniform unimodal exponential distribution. In order to increase the maximum flow rate higher energy atomizers are proposed [13]. These atomizers tend to produce a bimodal distribution. Due to higher energy input two different break-up mechanisms might occur (as for the airblast atomizer). Part of the liquid is sprayed through a wave instability mechanism as described by Sindayihebura & Bolle [14] and the rest is violently broken-up by cavitation inside the feeding channel of the atomizer. An attempt is made hereafter to predict high-energy ultrasonic spray droplets size distribution using maximum entropy formalism based on both Shannon and Tsallis form of information entropy thus trying to express the differences between the two atomization regimes.

This approach could first be used in bimodal spray characterization and, with further experience, could lead to universal spray prediction through the correlation between input energy-liquid mass ratio and Tsallis power law exponent.

**MAXIMUM ENTROPY THEORY**

The spray is a set of non-uniform droplets which sizes may be arranged into $n$ diameter classes. A quantitative criteria is defined in order to determine the part of the spray included in each size range. If this criteria is the number of the droplets inside a diameter class, the size distribution of the spray is a probability distribution in a statistical sense. The number distribution is not very useful for spray applications. Atomization is achieved in order to increase the free surface of a given liquid mass and/or to supply reduced volume parts of that mass. So, for practical purposes, the size distribution is often expressed in terms of surface or volume of droplets into each diameter class. Those surface or volume distributions are not "statistically correct" probability distributions. They are only distributions of given quantities (surface, volume) as a function of the droplet diameter. So, even if for comparison reasons the presented distributions are volume distributions, one should keep in mind that MEF yields number distributions.

The first step in spray prediction is to find the mean diameter by theoretical analysis or experimental trials. Once the mean diameter is known, the next step is to find the most likely probability distribution with respect to the calculated mean diameter and with the type of atomization. Unfortunately an infinity of size distributions may fit to the unique calculated mean diameter. The choice of the "right" distribution should rely on the atomization characteristics (break-up mechanism, fluid properties...). This is achieved by using the maximum entropy formalism. The MEF states that, when given some information about a statistic process (for ex., given the mean diameter of the spray) the probability distribution that agrees the best with that information is obtained by maximizing the information entropy subject to constraints describing the available knowledge:

$$S(p_i) = \sum_{i=1}^{n} p_i \ln p_i$$

Tsallis entropy: $$T(p_i) = \frac{1 - \sum_{i=1}^{n} p_i^q}{q - 1}$$

In the limiting case of $q=1$ Tsallis entropy reduces to Shannon expression. So Tsallis entropy is the generalized form of information entropy.

The constraints express mathematically the mean values information available. If $f_i(D)$ is a function describing droplet state (mass, for example) and its known mean value is $F_i$, the corresponding constraint written for all the size classes of the spray spectrum is:

$$\sum_{i=1}^{n} p_i f_i(D) = \langle f_i(D) \rangle = F_i$$
For spray applications of MEF, constraints may be derived from conservation laws. The problem of finding the most likely size distribution for the calculated mean diameter is expressed mathematically by a maximization problem. We need to find the maximum of the function described in eqn (1) subject to \( i+1 \) constraints. To the \( i \) constraints expressed as in eqn (2) is added the general probability constraint:

\[
\sum_{i=1}^{n} p_i = 1
\]  

The maximization problem is solved defining as many Lagrange multipliers as constraints (\( \lambda_1, \lambda_2, \ldots, \lambda_r \)). The solution is an exponential distribution when Shannon entropy is chosen and a power law distribution when Tsallis entropy (with \( q \neq 1 \)) is chosen:

\[
p^\varepsilon_i = \exp\left(-\lambda - \lambda_f (D_i) - \ldots - \lambda_f (D_i)\right) \quad \quad p^\varepsilon_i = \frac{q-1}{q} \left(-\lambda - \lambda_f (D_i) - \ldots - \lambda_f (D_i)\right)^{\frac{1}{1-q}}
\]

To evaluate the Lagrange multipliers eqns (2) are used. This results in a system of non-linear equations solved using an algorithm developed by Agmon and Alhassid [15].

**MAXIMUM ENTROPY APPLIED TO ULTRASONIC ATOMIZATION**

Ultrasonic nozzles produce low velocity droplets. Therefore the present work focuses only on droplets size distributions, although it is possible to predict both size and velocity distributions. Prior information to use for the MEF is the predicted value of mass mean diameter and atomization characteristics such as nozzle vibration frequency, liquid flow rate, liquid physical properties. When using Tsallis entropy the \( q \) exponent should also be determined. This exponent is an image of the input energy available for atomization. All we know till now is that wave instability mechanism leads to \( q=1 \) (Shannon entropy). Further investigation will provide values for other break-up mechanisms. Present study proposes \( q=1.3 \) for cavitation break-up.

