Spray drying of oil-in-water emulsions: oil droplet breakup during the atomization by pressure-swirl atomizers

Martha L. Taboada*, Heike P. Karbstein, Volker Gaukel
[1]Institute of Process Engineering in Life Sciences, Chair of Food Process Engineering,
Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany
*Corresponding author: martha.taboada@kit.edu

Abstract

The atomization of oil-in-water emulsions for spray drying purposes is a common task in the food industry. The oil droplet size in the resulting powder is of upmost importance as it affects the physical stability and sensorial properties of the product. Pressure-swirl nozzles are widely used in the food industry as atomization devices. The intense stresses acting on the liquid feed upon pressure-swirl atomization can lead to size change of the dispersed oil droplets. In the present study, model food oil-in-water emulsions were used to evaluate the influence of pressure-swirl atomization on the size of the dispersed oil droplets. Emulsions with varying initial droplet size were atomized at pressures ranging from 5 to 20 MPa by means of commercial hollow-cone pressure nozzles with different inlet geometries. Oil droplet breakup was visible for almost all atomization conditions, being the resulting oil droplet size highly dependent on the atomization pressure. In contrast, no effect of the atomizer design on the oil droplet size could be observed. Results of this study also show that the resulting oil droplet size after atomization can be controlled in a wide range almost independently of the spray droplet size.

Keywords

Atomization, pressure-swirl atomizer, oil droplet breakup, emulsion

Introduction

Spray drying is a widely used processing technique for the production of food powders such as infant formula and instant dairy powders [1]. In this process, atomization is used to create fine droplets from a liquid feed, which are dried into particles by subsequent contact with a hot air stream. Powders with encapsulated oily components can be produced by spray drying when an emulsion is used as the liquid feed. In this case, the oil droplets are dispersed in a continuous phase and remain entrapped in a solid matrix material after drying of the solvent [2]. The size of the dispersed oil droplets in the resulting powder is of upmost importance, as it affects the encapsulation efficiency, the physical stability of the product and the sensorial properties of the reconstituted food.

Several researches have studied the change of oil droplet size in emulsions during the atomization step for different types of atomizers [3–8]. However, in spite of their widespread industrial use, very few studies have systematically studied the oil droplet breakup during pressure-swirl atomization [5]. Most of the studies found in literature on atomization of emulsions with pressure-swirl atomizers focus on the spray characteristics and not on the change of the dispersed phase [9–11]. During pressure-swirl atomization, a swirling motion is imparted to the liquid feed, which is then strongly accelerated through a narrow gap to leave the atomizer and spread out in the form of a conical sheet [12]. As intense shear and elongational stresses act on the liquid feed upon pressure-swirl atomization, a deformation and breakup of the dispersed oil droplets can be expected.

From the emulsification literature it is known that dispersed droplets are disrupted when the local deformation stresses exceed the droplet capillary pressure and the deformation time exceeds the critical breakage time [13]. The capillary pressure is directly proportional to the interfacial tension, and inversely proportional to the droplet radius [13]. In laminar flow, droplet breakup in emulsions can be described by means of the capillary number Ca (Equation 1). For droplet breakup to occur, a critical value of the capillary number Ca_{cr} has to be exceeded. This value is known to depend on the viscosity ratio between the dispersed phase and the continuous phase [14] (Equation 2). The latter is replaced by the viscosity of the emulsion in case of emulsions of high dispersed phase content [15]. The relationship between the critical capillary number and the viscosity ratio depends on the type of flow acting on the droplet (simple shear or elongational flow) [14,16]. In the case of simple shear flow, a minimum value of the critical capillary number is obtained for viscosity ratios between 0.1 and 1. At values above 4 no droplet breakup is possible. In elongational flow, the critical capillary numbers are much lower and droplet breakup is not limited by high viscosity ratios. This theory on droplet breakup is valid for fully developed, stationary flows [14]. In real systems like spray nozzles, this might not be the case, and different flow types may occur simultaneously and/or

in sequence. For effervescent atomizers, for example, the simultaneous occurrence of shear and elongational stresses has been shown [4].

