Behavior of the Spraytec in the Presence of Multiple Light Scattering and of Bimodal Drop Size Distribution

Triballier K., Cousin J., Dumouchel C.

UMR 6614 – CORIA, Université et INSA de Rouen, 76801 Saint-Etienne du Rouvray, FRANCE

The present paper reports an experimental investigation on the performances of the Spraytec commercialized by Malvern. The Spraytec provides a volume-based drop-size distribution from the analysis of a diffraction pattern resulting from the interaction between a spray and a laser beam. This work focuses on the behavior of the Spraytec in two different situations. First, the influence of multiple scattering due to the measurement of large and spatially inhomogeneous sprays is investigated. The results show that the use of the Malvern Spraytec for such sprays can not guaranty a result free of multiple-scattering effects Second, a test of multi-modal drop-size distribution measurement is conducted. In the tested situations, the behavior of the Spraytec was found reliable.

1. Introduction

One of the techniques often used to measure liquid spray drop-size distribution is based on the Fraunhofer diffraction light theory thanks to the development due to Swithenbank *et al* [1]. Since the late 70's, this technique has been commercialized by Malvern.

The Malvern particle sizer provides a line of sight average measurement of the volume-based drop-size distribution derived from the diffraction pattern that is recorded on the receiver. The mathematical inversion procedure can be conducted according to three different models that are available in the data analysis software: the Rosin-Rammler, log-normal and model independent models. The two first models assume that the distribution follows a Rosin-Rammler or a log-normal distribution, both of them being two-parameter distributions. The third model is not associated to any mathematical distribution and reports a distribution calculated on the basis of 15 independent parameters.

One of the main drawbacks of the first Malvern particle-sizer generation was the assumption of individual photons scattering off single particles. Although this assumption is valid in many practical situations, it is not satisfied anymore when the number of particles in the measuring volume is too high, either because the drop density is high or the spray is large. In such conditions, multiple light scattering affects the diffraction pattern by increasing the diffraction angles. As a consequence, the small drop population is overestimated and the resulting drop-size distribution is narrower than the actual distribution.

Since 1984, many experimental or theoretical studies have considered the problem of multiple scattering in laser diffraction particle sizing. Dodge [2] reported an experimental investigation on the influence of the spray length on drop-size distribution measured with a Malvern equipment. The length of the spray was varied thanks to the use of seven identical nozzles that were arranged along the line of sight of the laser beam and operated singly, two at a time, three at a time and so forth up to all seven operating simultaneously. In this work, the mathematical inversion procedure used the Rosin-Rammler model. These experiments reported a non-negligible multiple scattering effect for obscuration greater than 50%. In addition, as explained by Felton *et al.* [3], the effect of multiple scattering is not only a matter of obscuration but depends on the size distribution also.

The mathematical inversion procedure of the last generation of Malvern particle sizer, the Spraytec, contains two major differences compared to the previous versions. First, the Spraytec employs the Lorenz-Mie theory to account for the contribution to the angular light energy distribution of refraction through small droplets. This considerably improves the behavior of the instrument when measuring very fine sprays. The experimental work conducted by Corcoran *et al.* [4] on nebulizer sprays illustrates this point. It reported that instruments entirely relying on Fraunhofer diffraction overestimate the small drop population (< 3 µm) and that the Spraytec is much more reliable for this drop category thanks to the use of the Lorenz-Mie theory. Second, the Spraytec mathematical inversion procedure includes a patented multiple scattering algorithm that allows successful measurements in extremely high concentrations with light obscuration as high as 95% (5% transmission). An application of this algorithm, presented in the Malvern Spraytec documentation [5] shows that it successfully accounts for multiple scattering in the case of dense sprays. This equipment is now used in experimental investigations where drop-size distributions of dense sprays have to be measured (Simmons and Hanratty [6], Al-Sarkhi and Hanratty [7]). However, considering the complexity of multiple scattering through the review of the previous works, one may wonder whether the algorithm can successfully treat any situations including large sprays with non-homogeneous spatial distribution. The experimental investigation reported in this paper deals with this very problem conducting a series of drop-size distribution measurements of sprays with a constant drop size distribution but a varying width. This was achieved through an approach similar to the one conducted by Dodge [2]. Furthermore, previous works (Hamidi and Swithenbank [8]; Gülder [9, 10]) reported that the Malvern particle sizer was reliable for the measurement of bi-modal and tri-modal distribution. However this was checked in specific situations, i.e., controlled suspensions, spatially homogeneous, with a maximum diameter that did not exceed 250 µm. In the present work, a test of multi-modal distribution measurement is performed with the Spraytec for a real liquid spray characterized by a large drop size interval and a non-homogeneous spatial distribution.

