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## Impact of effervescent atomization on oil drop size distribution of atomized oil-in-water emulsions

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### Abstract

In this work the application of effervescent atomization to spray drying of food liquids like emulsions is explored. Therefore the influence of the atomization process on the breakup of oil drops inside the emulsion is investigated. It is expected that the oil drop size distribution of the emulsion is influenced by the stress inside the nozzle orifice and the following atomization. According to Grace the viscosity ratio between disperse and continuous phase is a crucial factor for drop breakup. A model oil-in-water emulsion was used. The viscosity of the continuous phase was adjusted by adding maltodextrin or gelatinized corn starch thus varying the viscosity ratio in the range between 15 and 0.1. The dry matter content and corresponding viscosity show only low influence on the spray drop size distribution. However, the atomized emulsions contain mostly smaller oil drops compared to the original emulsions. The influence of the atomization on the oil drop size distribution decrease with decreasing viscosity ratios. An influence of increasing stress due to increased atomization gas mass flow is present but less significant. The viscosity ratio thus allows controlling the influence of the atomization on the oil drop size distribution in the spray. The invariance of the spray drop size distribution on minor changes in fluid properties like viscosity is a favorable characteristic in food processing where such changes are common.

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*Keywords:* effervescent atomization; emulsion; drop size distribution; viscosity ratio

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### 1. Introduction & Theoretical Background

In spray drying, higher dry matter content of the feed is favorable due to decreased energy consumption for water evaporation. However the corresponding increase in liquid viscosity leads typically to an increase in mean spray drop size and possibly to a deterioration of product quality.

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Pneumatic atomizers may overcome this but only with a high atomization gas consumption [1]. The effervescent atomization is a special type of internal mixing pneumatic atomizer being distinct in the formation of a two phase flow prior to the nozzle orifice outlet [2]. A sketch of the atomization process and the atomizer geometry is shown in figure 1. Atomization gas is injected into the liquid in the mixing chamber. In an ideal state a bubbly flow is formed in the mixing chamber which transforms into an annular flow due to contraction to the nozzle orifice. Exiting the nozzle orifice the thin liquid sheath breaks up into liquid filaments due to the expansion of the atomization gas and further into single drops [2]. Due to its lower atomization gas consumption [1] it is a promising method for economical atomization of liquids with high viscosity. The air-liquid mass ratio  $ALR$  of atomization gas mass flow to liquid mass flow characterizes the atomization gas consumption – see equation 1.

$$ALR = \dot{m}_g / \dot{m}_l \quad (1)$$

With: atomization gas mass flow  $\dot{m}_g$ , liquid mass flow  $\dot{m}_l$

The objective of this work is to explore the applicability of effervescent atomization to spray drying of food liquids. Food liquids are often emulsions. Here, the oil drop size distribution is an important characteristic in the spray dried particles and in the liquid after reconstitution. Therefore the influence of the atomization process – the first step in spray drying – on the breakup of oil drops inside the emulsion is investigated. Shear rates inside the nozzle orifice were estimated from prior investigations [3] to be up to  $10^6$  1/s. Therefore it is expected that the oil drop size distribution of the emulsion is influenced by the stress inside the nozzle orifice and the following atomization.

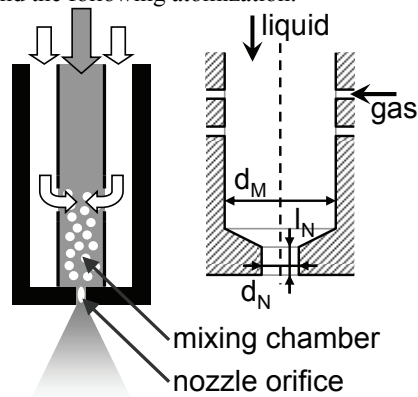


Fig. 1. Sketches of effervescent atomizer. Gas is injected into the mixing chamber filled with liquid and forms two phase flow which exits the atomizer through the nozzle orifice (left). Detail displaying the near orifice geometry with the mixing chamber diameter  $d_M$  and nozzle orifice diameter  $d_N$  and length  $l_N$  (right). [6]