The resulting droplet size of the dispersed phase is not only a function of the droplet disruption. After disruption, newly created droplets may recoalesce [13]. The superimposed effect of coalescence on droplet disruption can only be excluded for very low dispersed phase fractions, and when the new droplets can be stabilized quickly enough by the emulsifier in the system. This has been shown for the droplet breakup during atomization with effervescent atomizers [17].

$$Ca = \frac{\eta_c \cdot \dot{\gamma} \cdot r}{\sigma} \tag{1}$$

With: viscosity of continuous phase η_c, shear rate $\dot{\gamma}$, droplet radius r, interfacial tension σ

$$\lambda = \frac{\eta_d}{\eta_c} \tag{2}$$

With: viscosity of continuous phase η_c and viscosity of the dispersed phase η_c.

Various types of pressure-swirl atomizers have been developed for different applications. Their main difference is the way the swirl motion is imparted to the liquid feed. They include atomizers with tangential inlet holes and swirl chambers (Fig. 1b), and atomizers that impart swirling by means of axial helical slots (Fig. 1a) [12]. Differences in the atomizer geometry may lead to different elongational and shear stresses upon atomization and hence to different oil droplet breakup.
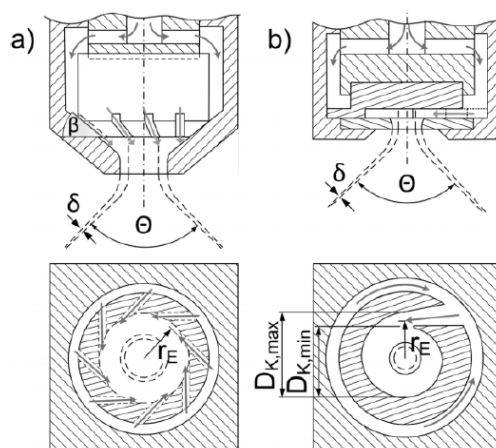

Figure 1. Schematic representation of hollow cone pressure-swirl atomizers with axial inlet with helical slots (a) and tangential inlet (b) [18].

The goal of this study was to evaluate the influence of pressure-swirl atomization on the size of the dispersed oil droplets of a model food oil-in-water emulsion. The impact of atomization parameters such as the atomization pressure and inlet design of the atomizer was studied, as this influences the stress history on the oil droplets. For this, emulsions with different initial droplet size consisting of maltodextrin, whey protein and medium-chain triglycerides (MCT) oil were atomized in an atomization rig and the resulting oil and spray droplet size distributions were analysed.

Material and methods
Model emulsions
For the performed investigations, food model oil-in-water emulsions were prepared. The continuous phase consisted of demineralized water and maltodextrin (Cargill C*Dry™ MD 01910, Germany). Medium chain triglycerides oil (WITARIX® MCT 60/40, Germany) was used as the dispersed phase and whey protein isolate (Lacprodan DI-9224, Arla Food Ingredients, Denmark) was used as emulsifier.

The emulsions were prepared in a two-step process. In the first step, the initial droplet size of the emulsions was adjusted by homogenizing a premix consisting of MCT-oil (50 wt.-% dispersed phase), water and whey protein isolate (5 wt.-%). Fine emulsions with initial Sauter mean diameter (SMD) of 20 ± 1.5 µm and 3.3 ± 0.2 µm were prepared by processing the premix for two minutes in a colloid mill (IKA magic LAB®, IKA®-Werke GmbH & Co. KG, Germany) at a gap width of 0.32 and 0.16 mm and a circumferential speed of 8.6 and 26 m/s respectively. Fine emulsions with a SMD of 0.24 ± 0.003 µm were prepared by means of a high pressure homogenizer (M110-Y,

Microfluidics) at 500 bar. The oil droplet size distribution of the emulsions was measured by laser diffraction spectroscopy (HORIBA LA950, Retsch Technology GmbH, Germany).

