ULTRASONIC ATOMIZATION SPRAY ANALYSIS WITH A THREE-PARAMETER GENERALIZED GAMMA FUNCTION

Dr. Christophe Dumouchel*, Dr. Daniel Sindayihebura and Prof. Léon Bolle*****
Christophe.Dumouchel@coria.fr
*** UMR 6614 – CORIA – Université et INSA de Rouen, Avenue de l'Université BP 12**
76801 Saint Etienne du Rouvray – France
**** Polyspray, Place du Levant, 2, 1348 Louvain la Neuve – Belgique**
***** Université Catholique de Louvain, Unité Term, Place du Levant, 2, 1348 Louvain la Neuve – Belgique**

Abstract
The work reported in this paper deals with the analysis of drop-size distribution of sprays produced by ultrasonic atomizers. A series of measurements is conducted as a function of the working frequency of the atomizer as well as of the physical properties of the liquid to be atomized. These measurements, performed with a diffraction technique, report the expected behavior as far as the influences of the working frequency and of the liquid properties are concerned. The measured distributions are then analyzed through the application of the Maximum Entropy Formalism. A recent application of this formalism to atomization problems led to a mathematical function for the volume-based drop-size distribution. This three-parameter function is a reduction of the generalized four-parameter gamma distribution. The present work shows that this function is very well adapted to represent volume-based drop-size distribution of ultrasonic sprays. Furthermore, it is found that among the three parameters introduced by the function one of them is constant for all the situations investigated and the two others are linked to a non dimensional group that includes the working frequency, the liquid-air surface tension and the liquid density. These results are very important as they suggest a possible development of a physical model of atomization based on the M.E.F. and that would allow the prediction of the spray drop-size distribution. Such a model does not exist so far.

Introduction

The existence of a mathematical model to predict spray drop-size distributions would be appreciated. Unfortunately such a model does not exist so far mainly because liquid atomization is a complex multivariable problem that has not been fully understood yet. A helpful prior step to the development of a liquid atomization model might be the establishment of a universal mathematical expression for the spray drop-size distribution. According to Lefebvre [1], one of the desirable attributes of such a mathematical expression should be to furnish some insight into the basic mechanism involved in atomization.

For almost twenty-five years the application of the Maximum Entropy Formalism (M.E.F.) has been seen as an interesting and promising alternative to elaborate mathematical drop-size distributions. The first applications of this formalism to predict spray drop-size distribution were conducted by Sellens and Brzustowski [2] and by Li and Tankin [3] simultaneously. In both approaches, the writing of the set of constraints, required by the formalism, was based on physical conservation laws: for the first time the determination of mathematical drop-size distribution rested on physical considerations. A different application of this formalism led recently to the following mathematical expression for the volume-based drop-size distribution [4, 5]:

$$f_v(D) = q\frac{\left(\frac{4}{q}\right)^{4/q}}{\Gamma\left(\frac{4}{q}\right)}\frac{(D-D_0)^3}{\left(D^*_{q+3,3}\right)^4}exp\left[-\frac{4}{q}\left(\frac{D-D_0}{D^*_{q+3,3}}\right)^q\right] \tag{1}$$

where D represents the drop diameter, $D_{q+3,3}$* is the constraint diameter, q is the order of the constraint diameter, D_0 is the diameter of the smallest drop and Γ stands for the classical Gamma function (for more details refer to [5]). The drop-size distribution given by Eq. (1) is a function of three parameters, namely, D_0, $D_{q+3,3}$* and q. It is a reduction of the more general four-parameter Generalized Gamma distribution.

In the present paper, drop-size distributions of sprays produced by ultrasonic atomizers are going to be analyzed by the function f_v given by Eq. (1). The objective is to see whether the function f_v is adapted to describe drop-size distribution of sprays produced by ultrasonic atomizer, and to which extend the parameters introduced by the mathematical distribution are related to the physics of atomization.

