

ORIGINAL ARTICLE

Oil droplet breakup during pressure swirl atomization of food emulsions: Influence of atomization pressure and initial oil droplet size

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Abstract

Atomization of emulsions with pressure swirl atomizers is a common task in food process engineering. Especially in spray drying processes for food materials like dairy products, it is the technology of choice. During atomization, emulsions are subjected to high stresses, which can lead to deformation and breakup of the dispersed droplets. In this study, the influence of atomization pressure (5–20 MPa) and initial oil droplet size (0.26, 3.1, and 20.8 μm) on the oil droplet breakup during atomization of food based oil-in-water emulsions with pressure swirl atomizers was investigated. It was shown that a significant oil droplet breakup takes place upon atomization. The size of oil droplets with an initial value of 3.1 and 20 μm was reduced up to 0.36 μm . No breakup of oil droplets with an initial value of 0.26 μm was observed. The breakup was highly dependent on the atomization pressure. The results were analyzed based on existing knowledge on droplet breakup in laminar flow. A concept to estimate capillary numbers during atomization was developed based on common models from different applications. The results of this study can be used to control the resulting oil droplet size after atomization with pressure swirl atomizers.

Practical application

Spray drying of emulsions is a widely used process in the food industry to produce products with encapsulated oily components. Product examples include infant formula, milk powder, and the encapsulation of aroma and coloring compounds. Breakup of the oil droplets during the atomization step of spray drying can change a previously adjusted and desired oil droplet size. As the oil droplet size in the final product can be responsible for several properties like sensorial aspects and stability, a control of oil droplet breakup is essential. Pressure swirl atomizers are widely used in industrial applications as atomization devices. In this study, oil droplet breakup during atomization with these atomizers was investigated. The findings in this study allow a better control of the oil droplet size during atomization in practical applications.

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1 | INTRODUCTION

Spray drying of emulsions is a common task in food engineering for the production of products with encapsulated oily components. Typical examples include products such as coffee creamers, infant formula, and the encapsulation of active ingredients, aroma, and coloring compounds (Reineccius, 2004). The process of spray drying starts with the atomization, by which the liquid emulsion is dispersed into small spray droplets. These droplets are subsequently dried to powder by contact with a hot air stream. Pressure swirl nozzles are widely used as atomization devices in the food industry (Barbosa-Cánovas et al., 2005). After drying, the oil droplets should be encapsulated in a matrix material, which acts like a barrier, providing protection against oxidation or losses.

During atomization, emulsions are subjected to intense stresses, which do not only deform and atomize the feed, but can also lead to deformation and breakup of the dispersed droplets therein. A breakup of the dispersed oil droplets results in a modification of a previously adjusted oil droplet size distribution (ODSD). The ODSD affects the stability of the powder and of the reconstituted emulsion, as well as the functional properties of the product. For example, the release and bioavailability of active compounds are directly related to the oil droplet size (McClements & Li, 2010). In addition, the oil droplet size determines the color impression of food coloring powders (Haas et al., 2019). Furthermore, oil droplet breakup during atomization has been correlated to a reduced encapsulation efficiency in the powder (Jafari et al., 2008). Therefore, it is of utmost importance to control oil droplet breakup during atomization.

Breakup of oil droplets during atomization of oil-in-water (O/W) emulsions has been already studied for different types of atomizers: Schröder et al. (2012) and Kleinhans et al. (2016) studied oil droplet breakup for atomization with effervescent atomizers and an air core liquid ring atomizer. Munoz-Ibanez et al. (2015) studied oil droplet breakup during atomization with rotary and external mixing pneumatic atomizers. In these studies, the breakup was shown to depend on the energy input of atomization, as well as on the initial oil droplet size and on the viscosity ratio of the emulsions. Few studies using pressure swirl atomizers have also reported breakup of the disperse phase during atomization of emulsions (Bolszo et al., 2010). However, in spite of their wide industrial use, this aspect has not been systematically studied yet for pressure swirl atomizers. Most of the studies found in literature on these atomizers focus on the spray characteristics and not on the changes of the disperse phase (Davanlou et al., 2015; Tratnig et al., 2009).

The atomization principle of pressure swirl atomizers is based on the conversion of pressure to kinetic energy. In this type of atomizers the liquid flows through tangential holes or slots into a swirl chamber, and then to a discharge orifice (Walzel, 2003). Due to swirling motion of the liquid, an air core is created that extends from the rear of the swirl chamber to the discharge orifice. In the orifice, a thin liquid film is formed, which then leaves the atomizer in the form of an annular sheet that spreads to a conical hollow spray (Lefebvre & McDonnell, 2017). A schematic view of a pressure swirl atomizer is

depicted in Figure 1. Acceleration of the liquid due to the diameter contraction is expected to result in elongational stresses in both radial and axial directions. In the liquid film at the atomizer orifice, high shear stresses are expected due to the high velocities and the proximity to the wall. By means of numerical simulations of the internal flow in commercial pressure swirl atomizers, Renze et al. (2011) demonstrated that shear rates up to $\sim 100,000 \text{ s}^{-1}$ and elongational rates up to $50,000 \text{ s}^{-1}$ occur in the liquid film close to the nozzle exit for pressures of 0.2 MPa. These stresses can also lead to deformation and breakup of the disperse droplets in emulsions.