In the second step, the feed emulsion for atomization was prepared by dilution. A solution of water and maltodextrin was used to dilute the fine emulsions to a dispersed phase content of 1 wt.-%. This concentration was chosen to exclude coalescence or coagulation phenomena of the oil droplets after atomization [17]. The concentration of maltodextrin after dilution was 35 wt-% by which the viscosity of the emulsion was adjusted to 35 mPa·s. Viscosities were measured by rotational rheometry (Physica MCR 101/301, Anton Paar, Austria) with a double gap geometry (DG26.7) at 20 °C. For this, a logarithmic shear rate controlled ramp of $1 - 1000$ s^{-1} was performed. All the studied emulsions presented Newtonian behaviour. Due to the low concentration of oil in the system, the viscosity of the emulsion and of the continuous phase were virtually the same. As the viscosity of the MCT-oil was 28 mPa·s, the viscosity ratio in the model system was 0.8, which is in the optimal area for droplet breakup in shear flow [14].

Atomization test rig and atomizers

For the atomization experiments, a spray test rig was used as depicted schematically in Figure 2. A high pressure three-piston pump (Rannie Lab, Denmark) was used to supply the solutions through the atomizer at pressures ranging from 5 to 20 MPa. The corresponding flow rate was measured with a flow meter (VSE0, 04/16, VSE GmbH, Germany). To avoid blockage of the nozzle orifice a metal filter with a mesh size of 440 µm was installed before the atomizer entrance. Prior to atomization, the emulsions were tempered to 20 °C in a jacketed vessel to ensure a constant viscosity during the experiments. The emulsions were gently stirred during the experiments with a propeller stirrer to avoid creaming of the oil droplets.

Spray droplet size was measured inline by means of a laser diffraction spectroscope with a 750 mm focal lens (Malvern Instrument, Malvern, UK) over a time of 25 s and with a data acquisition rate of 250 Hz. This resulted in 6250 recorded droplet size distributions, from which an averaged distribution was calculated. The measurements were performed 25 cm underneath the nozzle orifice, perpendicular to the spray cone axis. Emulsion samples were taken prior and after atomization and the oil droplet size was analysed offline by laser diffraction. For each experiment, two separately prepared emulsions were atomized, from which three samples were taken at each atomization condition.

To ensure that the pump and its periphery have no effect on the initial emulsion size distribution (prior to atomization) a sample was taken before the nozzle entry and the oil droplet size distribution was measured and compared with that of the initial emulsion. No difference between the two samples were observed, confirming that there is no change of the emulsion droplet size prior to atomization under the studied conditions.
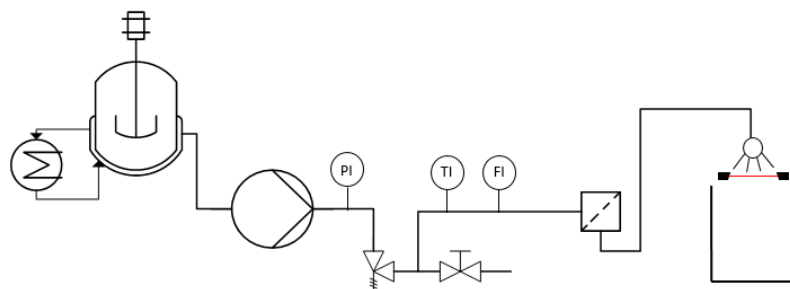

Figure 2. Schema of the experimental setup used for the atomization experiments

Atomization experiments were performed with commercially available, pilot scale hollow-cone atomizers with different inlet designs. Two nozzles with axial inlet and a grooved swirl body were used: the nozzle Schlick 121V (referred as Schlick in this work) from Schlick® with an orifice diameter of 0.3 mm, and the nozzle SKHN-MFP SprayDry® (core size 16, referred as SK) from Spraying Systems Co.® with an orifice diameter of 0.34 mm. Although both nozzles have the same swirl generation principle, the slots of the SK nozzle are substantially broader and shorter as the slots of the Schlick nozzle. This could result in a different oil droplet breakup upon atomization. A third hollow-cone nozzle with a single tangential inlet orifice and a swirl chamber was used: the Mini SDX® nozzle with a swirl chamber 32933-4 and an orifice disc 902-18 from Delavan® with an orifice diameter of 0.4 mm. From the broad variety of nozzles available on the market, these nozzles were selected due to their relevance in chemical and food engineering processes [1]. The throughput characteristics for different liquid pressures of the studied nozzles are shown in Table 1, where differences in the resulting volume flow for the different nozzles are evident. Experiments at 20 MPa were not possible with the Mini SDX nozzle due to maximal volume flow restrictions of the used high pressure pump (50 L/h).