Experimental investigation

The type of ultrasonic atomizer used in this work is schematized in Fig. 1. It is an acoustically resonant device composed of two piezoelectric rings (2 in Fig. 1) and a mechanical transformer (3 and 4). The free end of the mechanical transformer forms the atomizing surface. The liquid is supplied to the atomizing surface through the centered channel (5). The application of an electrical tension, characterized by a frequency and an amplitude, on the piezoelectric elements generates mechanical vibrations with the same frequency as the electrical input. When working at the resonant frequency, which is a function of the geometry of the atomizer, the amplifier concentrates the acoustical energy on the atomizing surface. Thus, as soon as the liquid is introduced on the atomizing surface, it spreads to form a thin liquid film and a stationary square-wave pattern develops on the interface of the liquid film. The wavelength of the square-wave pattern is a function of the working frequency and of the liquid-air surface tension. It can be estimated by the relation:

$$\lambda_s = \left(\frac{8\pi\sigma}{\rho_L f^2} \right)^{1/3} \tag{2}$$

When the intensity of the input signal is high enough, drops are emitted from the crests of the most rapidly growing unstable wave. In the present work, three atomizers were tested with seven different liquids (see Tables 1 and 2). This selection of atomizers and fluids allowed us to independently study the influence of the liquid viscosity, of the liquid-air surface tension and of the working frequency. For each atomizer fluid combination, the influence of the liquid flow rate was also examined in the range [0.4-l/h; 1.4-l/h] ([110-mm^3/s; 390-mm^3/s]).

The drop-size distributions were measured with a Malvern 2600D equipment. This particle sizer is a line-of-sight forward diffraction technique. The center of the laser beam was positioned at 5-mm from the atomizer. The diameter of the laser beam was 7-mm. The receiver of the Malvern was equipped with a 300-mm focal length lens allowing the measurement of drop diameter ranging from 5.8-µm to 564-µm. The main experimental results are summarized hereafter.

Influence of the liquid flow rate: An example of the influence of the mass flow rate on the drop size distribution is presented in Fig. 2 (Atomizer 2, Fluid 1). This figure shows that the mass flow rate has a reduced influence on the drop-size distribution up to 1.0-l/h. Above this limit the distribution slightly enlarges. This behavior is similar to experimental observations reported in the literature [6; 7]. Measurements presented in these studies show that the drop-size distribution of ultrasonic sprays is constant for a given flow rate interval. If the flow rate is outside this interval (under or above) the distribution enlarges as observed in Fig. 2 for the higher flow rates. When the flow rate ranges in this interval, the drop-size distribution of the spray is constant and is the narrowest for the considered atomizer-liquid combination. The efficiency of the atomizer is maximum is these conditions. The analysis presented in this work focuses on the distributions obtained in these working conditions only. For each atomizer-fluid combination tested, an average drop-size distribution was calculated on the basis of the distributions that were found independent of the flow rate. For the case of Atom.2 used with fluid 1, this average distribution is shown in Fig. 2. In this example, the distributions measured for the two greater flow rates were disregarded in the calculation of the average distribution.

Influence of the liquid viscosity: The influence of the liquid viscosity was investigated on the average drop-size distributions obtained for Fluids 1, 2, 3 and 4 used with Atom. 2. The results are presented in Fig. 3 where it appears clearly that the influence of the liquid viscosity on the drop-size distribution is negligible in the present range of working condition. This expected behavior comes from the fact that in ultrasonic atomization the characteristics of the internal liquid flow, which are mainly controlled by the liquid viscosity, do not influence the production of the liquid system to be atomized as in the case of pressure atomization. The behavior reported in Fig. 3 encourages us to reduce the distributions into an average distribution. This distribution calculated as a regular arithmetic mean is shown in Fig. 3.

Influence of the liquid-air surface tension: The influence of the liquid-air surface tension was investigated with Atom. 2 used with the fluids 2, 5, 6 and 7. Figure 4 compares the distributions obtained for each fluid. Each distribution is averaged on the flow rate. The distributions reported in this figure show that a decrease of the surface tension favors the production of a better-atomized sprays. This behavior is due to the fact that a decrease of the surface tension induces a reduction of the wavelength of the stationary wave that develops on the liquid film interface and that structures the disintegration process (see Eq. (2)).

