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# Measurement techniques concerning droplet size distribution of electrosprayed water

***Abstract.*** *The carefully-sized electrospraying of water is an important research problem because of wide use of sprays of water and aqueous solutions in many fields of industry and agriculture. The size of water droplets (or aqueous solutions) and its distribution is essential for various applications of aerosols. The measurement of size of electrosprayed water using laser-based measurement techniques is a non-intrusive and technologically advanced solution.*

***Streszczenie.*** *Wielkość mikrokropel wody oraz rozkład ich średnic mają istotne znaczenie w zastosowaniach aerosoli wytwarzanych metodą elektrostatycznego rozpylania. Pomiar średnic mikrokropel wody rozpylonej elektrostatycznie metodami opartymi na zastosowaniu lasera stanowi rozwiązanie bezkontaktowe i technicznie zaawansowane. (**Metody pomiaru rozkładu średnic kropel wody rozpylanej elektrostatycznie**).*

**Keywords:** electrospraying of water, droplet size distribution, droplet size measurement techniques.
**Słowa kluczowe:** elektrostatyczne rozpylanie wody, rozkład średnic kropel, metody pomiaru średnic i ładunku kropel.

## Introduction

The usefulness of many pulverized or dispersed substances in diverse fields and processes in both industrial end domestic activities (e.g. in paints, fuels, washing, or pharmaceuticals) is long known motivating factor for research and development of new techniques of pulverization, and measuring relevant features of the dispersed phase [1]. For many processes the most important characteristics is the particle size distribution (or, for sprays, the droplet size distribution, in short: DSD), since precise particle sizing has direct impact on the efficacy and quality of the product and helps reduce losses in both materials and energy. For example, ink jet printers or copier toners require carefully tailored DSD; similarly, saline aqueous solutions for drug inhalation can be targeted to specific part of the respiratory tract. Droplets under 3 μm in diameter are ejected during exhalation, whereas larger than 10 μm are trapped in the respiratory tract [2], so precise droplet sizing for a drug targeted to alveoli means large reduction of dosage.

For liquid spraying, the main efforts were focused on liquid fuels: a large number of spraying techniques and measurement methods have been developed [3], which in turn were adapted or incorporated in dispersion techniques for other liquids. The atomization of water has been of special interest because of the outstanding position of this liquid in human life and activities; mechanical and pneumatical atomizers have been invented.

The static electricity was applied to water atomizing (as well to other liquids) first in 1745 by Nollet, but the scientific origins of modern electrospraying of liquids were founded by theoretical works of Lord Rayleigh (1879) [4] and experimental works of Zeleny [5]. Adaptations of non-electric method in form of electrified rotating disc atomizer or HPMS (High Pressure Monodisperse Spraying) [6] supported by ring electrode were in common use. The scientific breakthrough happened in 1964, when Taylor formulated theory of a specific mode of electrospraying (ES), named cone-jet (or: Taylor jet) mode. In this mode, very fine aerosol of nearly monodispersed droplets is produced, moreover, the droplets are highly charged (in comparison with other modes of ES) with small energy consumed. As water is environmentally friendly medium, strong efforts are made for industrial use of water cone-jet electrospraying mode; unfortunately, this is difficult because of high surface tension of water, and the output is too low [7-11]. Because of this, the widespread scientific application of cone-jet mode is the Electrospray Ionization (ESI) [12].

Fast, non-intrusive and reliable measurements of DSD are important, among other fields, in meteorology, explosively generated water-sprays [13] or pharmaceuticals manufacturing; in the field of electrospraying of water (ESW), e.g. the EWICON project [6] of wind generator using charged water droplets is strongly dependent on accurate measurement of DSD.

## The distribution functions of ES water droplets

The DSD is a mathematical formula intended to characterise an experimental distribution without its plotting. The formula should be short and contain few parameters. The most common DSD functions for sprays are:

1 - the normal distribution (Eq.1) of droplet size is assumed when the range of droplets is narrow.

$$p(D)=\frac{1}{\sigma\sqrt{2\pi}}e^{\frac{-\left(D-\overline{D}\right)^2}{2\sigma^2}}, \tag{1}$$

where: $D$ – the current droplet diameter, $\overline{D}$ – the mean value of the droplet diameter, and $\sigma$ – the standard deviation.

2 - the log-normal (logarithmic normal) distribution, described by the Eq.2, is also used

$$p(D)=\frac{1}{\ln\sigma_{LN}\sqrt{2\pi}}e^{-\frac{\ln\left(D/\overline{D}\right)^2}{2\sigma_{LN}^2}}, \tag{2}$$

where $\sigma_{LN}$ is the standard deviation.