In order to estimate the stresses in the liquid film at the atomizer orifice, knowledge of the film thickness t is required. This information is not readily available and is not easy to determine experimentally at relevant industrial conditions. Several analytical and empirical correlations are available in the literature to estimate this value. A widely used theoretical model for the estimation of t is given by Suyari & Lefebvre (1986) in the form of Equation (1),

$$t = C \left(\frac{2r_o \dot{m} \mu}{\rho \Delta p_L} \right)^{\frac{1}{3}} \quad (1)$$

where r_o corresponds to the nozzle orifice radius, \dot{m} to the mass flow rate, μ to the liquid viscosity, ρ to the liquid density and Δp_L to the pressure differential during atomization. According to the original correlation by Rizk & Lefebvre (1985), the constant C is 3.66. The constant was corrected by Suyari & Lefebvre (1986) to 2.7 to better match experimental data. The correlation predicted the film thickness with high accuracy up to pressures of 3 MPa. Other recent studies with different atomizer geometries, pressures, and liquid properties have found that the expression estimates the liquid film thickness fairly well (Laurila et al., 2019; Wimmer & Brenn, 2013).

From the theory on breakup of dispersed droplets it is known that for droplet breakup the external forces imparted by the surrounding fluid must overcome the droplet capillary pressure (Karbstein & Schubert, 1995). When the external stresses are of simple shear nature, the breakup is characterized by the capillary number Ca :

$$Ca = \frac{\mu_c \dot{\gamma} X}{\sigma} \quad (2)$$

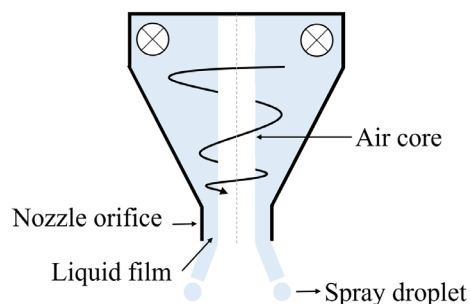


FIGURE 1 Schematic representation of a pressure swirl atomizer

where μ_c is the viscosity of the continuous phase, $\dot{\gamma}$ the shear rate, x the droplet radius, and σ the interfacial tension between the continuous and the disperse phase. In the case of elongational flow, the shear rate is replaced by the elongational rate $\dot{\epsilon}$ in Equation (2). For breakup to occur, a critical value of the capillary number Ca_{cri} has to be exceeded (Grace, 1982). This value depends on the viscosity ratio between the disperse and the continuous phase μ_d/μ_c . The denominator is replaced with the emulsion viscosity for emulsions with high disperse phase fractions (Armbruster, 1990). The correlation of Ca_{cri} with the viscosity ratio depends on the type of flow acting on the droplet interface. Grace (1982) characterized the relationship between the viscosity ratio and Ca_{cri} for simple shear and elongational flow at quasi-steady state. For simple shear, Ca_{cri} has a minimum value at viscosity ratios between 0.1 and 1. When the viscosity ratio exceeds a value of 4, no breakup is possible. For elongational flow Ca_{cri} is not limited by high viscosity ratios, and the values of Ca_{cri} are much lower than in shear flow. This theory, however, is based on a fully developed, stationary flow, which does not correspond to the flow conditions in spraying nozzles. Nevertheless, Munoz-Ibanez et al. (2015) demonstrated that the oil droplet breakup in rotary and pneumatic atomizers could be predicted fairly well with the calculated values of Ca_{cri} from Grace (1982).

For droplet breakup, it is also necessary that the droplet deformation time τ_{def} exceeds a critical value $\tau_{def,cri}$, that correlates with the droplet viscosity μ_d divided by the deformation stress, see Equation (3) (Walstra & Smulders, 1998):

$$\tau_{def,cri} \approx \frac{\eta_d}{\mu_c \dot{\gamma}} \quad (3)$$

In emulsions with high phase content, the resulting droplet size is not only a function of droplet breakup, but also of coalescence. In this study, the effect of coalescence was excluded by working at very low disperse phase fractions.

The aim of the present work was to investigate the impact of pressure swirl atomization on the oil droplet size of food emulsions. Specifically, the influence of the atomization pressure and the initial oil droplet size were investigated. For this purpose, the ODS of emulsions before and after atomization were compared. Additionally, stresses in the atomizer and capillary numbers were estimated in order to analyze the results based on the theory of droplet breakup in laminar flow.

