Two-Fluid Atomization and Spray-Impact

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Abstract
The possible applications of the spray-impact are versatile. For medical inhalers, in flame spectrometers or included in a spray dryer, the spray-impact can be used to produce smaller droplets in the small micrometer range. The mechanisms and phenomena after a single-droplet or spray-impact emerging from a single-fluid nozzle were examined by several researchers and the droplet formation mechanisms are well-understood.
This work focuses on the combination of a two-fluid nozzle and the spray-impact on a sphere. A two-fluid nozzle already produces a small amount of fine droplets (>5 µm) under moderate pressure conditions. However, the distribution is usually broad. Spray-impact can be used to increase the mass flow of the fine fraction. In addition, with two-fluid nozzles, the droplets are directly dispersed into the gas flow. The spray-impact was investigated experimentally and the influence of the atomizing parameters on the impact outcome was examined.
Two in-house build two-fluid nozzles with internal mixing are applied which show a good geometrical similarity. With the small-scale nozzle, an aerosol with a mass median droplet diameter of about 12 µm was produced under moderate conditions (Δp=5 bar). A multimodal distribution was measured after impact on a sphere (diameter 11 mm). However, an increased mass flow rate of small droplets (2 and 3 µm) was observed after impact for low liquid-to-gas mass flow ratios. For a detailed evaluation of the spray-impact outcome, a large-scale nozzle was applied, which produced an aerosol with a mass median diameter of about 80 µm (Δp=1 bar). In the large-scale set-up, a sphere with a diameter of 55 mm was used. After impact, the droplet size distribution was analysed at different regions around the sphere. There, the measurement position around the sphere after impact has no effect on the droplet size distribution. Compared to the aerosol before impact, the droplet size was reduced by the spray-impact. In addition, a smaller droplet size for decreased liquid-to-gas mass flow ratio and increased gas pressure was measured. The liquid film produced on the sphere surface during the impact influences the impact outcome. A characteristic film could be visualized during the experiments with the large-scale nozzle. However, a large variation in thickness was determined.
Finally, different droplet formation mechanisms such as splashing and crown formation were observed. The influence of different impact conditions on the mechanisms were analysed. The phenomena and mechanisms were compared to the investigations observed with single-fluid nozzles described in the literature.

Keywords
Spray-Impact, Two-Fluid-Nozzle, Primary Aerosols, Secondary Aerosols, Weber Number, Reynolds Number

Introduction

Spray-impact onto a solid surface is a key element in various technical applications, such as ink-jet printing, rapid spray cooling of hot surfaces, spray painting and coating [1]. The relevance of the spray-impact depends on the application [2]. In spray coating, the deposition of the liquid film on the impact surface is relevant and prevention must be done for the generation of secondary droplets [3]. In the previous decades, the research on spray impact had been performed using single-fluid atomizer. However, the investigation of the droplet impact outcome by means of a two-fluid nozzle is relevant for different technical applications. The spray-impact can be an opportunity to increase the mass flow of droplets in the small micrometer range. The production of submicron particles by spray drying processes demands droplets smaller than 3 µm. The formation of a secondary aerosol after impact should increase the fraction of useful droplets and should be applicable to produce nanoparticles for pharmaceutical application. [4]
Drop impact onto a solid dry surface or liquid surface has been investigated by many researchers for several years to study the phenomenon in detail which leads in recent advancement in high speed visualization, theoretical modelling and developing and improvement of numerical methods for the simulation of interfacial flows [1]. The parameters such as droplet size and velocity before impact, the impact angle, the liquid properties, the surface geometry and roughness affect the outcome of single droplet impact. The presence of a liquid film may especially influence the disintegration of the impinging droplet. The possible outcomes of the single droplet impact are prompt splash, corona splash, spreading or rebound based on the mentioned parameters above. [1,5–8] Typical values to describe the splashing threshold are the Weber and Reynolds numbers, which depend on the