The log-normal distribution is less popular than the normal Gaussian, but there are strong arguments about the range of applications of log-normal instead of normal distribution [14]. It was introduced by Kolmogoroff for continuous grinding of solid particles.

3 - the Rosin-Rammler (Eq.3) distribution, introduced in 1933 [2], is used for fine sprays generated from polydispersed (by electrostatic, or other method) jet of a liquid:

$$\Phi(D)=1-e^{-\left(\frac{D}{X}\right)^q}, \tag{3}$$

where: $\Phi(D)$ – the cumulative distribution function of droplets diameter relative to liquid volume, $X$ –the value of $D$ for which is $\Phi(D)$=0.632, $q$ – the coefficient of spraying monodispersion (typical values are 2 to 4). That means, that 63.2% of the volume of atomized liquid has the droplet diameter smaller than $D = X$.

4 - the Nukiyama-Tanasawa distribution (first shown in 1939 [2]) is used for coarse sprays

(4) $$p(D) = BD^2 e^{-bD^{\lambda}},$$

where $B$ is a constant calculated using the $\Gamma$ distribution, $b$ – a dimensional parameter, $\lambda$ – a parameter characterising the distribution.

In any case, the initial step is to make a histogram of experimental data. Depending on the type of droplet-sizing instrument used, the distribution may be temporal or spatial. If the droplets are measured at a small particular location in the spray flux volume over some time interval (that is the case of single particle counters, SPC), the result is a temporal distribution. If a set of droplets is measured by an ensemble sizing system in a large sample volume at some instant, then for that time instant the whole group is characterized by a spatial distribution. To convert a spatial distribution into a temporal one (for the purpose of comparison between the two instruments), the velocities of droplets in the further case must be known. It means that the opposite conversion is not feasible unless supplementary information about the droplet velocities is supplied.

**Measurement techniques for droplet size distribution**

The knowledge of DSD is useful for characterizing a device (e.g. a nozzle), controlling a process, or to check a measuring system.

The measurement techniques for DSD measurements evolved from the techniques elaborated for solid particles. Those methods are mechanical (intrusive) and optical (non-intrusive). Some mechanical methods for solid particles can not be adapted to droplets (like sieving or sedimentation); some other (e.g. impaction) can. The classic optical methods comprise photography, microscopy, holography, and Particle Imaging Velocimetry (PIV). More sophisticated are the methods based on light scattering, which are laser-based.

Laser as a tool for getting droplet size information based on properties of scattered light was introduced about 1960. Its advantages include: nearly planar wave, monochromatic light, coherence, and spectral power sufficient for assuring proper amount of scattered light; gas lasers are preferred as more stable than diode ones. The laser-based methods can be categorized into amplitude- independent (e.g. phase-Doppler) and amplitude dependent. Another classification comprises single particle counting, and ensemble methods.

The theories supporting the scattered-light based methods are: the Lorenz-Mie theory (1908, often called Mie), the generalized Lorenz-Mie theory (GLMT), the Fraunhofer diffraction theory, and geometrical optics.

In 1964 Laser Doppler Anemometry technique was invented. The most popular single particle counting instrument of this type is the phase-Doppler particle analyzer (PDPA). The principle of operation is depicted in Figure 1. A laser beam is splited into two beams of the same wavelength, which are focused with lens and interfere within the beam waist region. When a droplet crosses the sample volume, it sends scattered light, which is collected in detectors. The interference fringe pattern reflected and refracted by the droplet conveys information of the spatial frequency of the fringes, which is a function of the optical parameters of the system, the refractive index and the diameter $D$ of the particle. The phase difference between the signals in the detectors is measured as the time interval $\Delta T$; the Doppler period of the signal is measured as $T$; then the phase shift $\Phi$ is equal: $\Phi = 2\pi\, \Delta T/T$. The phase shift $\Phi$ between detectors is a linear function of the droplet diameter $D$:

(5) $$D = \frac{\lambda}{2\pi F}\Phi$$

where: $D$ – the droplet diameter, $\lambda$ – the laser wavelength, and $F$ – a function of the optical configuration (the sinus and cosine functions of the angles $\theta$, $\psi$ and $\varphi$) and the relative refractive index between the droplet and the medium.