2 | MATERIALS AND METHODS

2.1 | Model emulsions

Model food oil-in-water emulsions with different oil droplet sizes were prepared for the investigations. The continuous phase consisted of demineralized water, maltodextrin (Cargill C*Dry™ MD 01910, Germany), and whey protein (Lacprodan DI-9224, Arla Food Ingredients, Denmark) as emulsifier. Whey protein and maltodextrin were

used to resemble typical industrial formulations for encapsulation by spray drying. The disperse phase consisted of medium chain triglycerides oil (MCT oil, WITARIX MCT 60/40, Germany). All reported mass fractions refer to the mass of the total emulsion.

In a first step, fine emulsions consisting of 50 wt% MCT oil, 5 wt% whey protein and 45 wt% water were prepared. For this, whey protein was dissolved in water and the pH was adjusted to seven with a 0.5 M solution of NaOH. The solution was stored overnight to ensure complete hydration of the protein. Emulsions with different Sauter mean diameters (SMD) were prepared by homogenizing MCT oil with the protein solution. A colloid mill (IKA magic LAB, IKA-Werke GmbH & Co. KG, Germany) operated at a gap width of 0.32 and 0.16 mm and a circumferential speed of 8.6 and 26 m/s was used to obtain emulsions with a SMD of $20.8 \pm 2.3 \mu\text{m}$ and $3.1 \pm 0.08 \mu\text{m}$, respectively. A high-pressure homogenizer (M110-Y, Microfluidics) operated at 500 bar was used to obtain emulsions with a SMD of $0.26 \pm 0.01 \mu\text{m}$. In a second step, the fine emulsions were diluted with a solution of maltodextrin in water to obtain the emulsions for atomization. The dilution ratio of maltodextrin solution to emulsion was 4.9:1. The concentration of maltodextrin after dilution was 34.3 wt%, the protein concentration was 0.1 wt% and the oil concentration was 1 wt%. At this low oil concentration, coalescence of the oil droplets after atomization can be excluded (Walstra & Smulders, 1998). The two-step procedure was performed to produce a large amount of emulsion for the experiments with the exact oil droplet size in a fast manner.

ODSD were measured with laser diffraction spectroscopy (HORIBA LA950, Retsch Technology GmbH, Germany). The scattering data were analyzed using the Mie theory with a standard optical model for MCT oil in water. The ODS of emulsions were measured before and after dilution with the maltodextrin solution. No differences in the ODS were observed between the concentrated and diluted emulsions.

2.2 | Physical properties

Viscosities (μ) were measured with rotational rheometry (Physica MCR 101/301, Anton Paar, Austria) with increasing shear rate from 1 to $1,000 \text{ s}^{-1}$ at 20°C by means of a double gap geometry (DG26.7). In the inspected shear rate range the viscosity of the emulsions and of the oil were found to be independent of the shear rate. The viscosity of the MCT oil at 1000 s^{-1} was $28.8 \pm 0.2 \text{ m Pa}\cdot\text{s}$, while the viscosity of the emulsions was $32.3 \pm 1.3 \text{ m Pa}\cdot\text{s}$. The viscosity of the emulsion was used to calculate the viscosity ratio, as the viscosity of the emulsion and of the continuous phase are virtually the same. Thus, the viscosity ratio of the model system was 0.9, which is in the optimal region for droplet breakup in shear flow (Grace, 1982). Due to the Newtonian behavior of the emulsions, this value is expected to remain constant during atomization.

Besides viscosity, other relevant physical properties of the emulsions were determined. The density ρ of the emulsions was measured with a tensiometer (DCAT 21, DataPhysics Instruments GmbH,

Germany). The interfacial tension σ between the continuous and disperse phase was measured with the pendant drop method (OCA 15 LJ, DataPhysics Instruments GmbH, Germany). Reported values are the measured interfacial tension after 1 hr. A summary of the physical properties is given in Table 1. All measurements were performed in triplicate at a temperature of 20°C.

2.3 | Pressure swirl nozzle

A pressure-swirl atomizer of the type SKHN-MFP SprayDry (core size No. 16, orifice insert No. 80) from Spraying Systems was used for the atomization experiments. These nozzles have the advantage of providing relatively low flow rates, which allows their application in pilot scale, while having a very similar geometry to industrially sized nozzles. The nozzles from Spraying Systems are widely used in spray drying of food products (Barbosa-Cánovas et al., 2005). The nozzle consists of an orifice insert with conical shape, a slotted core and a nozzle casing. The used orifice insert has a diameter of $d_o = 0.34$ mm. The slotted core consists of two slots with a nominal width of $d_s = 0.41$ mm. A photograph of the nozzle configuration is depicted in Figure 2.