2. Influence of multiple scattering

Six identical low-pressure gasoline injectors were used in this part of the work. The nozzle of these injectors is composed of four holes and the resulting spray is highly non-homogeneous in space. Throughout the study the liquid atomized was CSL2 that has similar physical properties as gasoline and injectors operated with steady state operations only.
For the relevance of the present work, the six injectors must produce sprays with similar characteristics. First, we verified that all the six injectors deliver the same static flow rate at any injection pressure. Second, drop-size distribution measurements were performed with a Spraytec for each injector at different injection pressures and distances from the injectors. Throughout the present study, the Malvern Spraytec was equipped with a 450 mm focal-length lens. A comparison of the injector performance is presented in Fig. 1. The measurements reported in this figure were conducted at ΔP_i = 5 bar and at a distance from the nozzle h = 30 mm. The six volume-based drop-size distributions are very much alike: the peaks are located at the same drop diameter, and both tail and width are identical from one

distribution to another. It has been also verified that the spray produced by the six injectors are very much alike even in terms of mean and characteristic drop diameters.

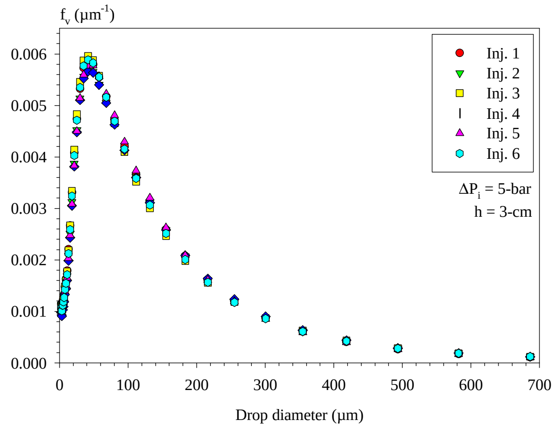

Fig. 1 Comparison of the drop size distribution of the 6 injectors

The aim of the present experimental study is to investigate the influence of the multiple scattering on Spraytec behavior when the length of the spray is increased. As done by Dodge [2], this was achieved by aligning the six injectors along the laser beam, and by making measurements with one operating injector, then two operating at a time, up to all six operating simultaneously. Such an experimental protocol is acceptable if the position of the spray in the measuring volume has no effect on the measurement. Due to possible vignetting, the length of the examination region of the instrument is limited within the working distance of the lens equal to 1.5 times the focal length of the Fourier lens. In the present case, the maximum working distance from the receiver lens is equal to 675 mm.
The six injectors were arranged on a straight feeding pipe that permitted to operate them individually. The distance between each injector was set to 60 mm. This arrangement allowed us to perform measurements down to 40 mm from the injector without observing any spray overlapping for all tested injection pressures. Thus, throughout the work, the spray evolved within the working distance of the Spraytec. Moreover, in order to control the validity of this arrangement, it has been verified that the volume-based drop-size distribution is not affected by the position of the injector on the feeding pipe.

Drop-size distribution measurements were performed at three distances from the injector (h = 20, 30 and 40 mm) and for four injection pressures (ΔP_i = 2, 3, 4 and 5 bar). For each operating condition, the spray produced by one single injector up to the six injectors operating simultaneously was measured provided that the transmission was not lower than 5%. This limit is imposed by Malvern. The drop-size distributions were first calculated without selecting the multiple scattering correction option. For each operating condition, the result obtained for one single injector is taken as the reference distribution throughout the analysis.

