THEORETICAL PREDICTION OF ULTRASONIC SPRAY CHARACTERISTICS USING THE MAXIMUM ENTROPY FORMALISM

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ABSTRACT
The present study aims at providing a theoretical procedure for the determination of droplets mean diameter and probability distribution. It is developed for the particular case of ultrasonic atomisation. First the mean droplet diameter is predicted based on the stability analysis of the liquid film. Then, the maximum entropy formalism (MEF) is used to find the most likely droplets diameter probability distribution with respect to the predicted mean diameter and with the type of atomisation. Finally, theoretical results are compared with experimental measurements. MEF provides a logical method for choosing a distribution in agreement with the knowledge we have about atomisation phenomena. As it is not relying on fitting to experimental data it may be used to find diameter probability distributions when and adequate theoretical approach provides a mean droplet diameter.

NOTATION

\( A \) Liquid film wave amplitude
\( A_0 \) Atomising surface wave amplitude
\( D_i \) Arithmetic mean diameter of the size class \( i \)
\( D_{\text{m}} \) Mass mean diameter
\( E_W \) Mean wave energy
\( E_S \) Liquid film surface energy
\( E_{DS} \) Droplets surface energy
\( E_{DK} \) Droplets kinetic energy
\( f \) Ultrasonic frequency
\( f_k \) Function describing droplet state
\( f_x \) Mean of \( f_k \) over all the droplets
\( h \) Liquid film height
\( k \) Energy constant
\( m_i \) Total mass of the droplets in the size class \( i \)
\( M \) Liquid film mass
\( n \) Number of droplets size classes
\( N \) Number of droplets inside the control volume
\( p_i \) Probability that a droplet diameter is into the size class \( i \)
\( r \) Number of constraints
\( S \) Shannon's information entropy
\( V \) Mean droplets velocity
\( \Phi \) Potential function
\( \lambda \) Liquid film oscillation wavelength
\( \lambda_1, \ldots, \lambda_r \) Lagrange multipliers
\( \sigma \) Surface tension
\( \omega \) Atomising surface wave angular frequency

INTRODUCTION
A spray is basically a set of non-uniform droplets which requires expensive measurement procedures in order to determine the size distribution. To perform further calculations, several analytical probability density functions (Rosin-Rammler, log-normal, log-hyperbolic, etc.) are commonly used to fit the experimental data. The choice of the probability density function is only based on the best fit procedure: it cannot be founded upon neither physical nor logical basis. This common approach provides no physical link between the atomisation parameters (type of atomiser, liquid properties) and the resulting spray characteristics. The extrapolation of available data to other working conditions is extremely uncertain. This is the first reason why spray research should use more extensively the maximum entropy formalism (MEF) to provide a probability distribution function closely related to the physics of atomisation.

A global characteristic of the spray is the mean diameter (several definitions of the mean diameter may be chosen depending on research purposes). When the size distribution is known, the mean diameter can easily be calculated. But when only a mean diameter is known, either by theoretical calculation or by data extrapolation, an infinite number of size distributions may fit to it. This is another reason why spray research needs a logical tool (as MEF) to operate the best distribution choice.

The work presented here involves a theoretical description of the liquid film break-up phenomenon that leads to the mean diameter prediction and the maximum entropy formalism as a logical method for the choice of the droplet size.
distribution related to this diameter. The knowledge of the operating conditions (ultrasonic frequency, flow rate) and of the liquid properties (surface tension, density) is sufficient.

Drops formation in atomisation process is mainly caused by the development of rapidly growing waves on a liquid surface. The instability of those waves is a result of internal causes (nozzle vibrations, liquid swirling, liquid pressure drop,...) or/and external causes (interactions with the surrounding medium). For the ultrasonic atomisation the main source of wave growth is the vibration of the piezoelectric nozzle. The wavelength of the most rapidly growing surface wave is predicted by the analysis of liquid film instability achieved by Sindayihebura (1). The mean droplet diameter may then be determined assuming that droplets form from the crests of the unstable surface waves (Sindayihebura & al. (2)).

The next step is to find the most likely probability distribution with respect to the calculated mean diameter and with the type of atomisation. This can be achieved by using Jayne’s maximum entropy formalism (3), a widely used method which allows to choose among all the possible probability distributions the most suitable one to the available knowledge about the phenomena. The MEF is applied to sprays since 1986 in order to predict droplets size and eventually velocity probability distributions. First works in this field done by Sellens and Bruztowski (4) and Li and Tankin (5, 6) used to find the probability distribution based on the measured mean diameter. As knowledge about the atomisation mechanism increases, theoretical mean diameter calculation becomes available for some specific atomisers. The challenge now is to combine mean diameter prediction and MEF in a complete theoretical description of spray characteristics.