2.4 | Atomization of emulsions

Atomization experiments were performed in a spray rig equipped with a high-pressure three-piston pump (Rannie LAB, Typ 8.5) to pump the emulsions through the atomizers. All atomization experiments were performed in triplicate. Prior to atomization, the emulsions were tempered to 20°C in a double wall vessel to ensure constant physical properties during atomization. To avoid creaming of the oil droplets, the emulsions were gently stirred in the vessel during the experiments with a propeller stirrer. A metal filter was installed before the atomizer entrance to avoid blockage of the exit orifice. The emulsions were atomized at pressures p_L of 5, 10, and 20 MPa and the corresponding liquid flow rates Q_L were measured with a flow meter (VSE0, 04/16, VSE GmbH, Germany). This pressure range was chosen because it covers the typical range in industrial applications. The pressure was measured with an analog pressure gauge (KOBOLD Messring GmbH, Germany) with a reading accuracy of 0.5 MPa. The applied liquid pressures as well as the measured flow rates are given in Table 2. To

ensure that the filter and the pump periphery have no effect on the initial ODS, emulsion samples were taken right before the nozzle entry. No significant difference was observed between the ODS of these samples and of the initial emulsions. During atomization, a sample of the spray was taken with a beaker ~25 cm below the nozzle exit. The oil droplet size of the emulsion was measured offline with laser diffraction spectroscopy (HORIBA LA950, Retsch Technology GmbH, Germany), as described before.

After atomization, the size distribution of the spray droplets was measured. Spray droplets are atomized emulsion droplets in which the oil droplets are dispersed (see Figure 1). Spray droplet size distributions were measured inline using a similar setup as in previous studies (Kleinhans et al., 2016). The spray rig was equipped with a laser diffraction spectroscope (Spraytec, Malvern Instruments GmbH, Germany) with a 750 mm focal length lens. The laser was placed perpendicular to the nozzle axis center line at a distance of 25 cm underneath the exit orifice. Spray droplet size distributions were measured for 30 s at each atomization condition. The diffraction patterns



FIGURE 2 Pressure swirl nozzle SKHN-MFP. left: slotted core, orifice insert, and nozzle casing. up right: slotted core (front view). Down right: orifice insert (back view)

TABLE 1 Physical properties of the liquids measured at 20°C (viscosity of emulsion μ_e , and disperse phase μ_d , liquid density ρ , interfacial tension σ) and corresponding relative uncertainty values u_r

Physical property		$u_r/\%$
$\mu_e/\text{Pa}\cdot\text{s}$	0.032	4.06
$\mu_d/\text{Pa}\cdot\text{s}$	0.029	0.69
$\rho/\text{kg}/\text{m}^3$	1,153.7	0.15
$\sigma/\text{N}/\text{m}$	0.012	5.28

TABLE 2 Liquid pressures p_L supplied to the pressure swirl atomizer and corresponding liquid volume flow rates q_l with their relative uncertainty values $u_r(Q_L)$

Atomization pressure p_L	Volume flow Q_L	$u_r(Q_L)$
MPa	L/h	%
5	20.5	6.2
10	28.8	1.9
20	32.6	2.4

were processed according to the Fraunhofer theory and a time averaged mean value was calculated.

3 | RESULTS AND DISCUSSION

3.1 | Oil droplet size after atomization

To assess the influence of the atomization pressure on the oil droplet breakup, emulsions were atomized at pressures p_L of 5, 10, and 20 MPa. The resulting mean volume cumulative distributions of oil droplet sizes after atomization at the different pressures are depicted in Figure 3. The results correspond to atomized emulsions with an initial Sauter mean diameter (SMD_i) of 3.1 and 20.8 μm . It can be seen, that even at a relatively low pressure of 5 MPa, the oil droplets after atomization are significantly smaller compared to the oil droplets in the feed emulsions. These results imply that the stresses the emulsions are subjected to during atomization are high enough to overcome the capillary pressure of the oil droplets, resulting in breakup of the dispersed oil droplets. A decrease in the oil droplet size with increasing atomization pressure is also observed. Several authors have reported a decrease in the oil droplet size with increasing energy input for atomization for different atomization systems (Kleinhans et al., 2016; Munoz-Ibanez et al., 2015; Schröder et al., 2012). In the case of pressure swirl nozzles an increase in the atomization pressure is expected to increase the velocity of the liquid and also reduce the film thickness at the atomizer outlet (Lefebvre & McDonell, 2017). Therefore, the stresses that lead to oil droplet breakup are expected to increase with increasing atomization pressure.