Drop breakup can be described by the Capillary number  $Ca$  – see equation 2. The respective critical Capillary number  $Ca_{cr}$  has to be reached for breakup. Grace [4] demonstrated that the viscosity ratio  $\lambda$  between disperse and continuous phase is a crucial factor for the critical Capillary number. Grace stated also that between a viscosity ratio of 0.5 and 1 a minimum in critical Capillary number occurs and no drop breakup is possible above a viscosity ratio of 4 in laminar shear flow. Armbruster [5] showed that in case of a high concentrated system the viscosity of the emulsion is to be used instead of the viscosity of the continuous phase – see equation 3.

$$Ca = \frac{\eta_c \cdot \dot{\gamma} \cdot x}{2 \cdot \gamma} \quad (2)$$

With: viscosity of continuous phase  $\eta_c$ , shear rate  $\dot{\gamma}$ , drop size  $x$ , interfacial tension  $\gamma$

$$\lambda = \eta_d / \eta_e \quad (3)$$

With: viscosity of disperse phase  $\eta_d$ , viscosity of emulsion  $\eta_e$

## 2. Materials & Methods

A model oil-in-water emulsion was used composed of demineralized water, vegetable oil (Floreale Haagen GmbH) and whey protein Lacprodan 87 (Arla Foods GmbH) as emulsifier. The disperse phase fraction was kept constant at 20 wt.-% and the emulsifier concentration at 2 wt.-%. The viscosity of the continuous phase was adjusted by adding maltodextrin (MD, Cargill C\*Dry MD 01910) or gelatinized corn starch (CS, Cargill C\*Tex 03402) at concentrations from 30 to 50 wt.-% and 2 to 5.4 wt.-% respectively. This allowed for the adjustment of viscosity ratios in the range between 15 and 0.1.

A colloid mill (FrymaKoruma GmbH) was used for emulsification. Vegetable oil and emulsifier were processed with 18 wt.-% (total) water at a gap width of 0.3 mm and a rotor speed of 2885 min<sup>-1</sup>. The carbohydrates and the respective remaining amount of water were mixed separately to keep the viscosity constant during the formation of the initial emulsion. The corn starch suspension it was gelatinized at 75 °C for 60 min. Finally, the initial emulsion and the carbohydrate solution / suspension were mixed gently to avoid a change in oil drop size distribution.

An effervescent atomizer was used with a mixing chamber diameter  $d_M$  of 6 mm, a nozzle orifice length  $l_N$  and diameter  $d_N$  of 1.5 mm – see figure 1 – according to prior investigations [6]. Samples of the emulsions were taken prior to atomization (untreated) and from the spray at two different *ALR*. Laser diffraction spectroscopy was used for determination of the spray drop size distribution (Malvern Spraytec) and oil size distribution (Beckman Coulter LS 13320) of the emulsions. Due to strong influences of beam steering effects in the spray measurements the scattering data of the two detectors nearest to the laser axis were not considered for the analysis as proposed by Mescher and Walzel [7]. The scattering data were analyzed according to Fraunhofer theory without consideration of the refractive indexes. Sauter mean diameter  $x_{1,2}$  was used as characteristic mean value. In case of the emulsion measurements the scattering data were analyzed according to Mie theory with a standard optical model for vegetable oil in water (refractive indexes: water 1.332, oil 1.473). Spray and oil drop size distributions were measured in triplicate from separate experiments.

Viscosities were measured by rotational rheometry (Anton Paar Physica MCR 301) with increasing shear rate from 0.1 to 1000 1/s at 25 °C. For emulsion containing maltodextrin a cone-plate geometry with a diameter of 50.0 mm (CP-50-1/TG) and for emulsions containing corn starch a coaxial cylinder geometry with an inner diameter of 26.7 mm (CC27) were used. Viscosities were measured in duplicate from separate experiments.

## 3. Results & Discussion

The results of viscosity measurements of emulsions with different carbohydrate concentrations are shown in figure 2. At the respective lowest concentration (30 % maltodextrin and 2.0 % corn starch) a Newtonian behavior is observed whereas at higher concentrations a shear thinning behavior occurs. The slight viscosity increase for 2.0 % corn starch above 400 1/s is likely to be due to Taylor instabilities. The vegetable oil itself has Newtonian behavior and a dynamic viscosity of 0.055 Pa·s (data not shown). The

viscosity ratio  $\lambda$  is calculated according to Armbruster from the measured viscosities at 1000 1/s and shown in table 1.