Influence of the atomizer working frequency: Finally, the three atomizers presented in Table 1 were used with Fluid 1 to investigate the influence of the working frequency on the drop-size distribution. The results are presented in Fig. 5 where it can be seen that an increase of the working frequency induces the production of smaller drops. As for the influence of the liquid air surface tension, this behavior is due to the fact that the

wavelength of the stationary wave decreases when the atomizer frequency increases leading then to the production of smaller drops (see Eq. (2)).

Application of the Maximum Entropy Formalism

The objective of this paper is to analyze the drop-size distribution of ultrasonic sprays through the application of the Maximum Entropy Formalism, i.e., through the application of the mathematical function given by Eq. (1). First, the analysis consists in determining the propensity of the mathematical distribution to fit the measurements. For each working condition this requires the determination of the three parameters q, $D_{q+3,3}$* and D_0. Second, the influence of the liquid surface tension and of the atomizer working frequency on these parameters is investigated.

From a mathematical point of view, the determination of the three parameters can be achieved by using any combination of three independent characteristics of the distribution such as mean drop diameters for instance. However, the resulting mathematical function is highly dependent on the characteristics used and the desired solution is usually obtained for a unique set of information. This problem is overcome here by determining the parameters on the basis of a numerical procedure, which uses the Kullback-Leibler number I defined by:

$$I = \sum_i p_i \, ln\left(\frac{p_i}{p_{ei}}\right) \tag{3}$$

where p_i and p_{ei} are two probability distributions. The number I constitutes a measure of the nearness of the two probability distributions. In the present application, p_{ei} and p_i represent the experimental probability distribution and the mathematical probability distribution, respectively. They are calculated by the following relations:

$$p_{ei} = m_v(D_i)\Delta D_i \quad \text{and} \quad p_i = \int_{Class\ i} f_v(D)dD \tag{4}$$

In Eq. (3), $f_v(D)$ is given by Eq. (1) and $m_v(D_i)$ corresponds to the measured probability density function. The value of I (Eq. (3)) is a function of the parameters q, $D_{q+3,3}$* and D_0 introduced by $f_v(D)$ and it is minimum when the probability distribution p_i is as near as possible to the experimental probability distribution p_{ei}. Therefore, for each situation, a numerical procedure seeks the parameters q, $D_{q+3,3}$* and D_0 that minimize the number I. This calculation is conducted with the Matlab software.

The numerical procedure is first tested for the average drop-size distribution obtained for Atom. 2 (Fig. 3). The comparison between the experimental average-distribution and the mathematical distribution is presented in Fig. 6 where the values of the three parameters are also indicated. The result presented in this figure shows a very good agreement between the two distributions. This shows that the three-parameter generalized gamma function deduced from the application of the M.E.F. seems adapted to represent volume-based drop-size distribution

The determination of the mathematical distribution was performed for the four distributions showing the influence of the liquid-air surface tension (Fig. 4). For each situation, the agreement between the calculated and the measured distributions was rather good. As far as the evolution of the parameters is concerned, two points were noted. First, it was found that a decrease of the liquid-air surface tension induces a reduction of both diameters D_0 and $D_{q+3,3}$*. Second, the parameter q was found to be almost independent of the surface tension coefficient. The average q value deduced from these four applications was equal to 1.35, which is close to 1.4 reported in Fig. 6. It was decided to start over again the parameter determination for the same cases but with a parameter q set to 1.4. The results of this second application are presented in Fig. 4. For each situation, the agreement is rather good. Thus, a constant parameter q equal to 1.4 seems to be well adapted to represent the volume-based drop-size distribution of sprays produced by Atom. 2.

This very value of the parameter q is used to analyze the drop-size distributions measured as a function of the atomizer working frequency. The resulting distributions are presented in Fig. 5 together with the experimental results. For the three situations shown in this figure, the agreement between the calculated and the measured distributions is very good.

All these results confirm that the three-parameter generalized Gamma distribution deduced from the application of the M.E.F. is very well adapted to represent volume-based drop-size distribution of ultrasonic sprays. Above this, it appears that the parameter q can be considered constant and equal to 1.4 whatever the atomizer and the fluid provided that it is working at the optimum atomization efficiency. This suggests that the distribution calculated with q = 1.4 would characterize the finest spray obtainable with the atomizer and the fluid used.