It can also be noted from the results shown in Figure 3 that very similar ODSD resulted after atomization of emulsions with SMD_i of 3.1

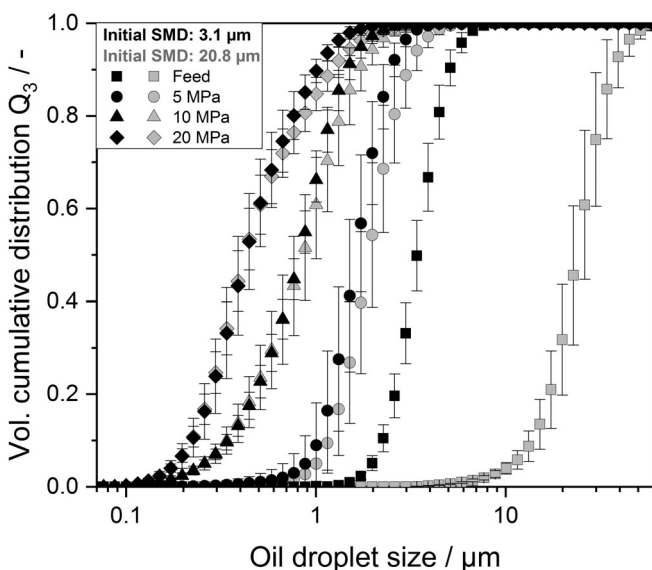


FIGURE 3 Cumulative oil droplet size distributions after atomization of emulsions with $SMD_i = 3.1 \mu\text{m}$ (black) and $SMD_i = 20.8 \mu\text{m}$ (gray) at pressures of 5, 10, and 20 MPa

and 20.8 μm at each studied pressure. From these results it is clear that the oil droplets are broken up to the same value independently of their initial droplet size. This effect can be further seen in Figure 4, where the resulting SMD for these emulsions are depicted. Analysis of variance (ANOVA) was carried out to compare the resulting SMD for each pressure, and no significant difference ($p < .05$) between the SMD of emulsions with different initial droplet size was observed. To further investigate the influence of the initial oil droplet size on the breakup behavior during atomization, emulsions with SMD_i of 0.26 μm were also atomized. The resulting SMD are also depicted in Figure 4. In the case of these submicron droplets, the SMD remained unchanged at all studied atomization pressures, indicating that no breakup of the oil droplets took place during atomization. The results imply that in the case of the small, submicron droplets, the capillary pressure is large enough to overcome the external stresses during atomization.

In the emulsification literature, the SMD of the disperse phase correlates with the energy input for emulsification according to the expression in Equation (4). In this equation, C is a constant that depends on the viscosity, and the exponent b gives insights on the breakup mechanisms of the disperse phase: for breakup due to laminar stresses, b takes a value close to one. For breakup due to inertial (turbulent) stresses, b takes values between 0.2 and 0.4 (Karbstein, 1994).

$$SMD = C \cdot p_L^{-b} \quad (4)$$

To evaluate the breakup mechanisms of the oil droplets during atomization, the resulting oil SMD were correlated to the expression in Equation (4). In this study, the energy input for emulsification corresponds to the atomization pressure. The resulting constants C and b for the emulsions with different SMD_i , as well as the coefficient of determination R^2 are summarized in Table 3. The resulting fit is also depicted in Figure 4 for emulsions with SMD_i of 3.1 μm . In the case of the emulsions with SMD_i of 3.1 and 20.8 μm , the exponent b takes a

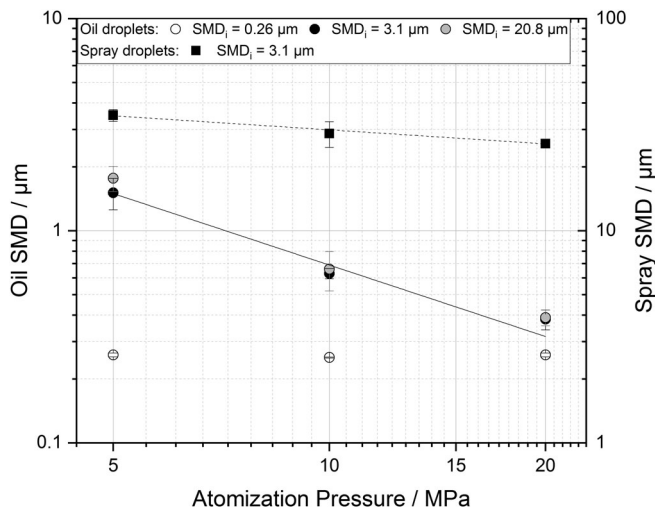


FIGURE 4 Sauter mean diameter of spray and oil droplets after atomization; fit to Equation (4) for spray droplets (dotted line) and oil droplets (solid line) for emulsions with $SMD_i = 3.1 \mu\text{m}$

	Oil droplets			Spray droplets
	SMD _i = 3.1 μm	SMD _i = 20.8 μm	SMD _i = 0.26 μm	
C	119.2	107.6	0.25	82.3
b	1.1	1.1	2E-10	0.2
R ²	0.98	0.96	4E-13	0.99

TABLE 3 Constants C and b as well as coefficients of determination R^2 for oil and spray droplets for the fit using Equation (4)

value of 1.1. This suggests that laminar stresses inside the atomizer dominate the oil droplet breakup. In the case of emulsions with a SMD_i of 0.26 μm, the exponent b takes a value close to zero. In this case, oil droplet breakup does not take place and the SMD is independent of the atomization pressure.