droplet characteristics before impact. Additionally, correlations to predict the size of secondary droplets after impact were developed, considering the mentioned impact conditions.
Due to the interaction of droplets with different size and velocity and the complex hydrodynamic conditions, single droplet impact and the mechanisms that govern multidrop impact in a spray are difficult to connect [9]. One possibility for predicting spray characteristics after impact is using the principal of superposition i.e., simple extrapolation of many non-interacting single droplets using an Euler/Lagrange numerical simulation [2,10,11].
Nevertheless, from the experimental studies, the applicability and universality of these models on the multidrop or spray impact is highly limited due to the impact parameters and operating conditions [12,13].
Another possibility is the experimental investigation of the spray impact which enables the formulation of empirical correlations [1,3,13]. However, the validity of empirical correlations is limited to the used experimental conditions, such as nozzle type, initial spray characteristic and surface properties. Therefore, the empirical models are applicable to a narrow parameter range only. It has been observed, that the spray impact leads to an accumulation of droplets on the surface, forming a liquid film that in turn affects the velocity and size of the ejected droplets. [1,9] The application of spray impact using two-fluid atomizer is mostly found in the industry of inhalable pharmaceuticals products. In industrial applications like flame spectroscopy, smaller droplets were produced using two-fluid atomizer and the impact on sphere. This spray impact produces flow of secondary droplets known as secondary aerosols after splashing onto a rigid surface like sphere or wall. There is an increase in number of smaller droplets after splashing. It is relevant to study the mechanism of an impact producing flow of secondary droplets after an impact. In order to understand the complete impact phenomena for the generation of secondary aerosols, a detailed experiment investigation on the droplet size, velocity and mass flux near the impact surface and the influence of the process parameter and liquid properties is necessary.
An attempt is made to analyze the spray-impact outcome with a two-fluid nozzle and to compare the mechanisms with the literature.
Moreover, the use of two-fluid nozzles is of interest due to the direct dispersion of the produced secondary aerosol into a gas flow [14]. It is known that the velocity of secondary droplets after spray impact is small, compared to the primary droplets i.e. primary aerosols. Therefore, secondary droplets remain close to the impact surface and may be transported by the gas flow. The secondary droplet characteristics were analyzed in the area between the nozzle and impact surface in experimental investigations thus far. [3,9]
The spray-impact on a sphere with a two-fluid nozzle will be examined experimentally. Different process parameters and their effect and role on the secondary aerosol outcome were analyzed.

Material and methods

A schematic drawing of the experimental setup is presented in Figure 1. For all atomization and impact experiments, deionized water was utilized.
Two inner mixing two-fluid nozzles with different scales were used for the production of the primary aerosol. The liquid is introduced into the mixing chamber with seven capillaries with an inner diameter of 0.5 mm in the small-scale and 2 mm in the large-scale. The gas stream is introduced by two sidewise ports. The nozzle orifice was 1 mm in the small and 5 mm in the large-scale.
For the small scale, a pump was used for the liquid and the liquid flowrate ranges between 1.2-6 kg/h and the atomizing gas mass flow was measured with a flow meter and ranges between 1-4 kg/h. The geometrical similar larger scale nozzle was designed with a scaling factor of 5. A flow meter was used to adjust the liquid mass flow in the range of 60-100 kg/h and the gas mass flow ranges betwenn 11-17 kg/h. The liquid-to-gas mass flow ratio $\mu = \dot{m}_L/\dot{m}_G$ is a relevant value to describe the atomization process.

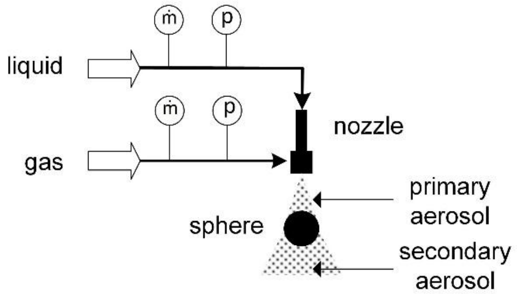

Figure 1 Schematic drawing of the spray impact setup [15]