ULTRASONIC ATOMISATION CHARACTERISTICS

A thin liquid film formed on a high-frequency vibrating surface (30-70 kHz for the devices designed at UCL) will break-up in a fine uniform spray. The ultrasonic vibration induces surface waves in the liquid film. As frequency is tuned, very regular square cells was observed by Sindayihebura (1) on the free surface just before reaching the resonance frequency (Fig 1). When resonance is reached, the amplitude grows till droplets break-up (Fig 2). The very regular square cells generate uniform size droplets.

Photographs of free liquid film surface just before atomisation showed that the wavelength decrease when ultrasonic frequency increases. Measurement of droplets size by Sindayihebura & al. (7) proved that mean droplet diameter also decrease with increased frequency. Wavelength and mean droplet diameter show similar evolutions when modifying liquid properties (2) both increase when increasing surface tension and tend to be insensitive to changes in viscosity. We may conclude that, in ultrasonic atomisation there is a strong correlation between the wavelength of free surface waves observed just before atomisation and the mean droplet diameter of the resulting spray.

THE DROPLET MEAN DIAMETER PREDICTION

Ultrasonic vibration of the nozzle induces surface waves in the liquid film formed onto the atomising surface. A linear stability analysis using linearised Navier-Stokes equations may be performed (1). This results in a prediction formula for the wavelength of the most rapidly growing unstable waves. The crests of these waves break up, then generating droplets. The droplet volume will be a fraction of the crest volume generated by the intersection of two unstable standing waves exhibiting a regular square pattern (as in Fig 1). Equating the mean droplet volume with a fraction of the crest volume leads to the following relation:

\[ D_{\text{m}}^3 \div A \lambda^2 \]  \[ \text{[1]} \]

Wavelength is calculated for the specific atomisation conditions (nozzle frequency, liquid properties). Assuming that the wave amplitude increases together with the wavelength, the mean volume diameter may be predicted as a
fraction of the calculated wavelength.

The proportionality coefficient is nearly invariant with working frequency (7) and liquid properties (2):$$D_{30} = 0.36\lambda$$

The mean diameter can therefore be predicted with respect to atomisation parameters, via the wavelength equation issued from stability analysis of the liquid film free surface. A more accurate prediction involves the knowledge of exact amplitude value when break-up occurs. Unfortunately the linear theory is not suited for unstable amplitude evaluation.

Measured size distributions are available for ultrasonic sprays of mixtures of water-methanol and water-glycerine at different flow rates. For further comparison with MEF distributions mean volume diameter is calculated in Table 1 for 5 different liquids.

<table>
<thead>
<tr>
<th>Mixture - mass fraction -</th>
<th>Viscosity $(10^2 \text{ kg/ms})$</th>
<th>Surface tension $(10^{-3} \text{ N/m})$</th>
<th>Density $(\text{kg/m}^3)$</th>
<th>Wavelength $(\mu\text{m})$</th>
<th>$D_{30}$ $(\mu\text{m})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>1.00</td>
<td>72.75</td>
<td>1000</td>
<td>91.32</td>
<td>33.4</td>
</tr>
<tr>
<td>16% Methanol</td>
<td>1.50</td>
<td>53.97</td>
<td>972</td>
<td>83.57</td>
<td>30.6</td>
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<tr>
<td>30% Methanol</td>
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<td>43.02</td>
<td>951</td>
<td>78.13</td>
<td>28.6</td>
</tr>
<tr>
<td>47% Methanol</td>
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<td>36.08</td>
<td>921</td>
<td>74.51</td>
<td>27.3</td>
</tr>
<tr>
<td>64% Methanol</td>
<td>1.50</td>
<td>31.84</td>
<td>885</td>
<td>72.45</td>
<td>26.5</td>
</tr>
<tr>
<td>21%Glycerol</td>
<td>1.80</td>
<td>72.36</td>
<td>1048</td>
<td>92.04</td>
<td>33.7</td>
</tr>
</tbody>
</table>

Table 1. Predicted mass mean diameter ($f = 50 \text{ kHz}$).

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. Atomisation 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.

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 atomisation. Unfortunately an infinity of size distributions may fit to the unique calculated mean diameter. The choice of the "right" distribution should rely on the atomisation characteristics (break-up mechanism, fluid properties…). This is achieved by using the maximum entropy formalism (MEF), a widely used method which allows to choose among all the possible probability distributions the most suitable one with respect to the available knowledge about the phenomena.