Figure 2 presents an example of the influence of the spray length on the mean drop diameter D_{43} as well as on the relative span factor Δ_V. As the spray length increases, corresponding to an increasing number of working injectors and a decreasing transmission, the measured diameters like D_{43} decrease. This behavior is representative of multiple scattering and was observed for all operating conditions. For 4 and 5 bar, the measurements with five and six consecutive injectors reported transmission below the 5% limit. In accordance with many previous investigations, it is observed that the influence of multiple light scattering has to be considered for transmission lower than 40%. The reduction of the mean diameters is a consequence of the overestimation of the small drop population. The corresponding increase of the relative span-factor indicates that the influence of multiple scattering is not the same for all drop categories. Paloposki and Kankkunen [11] showed that the influence of multiple scattering increases when the drop category characteristic-diameter decreases. Thus, as the transmission decreases, the diameter $D_{v0.1}$ is much more affected than the diameter $D_{v0.9}$ leading to a slight increase of the span-factor.

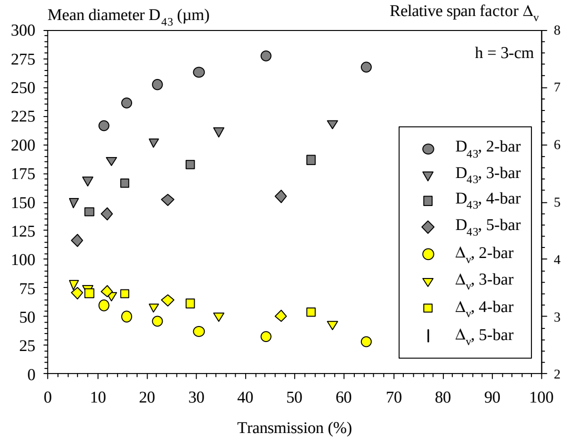

Fig. 2 Evolution of D_{43} and Δ_V with the transmission

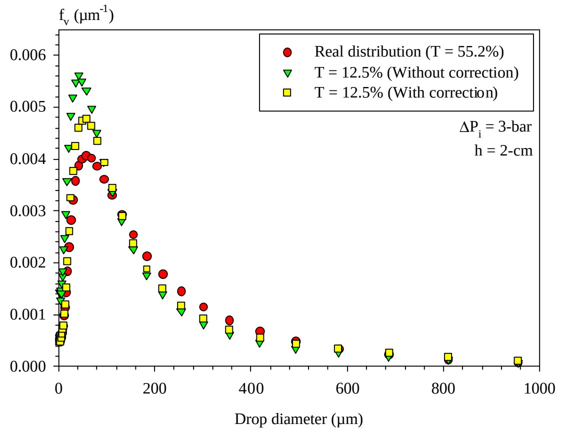

Fig. 3 Application of the multiple algorithm of the Spraytec

One of the objectives of the present work is to control the multiple light scattering correction-procedure provided by the Spraytec. To achieve this, all the measurements were analyzed once again with the Spraytec software by selecting this time the multiple light scattering option. An example of the influence of the Spraytec correction is shown in Fig. 3. This figure compares the uncorrected and the corrected drop-size distributions when four injectors were operating simultaneously (T = 12.5%). The real distribution (the one obtained with a single injector and calculated without selecting the multiple scattering option) is also presented for comparison. For this example it can be seen that the correction is not efficient enough. The corrected distribution is closer to the actual distribution than the uncorrected one but the small drop population is still overestimated.

The influence of the Spraytec correction was found to be a function of the transmission as shown in Fig. 4. This figure presents the evolution of characteristic diameters of the corrected distribution when the transmission diminishes for a constant spray drop-size distribution (ΔP_i = 3 bar, h = 2 cm, D_{43} = 234 µm). In this figure the characteristic diameters of the corrected distributions (denoted by a D without prime) are divided by the same characteristic diameter of the real distribution (denoted D’). Points located below the line D/D’ = 1 indicate insufficiently corrected diameters whereas points above this line correspond to overcorrected diameters. Surprisingly, it can be first observed in this figure that even with high transmission (55.2%) the correction shows an influence characterized by an increase of the characteristic diameters. For this case the three characteristic diameters are similarly affected by the Spraytec correction. We see that the influence of the correction is not negligible in a situation where it should be. Therefore the correction option should be prohibited when it is not required, namely, when T > 40%.