The droplet size distribution of the primary and the secondary aerosol after impact was evaluated with a laser diffraction device (Spraytec, Malvern, Worcestershire, United Kingdom). The representative diameters of the distribution $d_{10,3}, d_{50,3}, d_{90,3}$, standing for the diameters of the 10, 50 and 90 % quantile of the distribution were extracted. The width of the distribution can be characterized with the span value $(d_{90} - d_{10})/d_{50}$. In the small scale, the mass of the fines (2 and 3 µm) was calculated by extracting the representative quantile Q_3 from the distribution: $\dot{m}_{L,<2,3\,\mu m} = Q_3(2{,}3\,\mu m) \cdot \dot{m}_{feed}$. The droplet size for the primary aerosol was measured at a distance of 20 cm for the small- scale and 15 cm for the large-scale in order to ensure a representative measurement. The secondary aerosol was characterized after impact directly at the sphere.

In order to analyse the film deposition on the sphere, a high-speed camera (VEO 410L, Phantom, Wayne, New Jersey, USA) was utilized with a lens focal length of 105 mm and frame rates between 10 and 15 kHz. The contour of the film was evaluated with the image analysis tool ImageJ. The film was extracted from an image and superimposed on a picture of the dry sphere. With the known pixel-to-length ratio, the line tool of ImageJ was used to the determine the thickness of the film.

Results and discussion

Small scale nozzle

Droplets in micrometer range are produced using the small-scale two-fluid nozzle. For the applications such as inhalers and spray dryers, droplets in the small micrometer range are important. The droplet size distribution and the mass flow of small droplets were analyzed for the secondary aerosol. Figure 2 shows the generation of secondary aerosol after the spray impact.

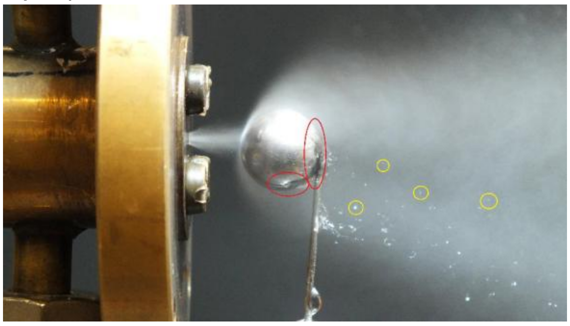

Figure 2: Spray impact on a sphere in the small-scale. Red circles: Film generation and droplet entrainment. Yellow circles: Entrained large droplets in the secondary aerosol. Sphere diameter 11 mm; distance between nozzle orifice and sphere: $\mathbf{5\ mm}$; differential gas inlet pressure: $\mathbf{\Delta p}$ =5 bar; feed mass flow: $\mathbf{2.6\ kg/h}$. [15]

Due to the spray-impact an increased mass of smaller droplets was observed. With the volumetric cumulative distribution of the primary and secondary aerosol an increase in mass of smaller droplet can be quantified (see Figure 3). For the primary aerosol, the mass of small droplets increases until a liquid-to-gas mass flow ratio of 5 and then remain constant. The increase in liquid feed rate is proportional to the mean droplet size and the decrease in mass of fine droplets is evident. For higher liquid-to-gas mass flow ratios the mass flow of fine droplets decreased and led to fluctuations.
After impact, the mass of fine droplets increases until a liquid-to-gas mass flow ratio of 2 and decreases afterwards. This is due to the increase in thickness of the liquid film formed on the surface with a higher feed flow rate.
Overall, the spray impact leads to an increase of the fines by a factor of 2 in the small liquid-to-gas mass flow ratio range, which is the common working range of pneumatic nebulizers. Additionally, a multimodal distribution was observed after spray impact for every impact condition.

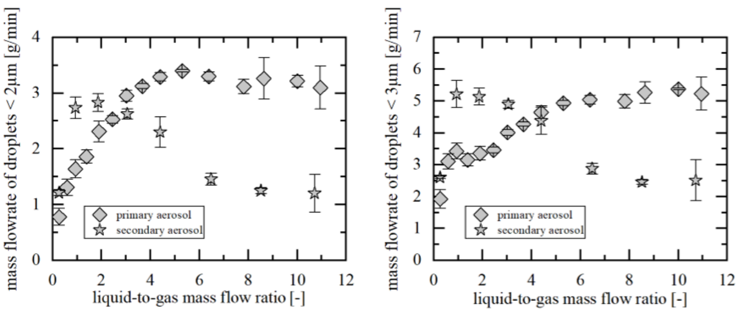

Figure 3: Mass flow rate of droplets < 2µm (left) and < 3µm (right) as a function of the liquid-to-gas mass flow ratio. [15]