For the ultrasonic atomization system, the conservation laws are written between two states of the liquid volume: the liquid film of mass \( M \) formed on the nozzle’s vibrating surface and a cloud of droplets of equal mass. The first natural constraint is mass conservation. If we suppose that no evaporation occurs, this constraint may be written as:

\[
M = \sum_{i=1}^{n} m_i
\]

If there are \( N \) droplets in the considered set then:

\[
\sum_{i=1}^{n} m_i = \rho \pi \frac{N}{6} \sum_{i=1}^{n} D_i^3 p_i
\]

If the liquid mass breaks up into \( N \) equal diameter (\( D_{30} \)) droplets, then:

\[
M = \rho \pi \frac{N}{6} D_{30}^3
\]

Mass conservation constraint for MEF results in:

\[
\sum_{i=1}^{n} d_i^3 p_i = 1 \quad \quad d_i = \frac{D_i}{D_{30}}
\]

For ultrasonic atomization the energy conservation is the best description of the break-up mechanism as it includes the transfer between ultrasonic wave energy and final droplets surface tension and kinetic energy.

Wave energy transforms mainly in surface tension energy and in a less important kinetic energy (droplets velocities in an ultrasonic spray are low – in the range of 1 m/s -).

The energy constraint includes wave energy (\( E_w \)) and liquid film surface energy before break-up (\( E_s \)), droplets surface tension (\( E_{DS} \)) and kinetic energy (\( E_{DK} \)). Only part of the available energy is used for the atomization thus an efficiency \( \eta_1 \) is applied to the energy conservation equation:

\[
\eta_1 \left(E_w + E_s \right) = E_{DS} + E_{DK}
\]

This energy constraint applies for the wave instability break-up mechanism. For high energy ultrasonic sprays, cavitation inside the feeding channel of the atomizer occurs and part of spray droplets are produced by bubble implosion due to the oscillating acoustic pressure. Pressure waves are emitted during bubble collapse. The energy constraint should contain this pressure wave energy as available input energy:

\[
\eta_1 E_{cavitation} = E_{DS} + E_{DK}
\]

Energy conservation constraint for MEF is:

\[
\sum_{i=1}^{n} d_i^3 p_i = k_E
\]

An expression for the \( k_E \) energy input factor has been found for the wave instability break-up mechanism and is
presented in [12]. The $k_E$ factor includes important information about the atomization device (frequency and amplitude of vibration) and about the sprayed liquid (surface tension, density).

There is not yet such an explicit analysis for the cavitation mechanism therefore $k_E$ may only be estimated from order of magnitudes of bubble collapse energy in ultrasonic field.

In order to find the probability distribution related to the predicted mass mean diameter, the MEF requires to maximize one of the forms of information entropy (eqns (1) subject to three constraints:

$$\sum_{i=1}^{n} p_i = 1$$

$$\sum_{i=1}^{n} p_i d_i = k_E$$

$$\sum_{i=1}^{n} p_i d_i^3 = 1$$

(12)

For exemplification purposes, hereafter we show the possible use of MEF based on combined Tsallis and Shannon information entropy to predict bimodal droplet size distributions. Such distributions are observed when increasing the input energy of ultrasonic atomizers. As one may see in figure 1, the experimental distribution observed for an ultrasonic high-energy nozzle spraying water at various flow rates, is clearly bimodal with second mode becoming more and more important as flow rate is increased.

![Figure 1](image-url)  

**FIGURE 1.** Experimental versus MEF predicted droplets volume distribution for a high-energy ultrasonic atomizer spraying water at increasing flow rates.

The first mode reflects the wave instability mechanism of droplet formation. It is well fitted by a Shannon exponential distribution law and is the classical mode observed in low-energy ultrasonic atomization. The second mode is well fitted by a Tsallis power distribution law of exponent $q=1.3$. This secondary distribution reflects droplet formation through cavitation effects.
Cavitation generates high energy pressure waves inside the atomizer feeding channel which causes violent droplet ejection. As flow rate is increased, velocity inside the channel increases therefore promoting cavitation. As a result the second break-up mode becomes more and more important.

Measured mass mean diameter for the entire distribution is the same for all four tested flow rates ($D_{\text{m}} = 44 \pm 0.5\% \, \mu m$). The mean diameter used in MEF for the derivation of the first mode probability distribution (based on Shannon entropy) was calculated from wave analysis as described in [16]. Unfortunately for the cavitation break-up mechanism no mean diameter theoretical prediction is available. We used a best fit procedure in order to determine the mean diameter for the second mode break-up distribution.