3.2 | Spray droplet size

The resulting SMD of spray droplet size distributions at the different atomization pressures are also depicted in Figure 4 for emulsions with SMD_i of 3.1 μm. No significant difference in the spray SMD was observed for emulsions with different SMD_i at the same pressure (data not shown). A reduction of the spray SMD with increasing atomization pressure is observed. However, it is noticeable that the effect of increasing pressure on the spray droplet size is in relation much lower than on the oil droplet size. In fact, an increase in the atomization pressure from 5 to 20 MPa resulted in a reduction of the oil SMD by 74%, while the SMD of the spray droplets was reduced only by about 26%.

To evaluate the breakup mechanism of the spray droplets, the resulting spray SMDs were also correlated to the expression in Equation (4). This expression has been widely used to correlate the SMD of spray droplets with the atomization pressure in pressure nozzles (Lefebvre & McDonell, 2017; Stähle et al., 2017). In this case, b usually takes values between 0.27 and 0.4 (Lefebvre & McDonell, 2017). The resulting constants C and b for the spray droplets, as well as the coefficient of determination R^2 are listed in Table 3. The resulting fit is depicted in Figure 4. In the case of spray droplets in this study, b takes a value of 0.22, which is an indicative of breakup in turbulent flow (Karbstein, 1994). The results imply that different mechanisms underlie the breakup of oil and spray droplets. From the literature on pressure swirl atomization it is known that spray droplets are generated due to the high relative velocity between the liquid and the gas outside of the atomizer. The liquid leaves the atomizer as a conical sheet and disintegrates into spray droplets by Kelvin-Helmholtz instabilities or by turbulence (Walzel, 2003). In the case of oil droplets, laminar shear stresses inside the atomizer, and specifically in the thin liquid film before the atomizer outlet, are expected to dominate the droplet breakup.

3.3 | Estimation of stresses and capillary numbers

To explain the observed dependences of the oil droplet breakup on the atomization pressure and on the initial oil droplet size, the laminar

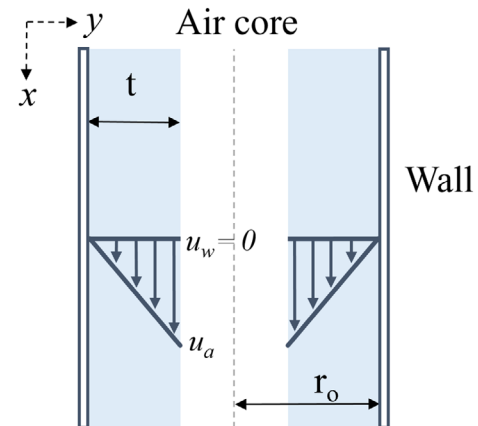


FIGURE 5 Model for the flow profile in the liquid film in the atomizer orifice. r_o : nozzle inner radius; t : liquid film thickness; u_a : velocity at the interface with the air core; u_w : velocity at the wall

stresses in the atomizer and the capillary numbers are estimated. The dominant stresses leading to oil droplet breakup are expected to occur in the thin liquid film at the atomizer outlet. Shear and elongational stresses can also occur in the slots of the slotted core, as well as in the swirl chamber. However, the named stresses are expected to be much lower in comparison to the stresses in the liquid film close to the atomizer exit (Nonnenmacher & Piesche, 2000; Rezaeimoghaddam et al., 2010). At this point, the small thickness of the liquid film leads to very high liquid velocities and velocity gradients.

In the liquid film, shear stresses are expected to dominate. A study of the flow inside similar pressure swirl nozzles confirms this: Renze et al. (2011) showed that the elongational rates in the liquid film at the nozzle outlet had a magnitude of half of the shear rates. It should be noted, however, that the critical capillary numbers for elongational flow are much lower than in shear flow (Grace, 1982). Therefore, oil droplet breakup in elongational flow requires reduced stresses compared to shear flow.

A schematic drawing for the liquid film in the orifice of the pressure swirl nozzle (as in Figure 1) is depicted in Figure 5. A model of the flow profile for the estimation of the shear rate in the liquid film is also depicted. A simplified linear flow profile is assumed, in which the liquid velocity at the wall u_w is zero and the maximum velocity u_a occurs at the air-liquid interface. In reality, the maximum liquid velocity occurs probably somewhere before the interface, as the liquid is slowed down by the air. This discrepancy is however not expected to change the rough magnitude of the shear rate estimation. Another important assumption is, that due to the high magnitude of the axial

velocity, the radial velocity in the liquid film can be neglected (Rezaeimoghaddam et al., 2010).