Large scale nozzle

A detailed view into the mechanisms of spray impact in a small scale two-fluid nozzle is challenging due to the small scale and the micrometer size range. A geometrical similar larger-scale nozzle was used to characterize and understand the outcome of the secondary aerosol.
The volume frequency is presented for different liquid-to-gas mass flow ratios for the secondary aerosol in Figure 4:

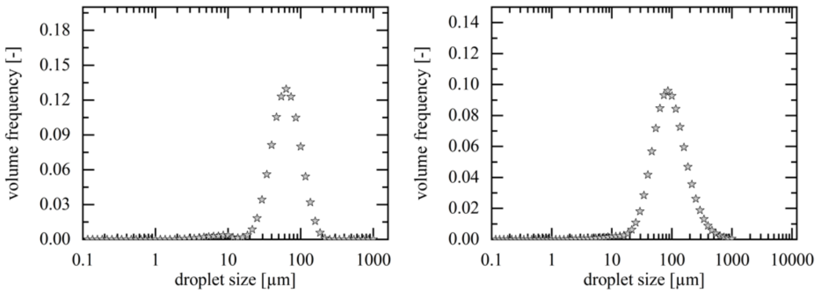

Figure 4: Volumetric frequency distribution of the secondary aerosol for a liquid-to-gas mass flow ratio of left: 4 and right: 9. [15]

After the spray impact, a unimodal distribution was observed, and larger droplets were produced when the liquid-to-gas mass flow ratio was increased. The distribution was much narrower with a span found to be 1.2 for a liquid-to-gas mass flow ratio about 4 compared to the higher liquid-to-gas mass flow ratio with a span value of about 2.3 (see Figure 4). The characteristic diameter of the distribution of the primary and the secondary aerosol was chosen as the mass median diameter $d_{50,3}$. The mass median droplet diameter of the primary and the secondary aerosol was compared for different liquid-to-gas mass flow ratios (see Figure 5).

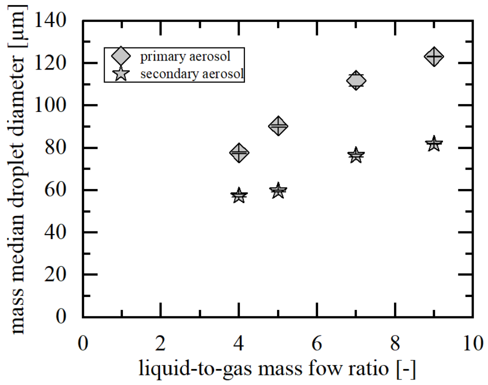

Figure 5: Mass median droplet size for the primary and the secondary aerosol as a function of the liquid-to-gas mass flow ratio (av ± s, n = 3) [15]

For the primary aerosol the droplet size increases with increasing liquid-to gas mass flow ratio. This behaviour was expected for the atomization with two-fluid nozzles [16]. The mass median droplet size increases with increased liquid-to-gas mass flow ratio. With the secondary aerosol the same trend and a smaller droplet size was measured. The multimodal distribution at the small-scale spray-impact may be due to the water flowing down the sphere resulting in bigger droplets in the distribution. The larger-scale enables the measurement of a more specific region around the sphere, avoiding bigger droplets in the measurement region.

After the impact of primary aerosol, the liquid film forms onto the surface of the sphere. The thickness of the liquid film plays an important role in the generation of the secondary aerosol and the splashing mechanism and may enhance the splashing outcome. The spray impact and the film generation were visualized clearly, at an instant in time is shown in Figure 6.