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 maximising Shannon’s information entropy subject to constraints describing the available knowledge:

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

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

$$\sum_{i=1}^{n} p_i f_k(D_i) = \langle f_k(D) \rangle = F_k$$

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 maximisation problem. We need to find the maximum of the function described in eqn [3] subject to $i+1$ constraints. To the $i$ constraints expressed as in eqn [4] is added the general probability constraint:

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

The maximisation problem is solved defining as many Lagrange multipliers as constraints ($\lambda_0, \lambda_1, ..., \lambda_n$). The solution is an exponential distribution:
\[ p_i = \exp \left( -\lambda_0 - \lambda_1 f_i(D_i) - \lambda_2 f_2(D_i) - \lambda_3 f_3(D_i) \right) \]  

To evaluate the Lagrange multipliers eqn [5] is used. This results in a system of non-linear equations. Agmon and Alhassid (8) showed that solving this system is equivalent to minimising a potential function \( \Theta \) such as:

\[ f_i = \frac{\partial \Theta}{\partial \lambda_i} \]

The form of the potential function is:

\[ \Theta(\lambda_1, \lambda_2, \cdots, \lambda_n) = \log \left( \sum_{i=1}^{n} \exp \left( -\lambda_0 - \lambda_1 g_1(D_i) - \lambda_2 g_2(D_i) - \cdots - \lambda_n g_n(D_i) \right) \right) \]

where \( g_k = f_k - F_k \)  

The minimum of the potential function with respect to all \( \lambda_k \) variables is also a solution of the Lagrange multipliers system.

MAXIMUM ENTROPY THEORY APPLIED TO ULTRASONIC ATOMISATION

In previous publications presenting spray applications of the MEF three different approaches may be noted. Li and Tankin work (5,6) and Sellens first publications (4) are dedicated to the joint size and velocity distribution of droplets. Only size distributions where then determined by Cousin & al.(9) and Ahmadi & Sellens(10). Finally Van der Geld and Vermeer (11) summed two size distributions, one for main droplets and another one for the satellite droplets which appear in pressure driven sprays.

Ultrasonic nozzles produce low velocity droplets and, due to their little sizes, secondary break-up does not occur. Therefore the present work focuses only on droplets size distributions. Prior information to use inside the MEF is the predicted value of mass mean diameter and atomisation characteristics such as nozzle vibration frequency, liquid flow rate, liquid physical properties. In other respects, experimental data (2) pointed out that ultrasonic atomisation phenomenon is driven by surface tension and that viscous dissipation has no effect on the resulting spray.

For the ultrasonic atomisation 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:

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

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

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

In order to find the probability distribution related to the predicted mass mean diameter and obeying only the mass conservation law, the MEF requires to maximise the information entropy (eqn [3]) subject to two constraints:

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

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

\[ d_i = \frac{D_i}{D_{30}} \]

According to Agmon and Alhassid () the associated Lagrange multipliers \( (\lambda_k) \) system has the same solution as the potential function \( (\Phi) \) minimum . For the two constraints the potential function is:

\[ \Theta = \ln \left( \sum_{i=1}^{n} \exp \left( -\lambda_i (d_i^3 - 1) \right) \right) \]  

In Fig 3 the MEF predicted probability distribution is compared with a measured distribution of similar mass mean diameter, generated by an ultrasonic atomiser. The MEF distribution is transformed into a volume distribution in order to present the results in a familiar form for spray literature.

We can notice that the MEF distribution based only on mass conservation is broader than the experimental one and thus fails to
predict the major characteristic of the ultrasonic spray: very regular droplets (narrow distribution). In fact no information was provided to the MEF on atomisation physics specifics. So, based on mass conservation only, the MEF cannot predict one specific distribution but a ‘general’ distribution we might describe as a mean of all size distributions produced by all possible atomisation mechanisms.

Therefore, at least one other constraint has to be added to the system. For ultrasonic atomisation 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.

The energy conservation equation is divided into two constraints by Sellens (4), one for surface energy and another one for kinetic energy. In ultrasonic atomisation surface energy is clearly not conservative. The surface tension energy of thousands of droplets is much greater than the liquid film surface 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 and liquid film surface energy before break-up, droplets surface tension and kinetic energy. As for mass conservation we choose to write a full conservative equation (without dissipation):

\[ E_w + E_s = E_{os} + E_{ke} \]  \[13\]

Wave energy is difficult to evaluate. If we assume that the liquid film oscillation has the same parameters (frequency, amplitude) than the ultrasonic nozzle vibration, and that the wave form is a sinusoid, we may write the mean wave energy:

\[ E_w = \frac{1}{4} M \omega^2 A_0^2 \]  \[14\]

But this form is valid for stable waves of finite amplitude \( A_0 \) only. In fact the break-up occurs when amplitude grows so the energy just before atomisation is much greater.