Table 1. Volume throughput for the studied commercial pressure-swirl atomizers by the atomization of an emulsion with a viscosity of 35 mPa·s at 20 °C

	Atomization pressure / MPa	Volume flow / L/h
Schlick	5 / 10 / 20	9.6 / 15.3 / 20.4
SK	5 / 10 / 20	18 / 26.1 / 32
Mini SDX	5 / 10 / -	28.2 / 41.8 / -

Results and discussion

Influence of atomization pressure and inlet atomizer design

The resulting oil droplet size distributions after the atomization of emulsions with an initial SMD of 3.3 µm with the three pressure-swirl nozzles at different pressures are depicted in Figure 3 (left). It can be observed, that with increasing atomization pressure, the oil droplet size distribution of the emulsion shifts to smaller sizes. A significant decrease in the oil droplet size can be seen even at a relatively low atomization pressure of 5 MPa. The spray droplet size distributions are also depicted In Figure 3 (right), where a reduction of the spray droplet size with increasing pressure can be observed.

The results depicted in Figure 3 imply that the capillary pressure of the oil droplets is not high enough to overcome the high stresses the emulsion is subjected to upon atomization. This results in a reduction of the oil droplet size. For pressure-swirl hollow cone nozzles it is well know that an increase in the atomization pressure leads to an increased velocity of the liquid feed and to a reduction of the film thickness at the exit of the nozzle [12]. Therefore, higher shear and elongation rates are to be expected as the atomization pressure is increased. This consequently results in increased droplet breakup, as observed in this study. A decrease in the oil droplet size with increasing energy input of atomization was also reported by other authors for different atomizers [3,6,17].
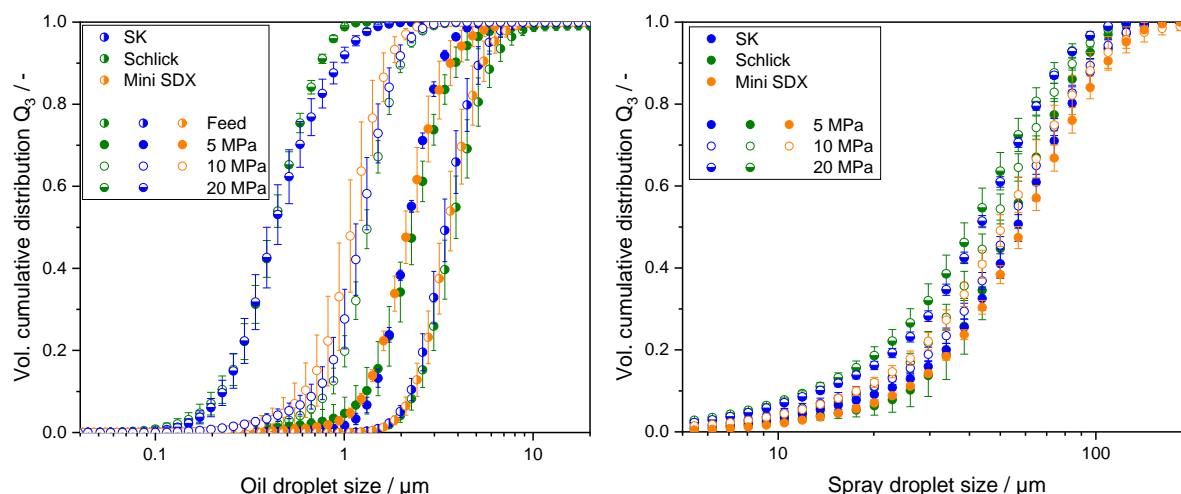

Figure 3. Oil droplet size distributions after atomization (left) of emulsions with initial SMD of 3.3 µm and viscosity of 35 mPa·s, and spray droplet size distributions (right) after atomization by means of different commercial pressure-swirl atomizers at pressures from 5 to 20 MPa.