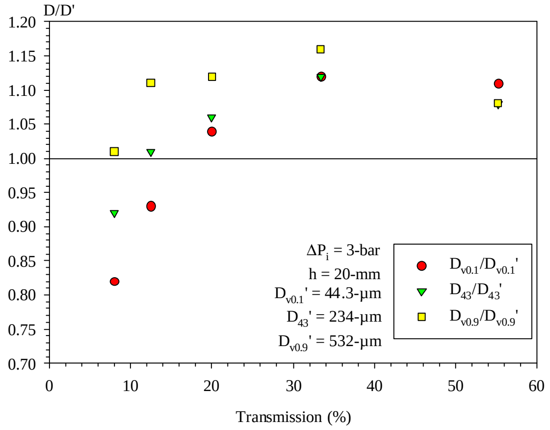

Fig. 4 Influence of the Spraytec correction on characteritic diameters

For medium transmission (33.4% and 20%) Fig. 4 shows that the corrected diameters are higher than the real values. Contrary to the highest transmission, the influence of the correction procedure depends now on the drop category. At T = 20% for instance, the overestimation of the multiple scattering effects is more pronounced for big drops than for small ones. When the transmission continues decreasing, this behavior amplifies. At T = 12.5%, the multiple scattering effects are underestimated for small drops, correct for medium drops and overestimated for big drops. Finally, for the lowest acceptable transmission for this case (T = 8%), the correction is appropriate for the big drop population but it is insufficient for all other drop categories.
In all tested situations, we can note that there is no transmission for which the correction is acceptable over the whole drop-size distribution.

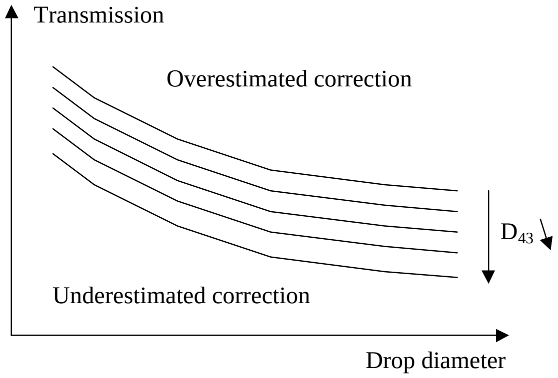

Fig. 5 Synthesis of the influence of the Spraytec multiple scattering algorithm

The obtained experimental results lead to conclude that the Spraytec multiple light scattering correction procedure is not acceptable for the kind of sprays examined here. The influence of the correction procedure can be summarized as schematized in Fig. 5. For a given spray, the correction is a function of both transmission and drop category. For a low transmission all drop categories are under corrected, for high transmissions all drop categories are over

corrected and, for medium transmissions, small drop categories are under corrected whereas big ones are over corrected. Thus, for a given spray, the limit between overestimated and underestimated corrections can be drawn as a monotonously decreasing line. Furthermore, the limit between underestimated and overestimated corrections translates downwards as the spray gets finer.
The fact that the multiple light scattering correction procedure of the Spraytec is not adapted to the sprays studied in the present work does not mean that this algorithm is incorrect. As shown in the Malvern documentation [5], the Spraytec algorithm seems appropriate when the spray density is high. In the present study, multiple scattering was the consequence of the use of large sprays. Furthermore, the sprays studied here were characterized by an inhomogeneous spatial distribution. This suggests that the influence of multiple scattering on diffraction technique particle sizing is complex and is a function of many spray characteristics that have not been all taken into account yet.