The definition of the shear rate $\dot{\gamma}$ is presented in Equation (5), while the solution for our model system is presented in Equations (6) and (7). In these equations u is the axial velocity and y is the coordinate perpendicular to the flow direction. t corresponds to the thickness of the liquid film. To solve Equation (6), the following boundary conditions are applied: the velocity at the wall u_w is equal to zero, while the velocity at the interface with the air core u_a is twice the average velocity \bar{u} of the liquid.

$$\dot{\gamma} = \frac{du}{dy} \quad (5)$$

$$\int_0^t d\dot{\gamma} = \int_{u_w}^{u_a} du \quad (6)$$

$$\dot{\gamma} = \frac{u_a - u_w}{t} = \frac{2\bar{u}}{t} \quad (7)$$

The average velocity \bar{u} is calculated according to Equation (8), in which Q_L corresponds to the experimentally measured volume flow and A_L is the flow area of the liquid. The flow area is calculated from the area of the nozzle orifice minus the area of the air core (Equation (9)), in which r_o corresponds to the radius of the nozzle orifice.

$$\bar{u} = \frac{Q_L}{A_L} \quad (8)$$

$$A_L = \pi(r_o^2 - (r_o - t)^2) \quad (9)$$

Solving Equation (9) requires knowledge of the liquid film thickness inside the atomizer. Equation (1) has been used to estimate the liquid film thickness in this study with a constant C value of 2.7. Unfortunately, no study has been found in literature, which validates the use of this correlation in the pressure range of this study. However, the correlation is based on a theoretical analysis of the flow conditions in the atomizer, which are expected to be valid at high pressures too.

The estimated film thickness, mean velocity, and shear rate in dependence of the atomization pressure are summarized in Table 4. As expected, the film thickness decreases and the velocity of the liquid increases with increasing pressure. By this, an increase in the

calculated shear rate with increasing atomization pressure is observed. The relatively high values of velocities are expected for atomizers with small orifices, as in this study (Wimmer & Brenn, 2013). The obtained values of shear rate are in good agreement with reported values in the literature for similar atomizers. For example, by means of numerical simulations, the shear rate in the orifice of a pressure swirl nozzle with $d_o = 1.8$ mm was estimated to be in the order of 10^5 s^{-1} for a pressure of 0.2 MPa (Renze et al., 2011). Due to the small diameter (0.34 mm) and higher pressures used in this study (up to 20 MPa), values up to 10^6 s^{-1} were obtained for the estimated shear rate. The increasing shear rate explains the increased oil droplet breakup with increasing atomization pressure.

The corresponding capillary numbers for shear flow for the different initial oil droplet sizes and atomization pressures were calculated by means of Equation (2) and are summarized in Table 5. These values are compared to the critical capillary number from Grace (1982) for droplet breakup in shear flow, also shown in Table 5. The estimated capillary numbers for the emulsions with a SMD of 3.1 and 20.8 μm are, for all atomization pressures, well above the critical capillary number. Therefore, oil droplet breakup in shear flow is possible for these emulsions at the studied atomization conditions. In the case of emulsions with a SMD of 0.26 μm , the capillary numbers are below the limits of droplet breakup in shear flow calculated by Grace (1982). In the case of the smaller oil droplets, the shear stresses during atomization are not high enough to overcome the capillary pressure and no oil droplet breakup is possible.

According to the work of Renze et al. (2011), the elongational stresses in the liquid film at the outlet of the atomizer are estimated to have a magnitude of half of the shear stresses. Based on this information and on the estimated shear rates listed in Table 4, the capillary numbers for elongational flow were also estimated and are listed in Table 5. Similar to the previous analysis on shear flow, the capillary numbers are compared to the critical capillary number from Grace (1982) for elongational flow. From Table 5 it can be seen, that for emulsions with a SMD_i of 3.1 and 20.8 μm the estimated capillary numbers are well above the critical capillary number for all atomization pressures. Therefore, in spite of the lower magnitude of the elongational stresses, oil droplet breakup due to elongational flow is also possible for these emulsions. The lower values of critical capillary numbers for elongational flow, compared to shear flow (Grace, 1982) explain this. In the case of emulsions with a SMD of 0.26 μm , the capillary numbers for a pressure of 10 and 20 MPa are slightly larger than the critical capillary number for elongational flow. Nonetheless, no droplet breakup was observed during atomization under these

TABLE 4 Estimated liquid film thickness, mean liquid velocity, and shear rate at different atomization pressures

Atomization pressure p_L MPa	Film thickness t^a μm	Mean liquid velocity \bar{u} m/s	Estimated shear rate $\dot{\gamma}$ 1/s
5	160	63.1	7.9E+05
10	147	89.7	1.2E+06
20	127	106.6	1.7E+06

^aAfter (Suyari et al., 1986).