Despite the difference in their geometry and the resulting volume flows, it can also be observed that there is no difference in the resulting oil droplet size distributions for the studied commercial atomizers. Analysis of variance (ANOVA) was carried out for each atomization pressure, in which the oil-SMD after atomization with the three atomizers were compared. No significance difference ($P < 0.05$) between the oil-SMD was observed. Different oil droplet breakup could be expected depending on the nozzle design. For example, higher elongational rates are expected in the nozzle inlet for the nozzles with axial inlet slots (SK and Schlick). The small diameter of the slots compared to the diameter of the inlet orifice of the Mini SDX would result in a higher acceleration of the liquid. Furthermore, as the slots of the nozzle Schlick are clearly longer as the ones from the nozzle SK, longer deformation times could be expected. Moreover, lower shear rates could be expected in the outlet of the Mini SDX due to its larger outlet orifice. However, the obtained results imply that the liquid feed was subjected to a similar stress history in the three atomizers, resulting in similar oil droplet breakup. This is also supported by the resulting spray droplet size distributions, as for each atomization pressure quite similar spray sizes are obtained for the three atomizers. But for a detailed discussion of these results, the exact stress and residence time profiles in these atomizers would have to be determined.

From Figure 3 it also clear that in the studied pressure range, the impact of the atomization on the spray droplet size is in relation much lower than the impact on the oil droplet size. This can be further appreciated in Figure 4, in which the resulting SMD for the oil and spray droplets after atomization at different pressures is depicted exemplary for the nozzle SK. It can be seen, that the SMD of both oil and spray droplets vary with the atomization pressure following the expression shown in Equation 3.

$$SMD = C \cdot P_L^{-b} \tag{3}$$

For the atomization with pressure-swirl nozzles this relation between spray SMD and atomization pressure has already been reported in literature. The constant C has been shown to depend on the viscosity, and values of the exponent b between 0.27 and 0.4 have been reported [12,19]. From the field of emulsification, a very similar expression for the variation of the SMD of the dispersed phase with the emulsification energy input is also well known [19]. Here, C is reported to depend on the viscosity of the dispersed phase, while the exponent b depends on the stresses the droplets are subjected to: for laminar stresses b takes a value of 1, while for turbulent breakup it takes values between 0.2 and 0.4 [20]. The energy input for emulsification would correspond in this study to the atomization pressure. In this study the exponent b takes a value of 0.25 for the spray droplets and 1.1 for the oil droplets, which suggests differences in the breakup mechanisms of the spray and oil droplets upon atomization. Thus, the oil droplet size after atomization can be controlled almost independently of the spray droplet size.
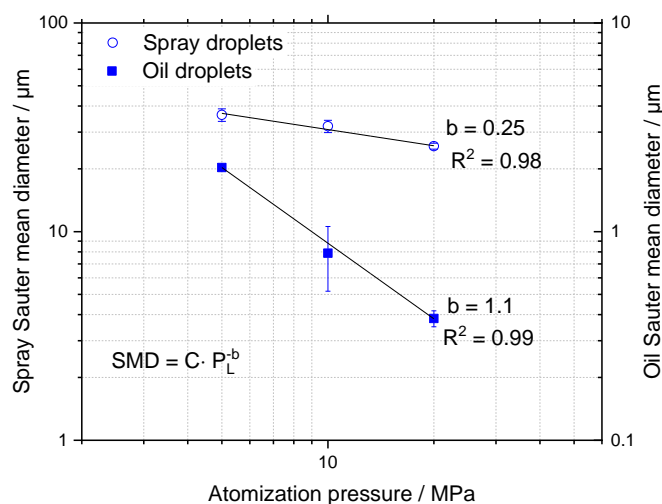

Figure 4. Oil and Spray Sauter mean diameters for emulsions with initial droplet size of 3.3 µm and viscosity of 35 mPa·s; Atomized with the nozzle SK at pressures from 5 to 20 MPa at 20 °C

Influence of emulsion initial droplet size
The resulting SMD with increasing pressure for the atomization with the nozzle SK of emulsions with different initial oil SMD are depicted in Figure 5. The nozzle SK was selected for these experiments due to its relevance in the food industry [1]. However, from the previous sections it is expected that these results also apply for the different atomizers. It is evident, that for emulsions with an initial droplet size of 3.3 and 20 µm the SMD decreases with increasing atomization pressure. In contrast, the SMD of emulsions with initial SMD of 0.24 µm remains unchanged upon atomization. In this case, the capillary pressure of these droplets was probably high enough to overcome the stresses upon atomization, by which no further breakup was achieved [21].