3. Multi-modal drop-size distribution

The aim of the experimental work presented in this section is to see whether the Spraytec equipment is reliable to measure multi-modal spray drop-size distributions. Previous works reported a reliable ability of the diffraction technique to measure bi-modal [8] and tri-modal [10] drop-size distributions. However, the situations examined concerned calibrated and homogeneous suspensions with a maximum particle diameter that did not exceed 250 µm. The present objective is to see whether the Spraytec behaves as well with real liquid spray, spatially inhomogeneous and with a large drop diameter interval.
To answer this question, the drop-size distributions of two different sprays were simultaneously measured. Each spray was first characterized independently. Second, the two sprays were aligned in the laser beam to be measured when operating at the same time. The drop-size distribution measured this way was then compared to the expected distribution calculated from the individual measurements weighted by the measured volume fraction of each spray.
The Spraytec receiver was equipped with a 200 mm focal length lens. Two identical swirl atomizers were selected. Swirl atomizers produce sprays through the disintegration of a conical and hollow liquid sheet. Therefore, in a plane perpendicular to the injector axis, the resulting spray drops are mainly concentrated in an annular section whose radius is a function of the conical liquid sheet angle and of the distance of the injector. Thus, the spray concentration and drop-size distribution are both highly spatially dependent.
Both injectors were connected to independent liquid feeding circuits and they were both used in steady state operations. One of the injector was fed with CSL2. The injection pressure was set to 10 bar and the drop-size distribution measurement was performed at 80 mm from the injector. For this injector the volume flow rate was equal to Q_c = 3.12 10^{-6} m^3/s. The second spray was a spray of water produced at 2 bar and the drop-size distribution was measured at 30 mm downstream the injector tip. The volume flow rate delivered by the second injector was equal to 1.62 10^{-6} m^3/s. To avoid interactions between drops and the subsequent drop recombination, the injectors were positioned at 140 mm from each other. Whatever the configurations, Spraytec measurements reported transmission not less than 45%. Such transmissions indicated the absence of unfavorable multiple light scattering effects. Then, as suggested previously, the Spraytec multiple scattering correction option for the present part of the study is deselected.
Figure 6 presents the individual drop-size distributions for the spray of CSL2 (f_{V1}) and for the spray of water (f_{V2}). The spray of CSL2 is mainly composed of small drops. The peak of the

distribution is located at around 50 µm. The spray of water carries bigger drops. The main peak diameter of this distribution is of the order of 200 µm. These results were expected as the CSL2 has a lower surface tension than the water and the spray of CSL2 was produced with a greater injection pressure. These two points favor the production of smaller drops.

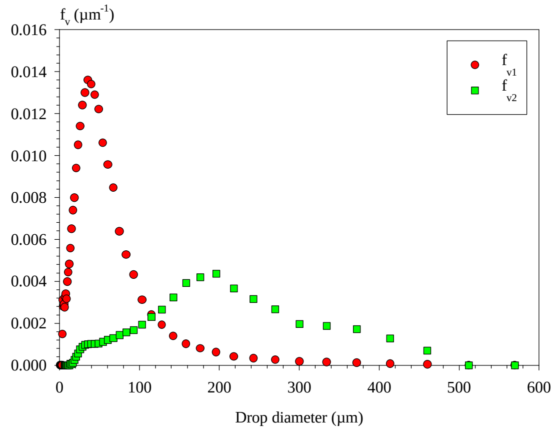

Fig. 6 Measured drop size distributions of sprays 1 and 2

Figure 7 presents the distribution resulting from the simultaneous measurement. It can be seen in this figure that the measured distribution shows two peaks located at 50 µm and 200 µm. To make sure that the measured distribution presented in Fig. 7 is representative, the expected multi-modal drop-size distribution can be estimated as follows.