As Shannon law is a limiting case of Tsallis law (when $q=1$) we may consider that the $q$ exponent quantifies the energy involved in the break-up mechanism. Wave instability low-energy break-up lead to distributions of exponent $q=1$ whereas cavitation high-energy break-up will form $q=1.3$ power law distributions.

**CONCLUSION**

The maximum entropy formalism provides a logical method for choosing the droplet distribution in agreement with the knowledge we have about atomization phenomena. Till now application in spray predictions used the Shannon entropy function. The resulting probability distribution function is exponential as most of the empirical functions used in spray predictions (Rosin-Rammler, log-normal,...). A generalized form of information entropy has been proposed by Constantino Tsallis. If Tsallis entropy function is used in MEF the corresponding probability distribution function is a power law. Power laws have been found in high energy phenomena such as fragmentation where scaling from exponential to power law occurs when increasing the impact energy.

Most of the spray break-up mechanisms are based on wave instability. Droplets formed by detachment from wave crests tend to be distributed following an exponential law. But some atomizers showed bimodal distributions and transition from an exponential to a power law. This phenomena occurs in airblast atomizers were part of the sprayed liquid is subject to backflow and violent break-up. A bimodal distribution may also been observed in ultrasonic sprays when input power is increased. Part of the droplets are obtained through a surface wave instability mechanism while another part is the result of cavitation inside the feeding channel.

When high-energy break-up mechanism (such as cavitation or droplets burst in combustion) are involved in spray formation, Tsallis form of entropy should be used in order to predict the droplets distribution function.

The example of high-energy ultrasonic atomization is presented here. The experimental bimodal distribution attests for two mechanism of droplets formations, wave instability and cavitation. The wave instability mode is predicted using MEF with classical Shannon entropy while the cavitation mode is better described by the power law obtained using Tsallis entropy with $q=1.3$. For a complete theoretical prediction the mass mean diameter of each partial distribution should be determined theoretically. This is done for wave instability break-up but is difficult for cavitation.

The described approach should be useful in all bimodal distributions as they are likely to originate in different break-up mechanism. Furthermore the exponent of Tsallis power law might be used as a quantitative description of input energy available for atomization.

**NOMENCLATURE**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_i$</td>
<td>Non-dimensional diameter of the size class $i$</td>
</tr>
<tr>
<td>$D_i$</td>
<td>Arithmetical mean diameter of the size class $i$</td>
</tr>
<tr>
<td>$D_{30}$</td>
<td>Mass mean diameter</td>
</tr>
<tr>
<td>$E_W$</td>
<td>Mean wave energy</td>
</tr>
<tr>
<td>$E_S$</td>
<td>Liquid film surface energy</td>
</tr>
<tr>
<td>$E_{DS}$</td>
<td>Droplets surface energy</td>
</tr>
<tr>
<td>$E_{DK}$</td>
<td>Droplets kinetic energy</td>
</tr>
<tr>
<td>$E_{cavitation}$</td>
<td>Cavitation energy</td>
</tr>
<tr>
<td>$f_k$</td>
<td>Function describing droplet state</td>
</tr>
<tr>
<td>$F_k$</td>
<td>Mean of $f_k$ over all the droplets</td>
</tr>
<tr>
<td>$k$</td>
<td>Information entropy constant</td>
</tr>
<tr>
<td>$k_E$</td>
<td>Energy constant</td>
</tr>
<tr>
<td>$m_i$</td>
<td>Total mass of the droplets in the size class $i$</td>
</tr>
<tr>
<td>$M$</td>
<td>Liquid film mass</td>
</tr>
<tr>
<td>$n$</td>
<td>Number of droplets size classes</td>
</tr>
<tr>
<td>$N$</td>
<td>Number of droplets inside the control volume</td>
</tr>
<tr>
<td>$p_i$</td>
<td>Probability that a droplet diameter is into the size class $i$</td>
</tr>
<tr>
<td>$p_i^{S}$</td>
<td>$p_i$ obtained with Shannon entropy</td>
</tr>
<tr>
<td>$p_i^{T}$</td>
<td>$p_i$ obtained with Tsallis entropy</td>
</tr>
<tr>
<td>$q$</td>
<td>Tsallis law exponent</td>
</tr>
<tr>
<td>$r$</td>
<td>Number of constraints</td>
</tr>
<tr>
<td>$S$</td>
<td>Shannon's information entropy</td>
</tr>
<tr>
<td>$T$</td>
<td>Tsallis information entropy</td>
</tr>
<tr>
<td>$\lambda_0, ..., \lambda_r$</td>
<td>Lagrange multipliers</td>
</tr>
<tr>
<td>$\eta_1, \eta_2$</td>
<td>Energy transformation efficiency</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Density</td>
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</table>

**REFERENCES**