The two other parameters, D_0 and $D_{q+3,3}$* are equivalent to drop diameters and are both functions of the liquid-air surface tension and of the atomizer working frequency. They both decrease when the frequency

increases or when the surface tension decreases. Many experimental investigations found in the literature reported that any mean drop diameter of ultrasonic sprays is directly proportional to the wavelength of the wave pattern that structures the disintegration process. Figure 7 shows that this behavior may be considered valid for the parameter D_0 and $D_{q+3,3*}$. This figure shows the relationship between the wavelength λ_s and the two parameters. For each situation, the wavelength was estimated thanks to Eq. (2). Despite the fact that the points in Fig. 7 show a slight scatter, it seems reasonable to assume that the parameters D_0 and $D_{q+3,3*}$ are both proportional to the wavelength λ_s. Considering Eq. (2), this result encourages us to consider the following number $\overline{D}$ defined by:

$$\overline{D} = \frac{\rho_L f^2 D^3}{\sigma} \tag{5}$$

This number has no dimension. It is equivalent to a Weber number where the velocity has been replaced by the product *f.D*, where *D* is a drop diameter. For each characteristic diameter of the spray drop-size distribution, the number $\overline{D}$ takes a constant value. For the two parameters D_0 and $D_{q+3,3}$*, this constant value is equal to 0.15 and 0.27 respectively. Thus, the drop-size distributions of sprays produced by the ultrasonic atomizers investigated in this study are well represented by the mathematical distribution given by Eq. (1) where the three parameters are given by:

$$\begin{cases} q = 1.4 \\ \overline{D_0} = 0.15 \\ \overline{D_{q+3,3}*} = 0.27 \end{cases} \tag{10}$$

Conclusion

The present paper reports an analysis of the behavior of ultrasonic atomizers. It concentrates on the influence of the fluid and of the working frequency of the atomizer on the spray drop size distribution. Measurements of the drop-size distribution were performed with a line-of-sight diffraction technique. The experimental results showed that the flow rate and the liquid viscosity have a negligible influence on the drop-size distributions that appear to be mainly functions of the liquid-air surface tension and of the atomizer frequency. The size of the drops globally decreases with an increase of the working frequency or a decrease of the liquid-air surface tension.

The measured drop-size distributions were analyzed via the application of the Maximum Entropy Formalism. Developed in a previous investigation, this formalism led to the establishment of a mathematical function to represent volume-based drop-size distribution. This function is a three-parameter generalized Gamma function. The purpose of this analysis is to see whether this function is adapted to represent the drop-size distribution of ultrasonic sprays, and to which extend the parameters introduced by the mathematical distribution are physically relevant.

All the applications reported in this paper show that the three-parameter generalized Gamma function is very well adapted to represent volume-based drop size distributions of sprays produced by ultrasonic atomizers. Furthermore it has been found that the three parameters introduced by the mathematical function show specific behavior. For all the situations investigated in this paper, the parameter *q* was found constant and equal to 1.4. This result is interesting and suggests that this parameter is related to the atomization process only, i.e., to the way the drops are produced. Similar behavior was observed in previous investigation ([4] for instance). The two other parameters, which are equivalent to drop diameters, have been found to depend on the atomizer frequency, the liquid density and the liquid-air surface tension. Their values were connected to a group without dimension that is equivalent to a Weber number. This last result is important. It indicates clearly that the parameters introduced by the mathematical function are not only fitting parameters but are representative of the physics involved in the atomization process investigated here. It must be added here that these results are relevant only in the cases where the influence of the flow rate and on the liquid viscosity on the drop-size distribution is negligible. In other words, they allow to predict the drop-size distribution of the finest spray an ultrasonic atomizer can provide with a given liquid.