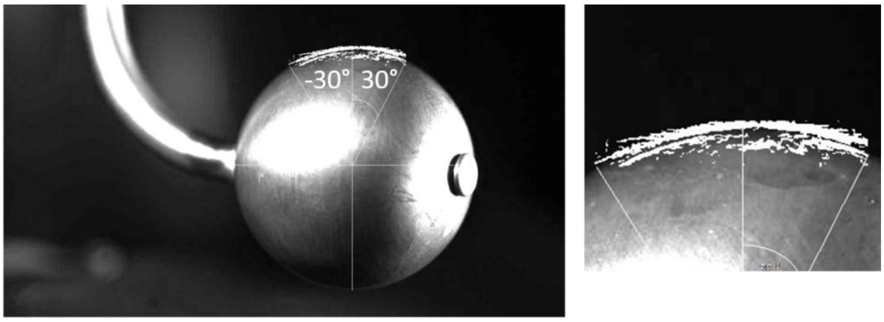

Figure 6: Left: Image of the spray impact with the film generation on the sphere. Right: detail view of the liquid film [15]

Under various spraying conditions, the film thickness was analysed. Thickness was measured at different positions of the film and the variation of thickness with its position is shown in Figure 7. In the experiment conducted within the operating range the film thickness was fairly high, ranging between 100 and 1300 μm (see Figure 7). It is evident that thickness at the center of the sphere is larger compared to that at the sides. As the center of the sphere faces the impact directly, the thickness is expected to be larger at that point. This is true for all the operating conditions implying at all liquid-to-gas mass flow ratios and at all pressures. The film thickness was calculated by using the average film thickness of three points in time for the same impact conditions. The variation in thickness is high. Due to the uncertainties in the evaluation of the film thickness and the fluctuations of the film, the measurement error is high. However, a tendency was shown that the film thickness decreases with increased gas inlet pressure and increases for increasing liquid-to-gas mass flow ratios.

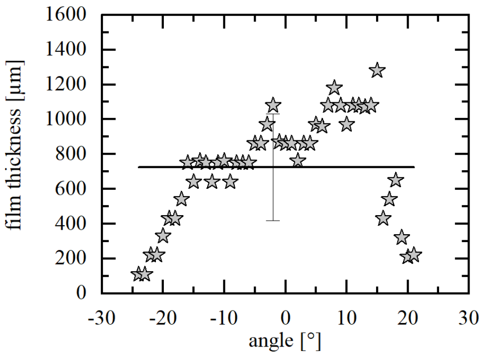

Figure 7: Film thickness as a function of the position over the impact surface. Stars: film thickness along the surface, line: mean film thickness with the standard deviation. (liquid-to-gas mass flow ratio: 4) [15]

Various outcomes have been observed after primary aerosols impacts onto the sphere and generate secondary aerosols. The outcome of the spray impact depends on the interaction of several process parameters such as differential gas pressure and liquid-to-gas mass flow ratios. The additional gas flow in the two-fluid nozzle may influence the splashing and crown formation. In general, for the large-scale atomizer, splashing, the generation of finger-like jets, crown interaction and spreading on the impact surface were observed. Since the range of parameter and atomized liquid is broad, a general statement predicting the outcome of spray impact is challenging. High speed camera was used for the observation of possible outcomes for the liquid-to-gas mass flow ratio and differential gas pressure range. Dimensionless numbers (impacting Reynolds and Weber number) separates the boundaries of the outcome of the spray impact. Splashing is observed for liquid-to-gas mass flow ratio of 4 and 5. At lower liquid-to-gas mass flow ratios, the kinetic energy of the drops is more as compared with higher ratios, leading the spray in splashing. The crown formation has been observed for water to air mass flow ratio of 7 and 9. Below this ratio, this phenomenon was not observed. At higher liquid-to-gas mass flow ratio, the thickness of the liquid film increases. Due to increases in the thickness of the liquid layer, splash gets weakened. Additionally, kinematic discontinuity has been observed at the impact region which results in a crown like sheet at the surface of the liquid film. From the crown like sheet, secondary aerosols are formed after an impact as outlined in Figure 8.