The surface tension energy of the film and of the droplets is written as a function of liquid mass:

\[ E_s = \sigma \frac{M}{\rho h} \quad E_{os} = \sum_{i=1}^{n} \sigma S_i = \sigma N \pi \sum_{i=1}^{n} p_i D_i^2 \]  \[15\]

Kinetic energy of droplets is calculated based on the mean droplets velocity \( V \):

\[ E_c = \frac{V^2}{2} \sum_{i=1}^{n} m_i = \frac{1}{2} M V^2 \]  \[16\]

The energy constraint expression is:

\[ \sum_{i=1}^{n} p_i d_i^2 = k \]  \[17\]

The constant includes important information about the atomisation device (frequency of vibration \( f \)) and about the sprayed liquid (surface tension, density). The wave amplitude \( A_0 \) is taken equal to the known amplitude of the nozzle’s vibration but it is certainly underestimated as instability involves amplitude growth.

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Energy constant (calculated)</th>
<th>Energy constant (estimated)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>0.047</td>
<td>0.85</td>
</tr>
<tr>
<td>16% Methanol</td>
<td>0.041</td>
<td>0.89</td>
</tr>
<tr>
<td>30% Methanol</td>
<td>0.0409</td>
<td>0.78</td>
</tr>
<tr>
<td>47% Methanol</td>
<td>0.0363</td>
<td>0.88</td>
</tr>
<tr>
<td>64% Methanol</td>
<td>0.0329</td>
<td>0.855</td>
</tr>
<tr>
<td>21% Glycerol</td>
<td>0.0505</td>
<td>0.925</td>
</tr>
</tbody>
</table>

Table 2. The energy constant for different liquids sprayed with the same nozzle \( (f=50kHz) \)

The MEF system of constraints becomes:

\[ \sum_{i=1}^{n} p_i = 1 \]
\[ \sum_{i=1}^{n} p_i d_i^2 = k \]  \[18\]
\[ \sum_{i=1}^{n} p_i d_i^3 = 1 \]

The potential function to minimise is:

\[ \Theta = \ln \left( \sum_{i=1}^{n} \exp \left( -\lambda_i (d_{i}^2 - k) - \lambda_i (d_{i}^3 - 1) \right) \right) \]  \[19\]

When the energy constraint is calculated as in eqn [17], the numerical algorithm does not achieve convergence. An analysis of the energy constant influence on the shape of the MEF distribution leads to the conclusion that the acceptable \( k \) values are twenty times greater than the calculated ones. The values of \( k \) calculated as in eqn [17] and
evaluated for predicted distribution best fit are listed in Table 2. Fig 4 shows the MEF predictions when the energy constant is varied.

![Fig 4](image)

**Fig 4.** Influence of the energy constant on the MEF predicted distribution.

When the energy constant is well evaluated, the prediction of the MEF fits well with the measured distributions and tends to be even narrower. To predict larger drops present in the measured distributions, it might be useful to add an information about the coalescence mechanism as it has been already done for secondary break-up (11).

**CONCLUSION**

The maximum entropy formalism provides a logical method for choosing a distribution in agreement with the knowledge we have about atomisation phenomena. As it is not relying on fitting to experimental data it may be used to find the probability distribution when an adequate theoretical approach provides a mean droplet diameter. Combining instability analysis for mean droplet diameter prediction with maximum entropy formalism to find the corresponding probability distribution leads to a complete spray characterisation provided that the physics of the atomisation phenomena is well described. The ultrasonic atomisation benefits from a well understood mechanism and thus the results obtained by MEF prediction are very good when both mass and energy conservation are used as constraints. We can notice that, as the mass constraint results in a mean diameter constraint, the predicted distribution is symmetrical around this value. But mean diameter information alone is not sufficient to choose the best probability distribution (besides mass conservation does not say anything about physics of atomisation: it should be kept in mind that many other different phenomena may conserve mass). The energy conservation constraint depends on a still unknown parameter – wave amplitude when break-up occurs. Wave amplitude is also involved in the mean diameter prediction. If further research would fill in this gap, the experimental adjustment which is still used in the present work will be avoided.

**REFERENCES**