Nomenclature

D	Drop diameter
D_0	smaller drop diameter (parameter of the mathematical distribution)
$D_{q+3,3}$*	constraint mean diameter (parameter of the mathematical distribution)
f	working frequency of the ultrasonic atomizer

f_v	mathematical volume-based drop-size distribution (Eq. (1))
I	Kullback-Leibler number
p_i	theoretical probability distribution
p_{ei}	measured probability distribution
q	parameter of the mathematical function f_v
Γ	Gamma function
σ	liquid-air surface tension
ρ_L	liquid density
λ_s	wavelength of the most unstable wave

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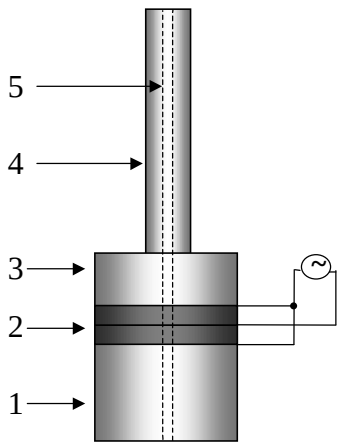

Figure 1. Schematic view of a low-frequency ultrasonic atomizer

Atomizer	Working frequency f (kHz)
1	41.1
2	49.9
3	135

Table 1. Frequency of the ultrasonic atomizers

Fluids (% in mass)	μ_L (10^{-3}-kg/ms)	σ (mN/m)	ρ_L (kg/m^3)
1. water	1.00	72.75	1000
2. water + glycerol (15%)	1.49	72.67	1033
3. water + glycerol (21%)	1.80	72.36	1048
4. water + glycerol (36%)	3.09	71.54	1087
5. water + methanol (16%)	1.50	53.97	972
6. water + methanol (47%)	1.80	36.08	921
7. water + methanol (64%)	1.50	31.84	885

Table 2. Physical properties of the fluids

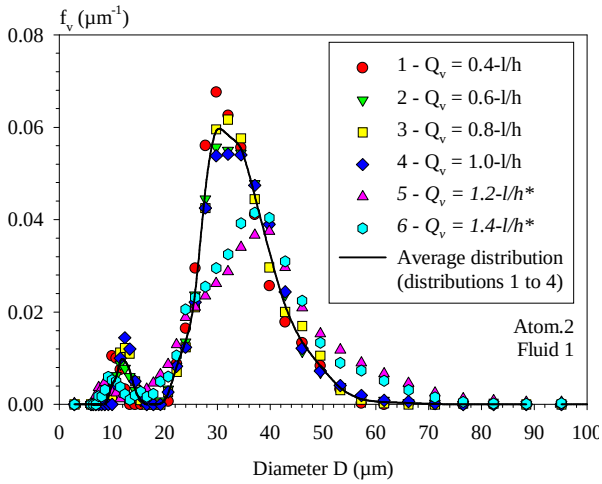

Figure 2. Influence of the liquid flow rate on the drop-size distribution (Atom. 2, Fluid 1) *not taken in the average

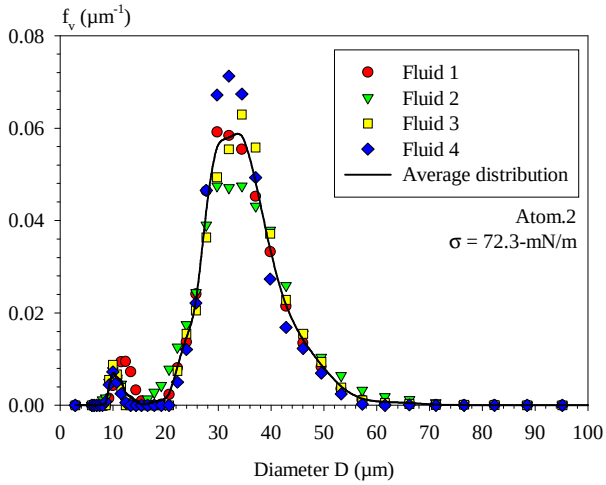

Figure 3. Influence of the liquid viscosity on the drop-size distribution (Atom. 2)