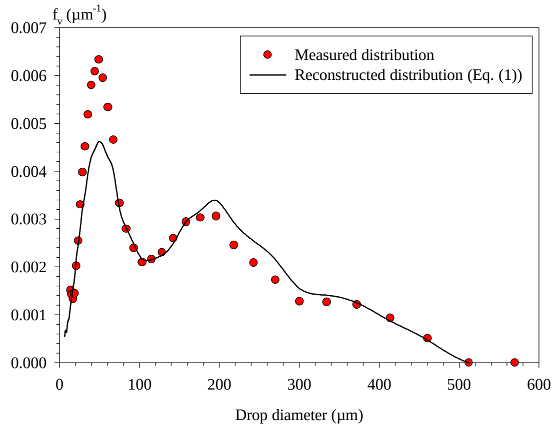

Fig. 7 Comparison between the measured and reconstructed distributions

At the measurement location, the laser beam embraced all the spray of water. Thus, the measured volume of water-spray per unit time was equal to the volume flow rate Q_W delivered by the injector. The spray of CSL2 was wider and it was partly covered by the laser beam. Per unit time, the measured volume of CSL2 was equal to βQ_c, where Q_c is the volume flow rate delivered by the injector and β is a weighting coefficient. This coefficient was determined on the basis of geometrical considerations and was found of the order of 0.13. Thus, the drop-size distribution obtained from the simultaneous measurement of the sprays is expected to be:

$$f_v(D) = \frac{\beta Q_c f_{v1}(D) + Q_w f_{v2}(D)}{\beta Q_c + Q_w}$$

where f_{v1} and f_{v2} are the drop-size distributions presented in Fig. 6. The drop-size distribution deduced from the previous equation is plotted in Fig. 7 and can be compared with the measured distribution. It can be noted that the two distributions agree reasonably to consider that the measurement is reliable. Indeed, the peak diameters as well as the distribution tails are in accordance. This result is important and indicates that the Spraytec is reliable to measure multi-modal spray drop-size distributions.

4. Conclusion

The work presented in this paper investigates the behavior of the Malvern Spraytec when large sprays or complex drop-size distributions have to be measured. Contrary to the previous Malvern particle sizers, the inversion procedure of the Spraytec is completed by a multiple light scattering algorithm. The application of this algorithm in the mathematical inversion procedure is optional. Multiple light scattering happens when the measuring volume contains a high number of drops either because the spray density is high or because the measuring volume is large. In the present work, the Spraytec multiple-scattering model was experimentally tested on large sprays but with a reasonable drop density. Furthermore, the sprays examined here presented inhomogeneous spatial distribution in terms of drop number and drop size.

The results show that the use of the Malvern Spraytec for the measurements of large and inhomogeneous sprays can not guaranty a result free of multiple-scattering effects. According to the information found in the Malvern documentation, the corrective algorithm is adapted for high concentrated sprays, namely, when multiple scattering is due to a high spray density. In the present case, the multiple scattering results from the spray dimension. For the sprays examined here, the influence of the Spraytec correction is found to be a function of the transmission, of the spray drop-size distribution and of the drop category.

The application of the Spraytec correction model performed in this work reported also that it should not be used when multiple light scattering is negligible. In accordance with previous works, it was found to be the case when $T > 40\%$. All these results show that the problem of multiple scattering in diffraction particle sizing techniques is not solved for any situations and must still be considered with care.

As far as the measurement of multi-modal distribution is concerned, the behavior of the Spraytec was found reliable. This result agrees with those of previous workers who reported the propensity of diffraction particle sizing technique to determine bi or tri-modal drop-size distribution of calibrated and spatially homogeneous suspensions.

5. References

[1] Swithenbank J Beer J M, Taylor D S, Abbot D and McCreath C G 1977 *Progress in Astronautics and Aeronautics* **53**, 421-447

[2] Dodge L G 1984 *Optical Eng.* **23**, 626-630

[3] Felton P G, Hamidi A A and Aigal A K 1985 *Proceedings of the ICLASS-85*, London

[4] Corcoran T E, Hitron R, Humphrey W and Chigier N 2000 *Journal of Aerosol Science* **31**, 35-50

[5] Malvern Website 2000 www.malvern.co.uk

[6] Simmons M J H and Hanratty T J 2001 *Int. Journal of Multiphase Flow* **27**, 861-883

[7] Al-Sarkhi A and Hanratty T J 2002 *Int. Journal of Multiphase Flow* **28**, 1617-1629

[8] Hamidi A A and Swithenbank J 1986 *Journ. Inst. Energy* **65**, 101-105

[9] Gülder O L 1987 *ILASS Americas 87* Madison USA

[10] Gülder O L *Aerosol Science and Techno*logy **12**, 570-577

[11] Paloposki T and Kankkunen A 1991 ICLASS'91, Gaithersburg, Paper **46**, 441-448