Interestingly, there is no significant difference in the resulting SMD of emulsions with initial droplet size of 20 and 3.3 µm. Other studies with a similar emulsion system showed that increasing the initial droplet size leads to larger droplets after atomization with an effervescent atomizer [7]. To explain these differences, stress histories would have to be evaluated for both atomizer types. Differences in stress and residence time profiles in the atomizers may be responsible for these effects, as larger droplets require longer deformation times to reach the deformation state corresponding to the applied stress. So, In the case of pressure-swirl atomizers, the residence time in the high stress areas is probably long enough to achieve droplet breakup to the equilibrium value even for the larger droplets. Thus, the same oil-SMD is obtained for these two emulsions.
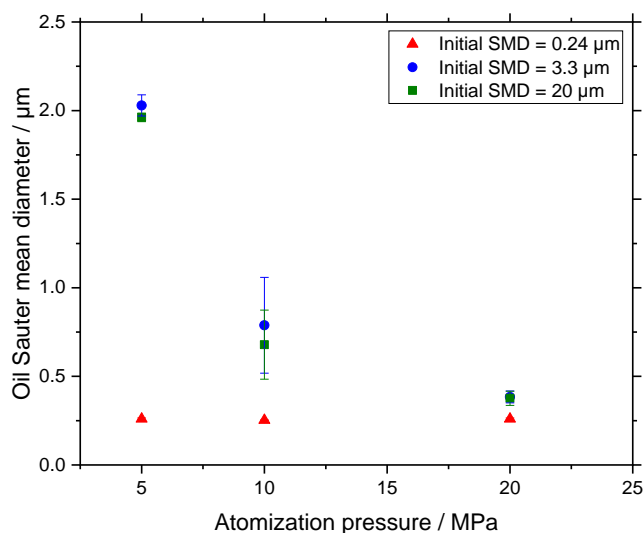
Figure 5. Sauter mean diameter for emulsions with different initial oil SMD after atomization pressures of 5, 10 and 20 MPa with the nozzle SK. Emulsion viscosity: 35 mPa·s

Conclusions

In this study it was shown that the stresses acting on the liquid feed during pressure-swirl atomization are high enough to deform and break the dispersed phase droplets of model food oil-in-water emulsions. The atomization pressure has the highest impact on the breakup of the emulsion droplets, while no influence of the nozzle inlet design and swirl principle generation could be observed. Increasing the initial oil droplet size did not lead to larger droplets after atomization, probably due to sufficient residence times of the droplets in the high stress areas of the nozzle. For a detailed discussion of these results, stress-time profiles would have to be deducted from local flow conditions. Results of this study also show that by adjustment of the atomization pressure, the desired oil droplet size after atomization can be controlled almost independently of the spray droplet size. This is of high relevance for spray dried emulsions in which physical stability and sensorial characteristics are influenced by the oil droplet size in the resulting powder.

Acknowledgements

Nomenclature

b	fitting parameter
C	fitting parameter
D_K	swirl chamber diameter [m]
Ca	capillary number
Ca_{cr}	critical capillary number
r	droplet radius [m]
r_E	radial distance to the inlet [m]
SMD	Sauter mean diameter [µm]
P_L	atomization pressure [MPa]
β	angle of the nozzle inlet [rad]
δ	film thickness [m]
λ	viscosity ratio
η_c	viscosity of the continuous phase [Pa·s]
η_c	viscosity of the dispersed phase [Pa·s]
$\dot{\gamma}$	shear rate [s^{-1}]
σ	interfacial tension [N/m]
Θ	spray angle

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