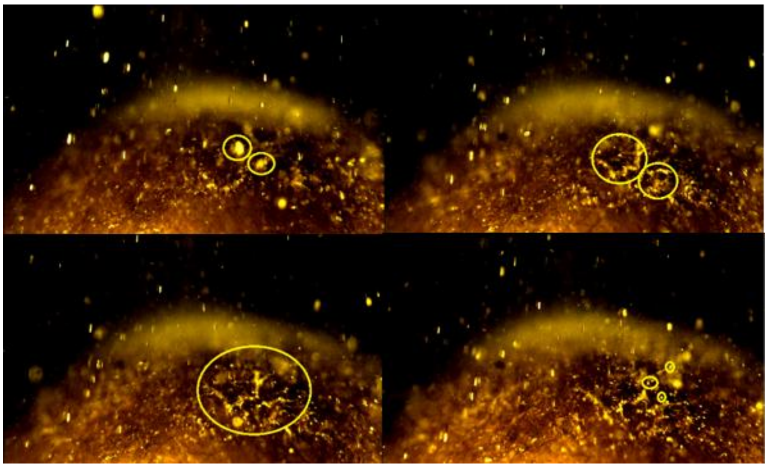

Figure 8: Representative moment recording of the crown interaction for a liquid-to-gas mass flow ratio of about 7. ($d_{50,3,\,PA}$= 111 µm, impact velocity= 61 m/s, differential gas pressure= 1 bar) [15]

After the impact of primary aerosol onto the surface, spreading had been observed for lower liquid-to-gas mass flow ratios as represented in Figure 9. Due to the higher kinetic energy of the droplets, spreading was observed. Higher kinetic energy result in spreading has been confirmed with experimental results of [17] in the application of spray coating on paper.

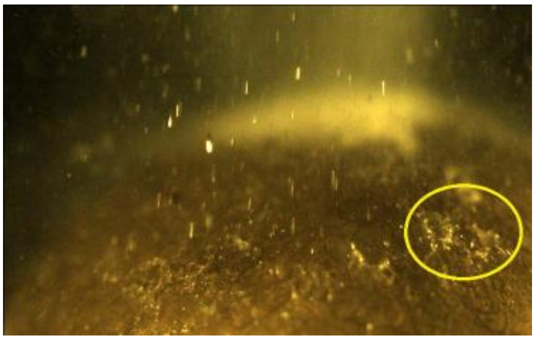

Figure 9: Moment recording of the spreading on the sphere surface [15]

Conclusion:

The spray impact was studied using the concepts derived from a single and the multiple drop impact onto a solid surface. In the literature, it was found that the spray impact onto a solid surface produces secondary droplets using single-fluid nozzle. The mechanism for the generation of secondary droplets was not well understood.

In this work two-fluid nozzles in two size scales were examined. It was possible to increase the mass flow of fine droplet (<3 µm) for small liquid-to-gas mass flow ratios due to the spray-impact on a sphere.

However, the mechanisms were not visible in the small scale. With a large-scale nozzle the mechanisms during impact were measured and visualized. The spray-impact outcome depends on different process parameters relevant for two-fluid atomization, such as the liquid-to-gas mass flow ratio, the differential gas pressure and the distance between the nozzle orifice and the impact surface.

The effect of liquid-to-gas mass flow ratio on the droplet size of primary and secondary aerosols was studied. With increase in the ratio, the droplet size for both also increases.

A film was produced on the sphere surface and the film thickness was measured at various locations onto the sphere for all the operating conditions. The average was taken for all the thickness measured at a given operating condition. With increase in liquid-to-gas mass flow ratio, film thickness increases while on increasing nebulization pressure and consequently a higher gas flow rate, the film thickness decreases.

The study of spray impact using two-fluid nozzle was compared to the mechanisms associated with single-fluid nozzles. Mechanism such as splashing and crown formation could be visualized by using the two-fluid nozzle. The combination of two-fluid atomization with an impact sphere is an opportunity to increase the mass flow of fine droplets. This process can be relevant for different technical applications, e.g. for spray drying of nanoparticles. However, in order to understand the entire impact mechanism, the influencing parameters should be varied, and the influence of the impact surface investigated.

Nomenclature

Δp	differential pressure [bar]
$d_{50,3}$	mass median diameter [µm]
$\hbar$	average film thickness [µm]
μ	liquid-to-gas mass flow ratio [-]

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