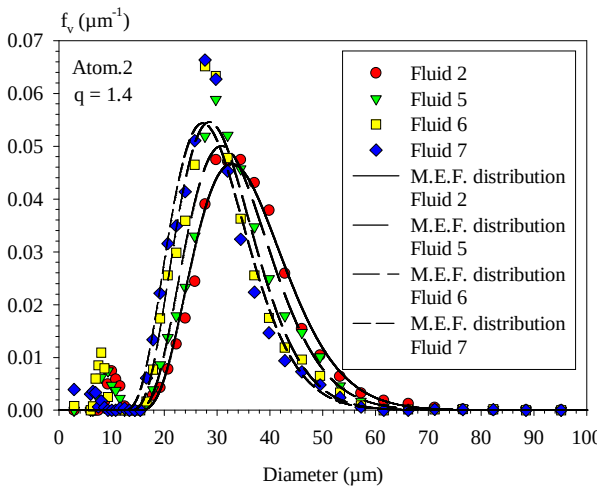

Figure 4. Influence of the liquid viscosity on the drop-size distribution (Atom. 2)

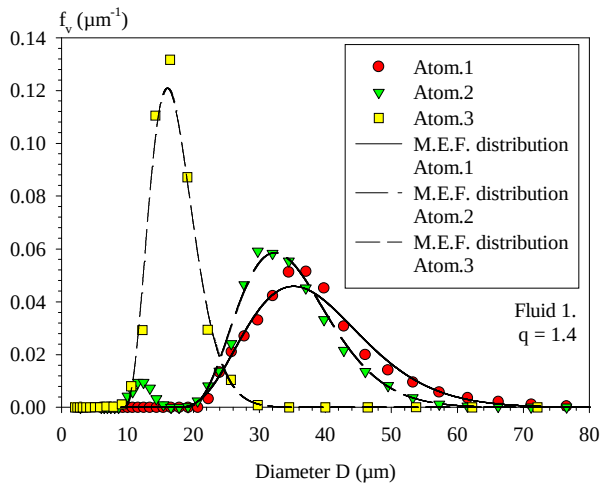

Figure 5. Influence of the atomizer working-frequency on the drop-size distribution (Fluid 1)

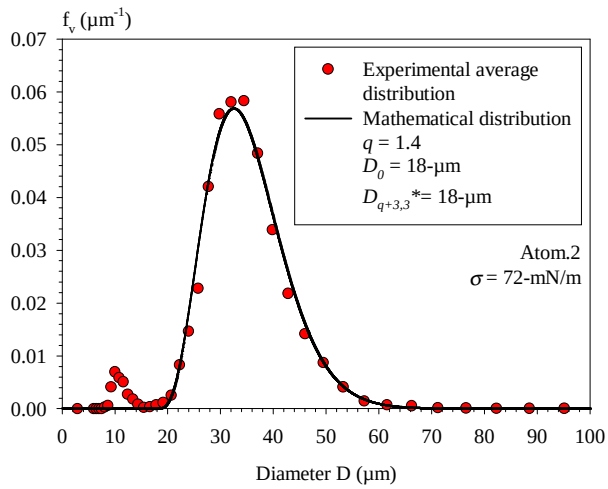

Figure 6. Comparison between the M.E.F. and the experimental volume-based drop size distribution

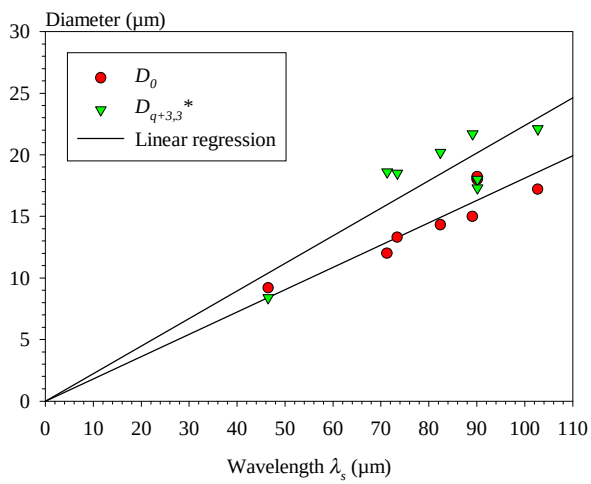

Figure 7. Evolution of the mathematical distribution parameters with the stationary wave wavelength