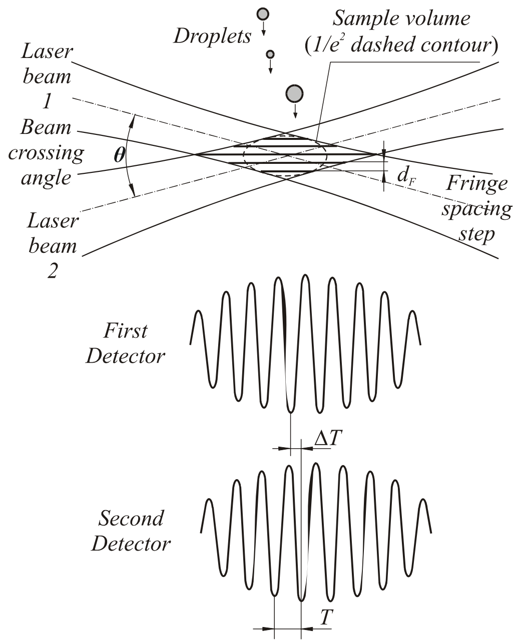


Fig.1. The optical arrangement and measuring principle of PDPA

This system also allows measuring the droplet velocity as proportional to Doppler frequency $f_D = 1/T$. For PDPA instruments, the measured droplet diameter ranges from 0,5 μm even to 10000 μm. This technique is restricted to spherical droplets only, since the measured signal depends on the local radius of curvature of the droplet [12].

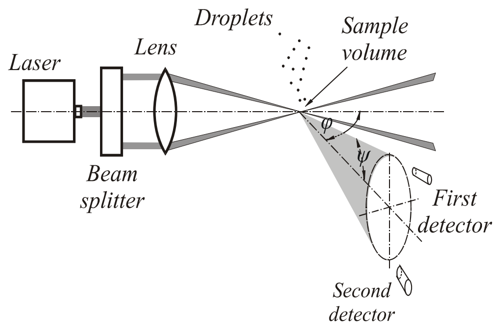


Fig.2. Schematic diagram of a PDPA instrument

The most popular ensemble method is based on the Fraunhofer diffraction (although most recently the supporting approximation algorithms are based on the Mie theory) [13]. Instruments based on this technique were introduced in the late 1970s (Fig.3).

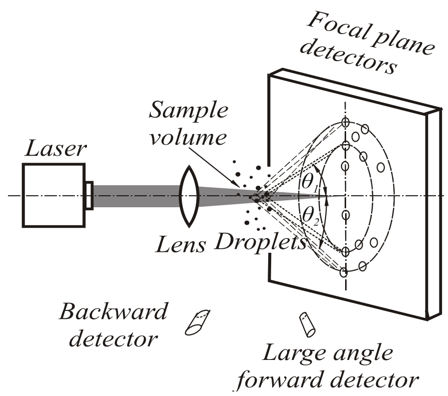


Fig.3. Schematic diagram of an instrument based on the Fraunhofer diffraction technique

A single droplet produces on the focal plane a pattern consisting of concentric circles – light and dark, alternately. The intensity $I$ depends on the Airy function:

$$\frac{I}{I(0)} = \left[\frac{2J_1(X)}{X}\right]^2, \tag{6}$$

where $J_1$ is the first-order spherical Bessel function, $I(0)$ is the scattered light intensity on the optical axis, and $X$ is a dimensionless parameter. Small droplets scatter light at large angles, while large droplets scatter light near the pattern centre. The Fraunhofer diffraction theory claims, that the DSD can be determined from the angular distribution of scattered light across the detectors (the optimum number of detectors is 16 to 32).

The range of measured droplet diameter is from 0,1 µm to to 2000 µm (e.g. accuracy 1 % - Spraytec, f-ma Malvern); but the method can be used only for spherical droplets.

For non-spherical droplets, the glare points (GP) techniques [15] are introduced, but the accuracy is less than the PDPA (a few percent is reported).

The charge of droplets is measured using the fall-velocity method [16], similar to the Millikan's method for measurement of the elementary charge. Also scaling laws, connecting water droplet diameter with the amount of charge induced on its surface in the process of electrospraying are derived [17].

**Conclusions**

The introduction of laser-based techniques allowed accurate *in-situ* measurement of droplet size and velocity, even in environments hardly accessible to other techniques. However, the particle-sizing laser-based instruments were originally designed for solid particle sizing, and only rarely are dedicated exclusively for spray droplet measuring.

The vast progress in measurement accuracy was made during last ten years, mainly due to considerable increase in computing power of software for processing the data collected by the systems. That has made possible the implementation of more sophisticated Mie theory into the algorithms for DSD computing.

Although the manufacturers claim that the laser-based particle-sizing systems do not require calibration as the operating principles are based on fundamental laws of optics, for the validation of measurements a check comparison against standard should be performed. As a rule, the standard is a wafer of latex particles (accuracy 1 %) and not a liquid droplet standard. The alternative is more reliable generator of monodispersed droplets, but the accuracy of checking becomes worse. The problem of calibration of droplet-sizing systems needs further considerations.

For the time being, there are no laser-based instruments dedicated for measuring DSD of sprayed water; just the relevant parameters of water should be entered when the instrument is adjusted for water droplets measurement. In the case of droplets of electrosprayed water, if would be useful to measure the droplet charge distribution simultaneously.

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