TABLE 5 Estimated capillary numbers and critical capillary number after (Grace, 1982) for shear and elongational flows

Atomization pressure p_L (MPa)	Ca for shear flow			Ca for elongational flow			Ca_{crit} for shear flow ^a (–)	Ca_{crit} for elongational flow ^a (–)
	SMD _i			SMD _i				
	3.1 μm	20.8 μm	0.26 μm	3.1 μm	20.8 μm	0.26 μm		
(–)	(–)	(–)	(–)	(–)	(–)			
5	3.1	21.0	0.23	1.6	10.5	0.11	0.5	0.15
10	4.9	32.6	0.33	2.5	16.3	0.16		
20	6.7	44.7	0.45	3.4	22.3	0.22		

^aAfter (Grace, 1982).

conditions. These results may be explained by several reasons. First of all, the critical capillary numbers from Grace (1982) were defined for quasi-equilibrium conditions under well-defined flow conditions. Neither wall effects, nor very quickly changing flow conditions at time scales well below the critical deformation time were considered, as should be done for pressure swirl atomizers. Therefore, a larger critical capillary number as the one reported by Grace (1982) can be expected. Similar results are reported in the study of Munoz-Ibanez et al. (2015) for rotary and pneumatic atomizers. Furthermore, a great uncertainty results from the rough estimation of the elongational stresses at about half of the shear stresses (Renze et al., 2011).

Nevertheless, the good agreement of the experimental results with the theory on droplet breakup in laminar flow supports the hypothesis, that the stresses in the liquid film at the atomizer outlet dominate oil droplet breakup during atomization. To confirm the findings in this study, a detailed deduction of the stress profiles inside the atomizer would be required.

To explain the fact that the same oil droplet size after atomization was achieved with emulsions with SMD_i of 3.1 and 20.8 μm , the residence time in the high stress area was estimated. From the emulsion theory it is known that larger droplets require longer stressing times for reaching the deformation state that corresponds to the stress applied (Walstra, 1993). Therefore, to achieve the equilibrium value of oil droplet size, the residence time in high stress areas must be long enough to allow deformation and breakup of the large droplets. By means of Equation (3) the required deformation time for oil droplet breakup can be estimated. For both initial oil droplet sizes it is in the order of $\sim 10^{-6}$ to 10^{-7} s. The mean residence time of the emulsions in the liquid film in the nozzle orifice is estimated to be in the order of $\sim 10^{-5}$ s. Therefore, the residence time should be sufficiently long for both emulsions to reach the equilibrium value of oil droplet size. The fact that the same oil droplet sizes were obtained for emulsions with SMD_i of 3.1 and 20.8 μm is explained as in both cases the residence time was long enough to break up the droplets to the equilibrium value. It should be noted that Equation (3) was derived for stationary shear flow conditions, which probably do not correspond to the flow conditions in the atomizer. For a detailed discussion of these results, stress-times

profiles inside the atomizer would have to be evaluated from local flow conditions.

4 | CONCLUSION

The experimental study of atomization of O/W emulsions with pressure swirl atomizers showed that a significant oil droplet breakup takes place during atomization. The oil droplet breakup is highly dependent on the atomization pressure, as the stresses in the liquid film of the atomizer orifice correlate with the atomization pressure. The impact of the pressure on the spray droplet size is relatively low compared with the impact on the oil droplet size. These results have the practical implication that an increase in the atomization pressure to achieve an adequate spray droplet size for the spray drying process, will necessarily lead to a reduction of a previously adjusted oil droplet size in emulsions. The results also suggest that oil droplet breakup occurs under laminar flow conditions, whereas spray droplet breakup is dominated by turbulent flow. In addition, the results suggest that the stresses in the atomizer and the residence time of the droplets are large enough to reduce the SMD to submicron values, even when emulsions with large initial oil droplet sizes are atomized. Therefore, the oil droplet size after atomization can only be controlled to a limited extent with the initial oil droplet size.

A theoretical approach for the estimation of stresses and capillary numbers during atomization with pressure swirl atomizers was developed. The good agreement of the experimental results with the theory on droplet breakup in laminar flow supports the hypothesis, that the stresses in the liquid film at the atomizer outlet dominate oil droplet breakup during atomization. The results also indicate that both shear and elongational stresses can lead to oil droplet breakup under the studied conditions. To confirm this, detailed stress-time profiles in the atomizer should be analyzed.

The concept developed in this study for the estimation of the capillary numbers can be used to control oil droplet breakup during atomization under given process conditions. The findings suggest that to avoid oil droplet breakup, the emulsion properties and operating conditions must be adjusted to obtain capillary numbers below the critical values. This concept could be used as a tool to control oil

droplet size during atomization in practical applications. These results are of high relevance for spray drying applications in which the physical stability and sensorial properties are affected by the oil droplet size. To further validate this, more investigations are required covering a wider range of viscosity ratios.

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AUTHOR CONTRIBUTIONS

Martha L. Taboada: Conceptualization; data curation; investigation; methodology; project administration; validation; visualization; writing-original draft. **Ann-Christin Schäfer:** Investigation; methodology; writing-review and editing. **Heike P. Karbstein:** Funding acquisition; supervision; validation; writing-review and editing. **Volker Gaukel:** Funding acquisition; project administration; supervision; validation; writing-review and editing.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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