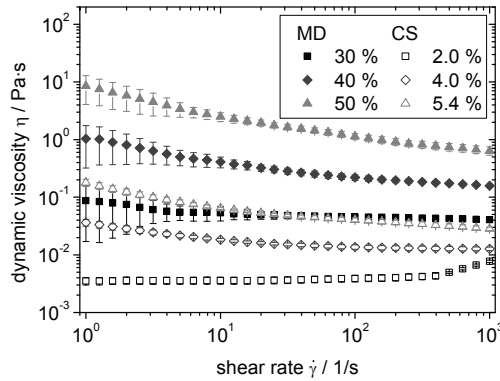


Fig. 2. Viscosity curves of emulsions for different carbohydrate content measured at 25 °C.

Table 1. Dynamic viscosities and viscosity ratios of disperse to continuous phase at 1000 1/s for different carbohydrate concentrations (left maltodextrin, right corn starch). Interpolated value of corn starch 2.0 %.

maltodextrin concentration	dynamic viscosity $\eta_e$ / Pa·s	viscosity ratio $\lambda$ / -	corn starch concentration	dynamic viscosity $\eta_e$ / Pa·s	viscosity ratio $\lambda$ / -
30 %	0.041	1.35	2.0 %	0.004	13.41
40 %	0.158	0.35	4.0 %	0.013	4.32
50 %	0.638	0.09	5.4 %	0.028	1.95

The corresponding spray drop size distributions as a function of the *ALR*, carbohydrate type and concentration are given in figure 3. With increasing *ALR* the Sauter mean diameter decreases. It increases with increasing carbohydrate content. The latter is more pronounced in case of emulsions containing maltodextrin due to the larger differences in the viscosity at different concentrations. Like in prior investigations [3,6] the dry matter content and the corresponding viscosity show in general only a low influence on drop size distribution of the spray compared to the *ALR*.

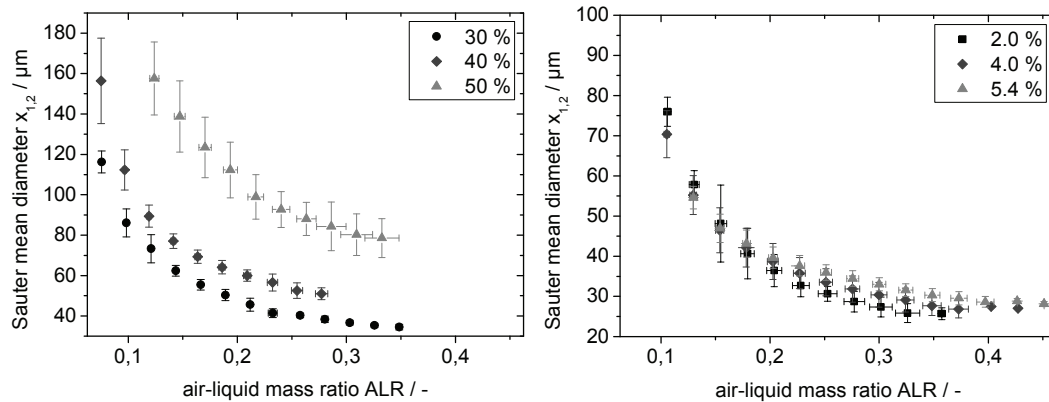


Fig. 3. Sauter mean diameter  $x_{1,2}$  of sprays from maltodextrin (left) and corn starch (right) containing emulsions at different carbohydrate content as a function of the *ALR*.

As the same initial emulsion was used for all emulsions no difference in the oil drop size distribution for the untreated emulsions is expected. The measured oil drop size distributions of the untreated emulsions with different carbohydrate content are shown in figure 4. As accepted the maltodextrin content has no effect on the oil drop size distribution of the emulsion whereas the corn starch content affects the oil drop size distribution. With increasing corn starch content more large drops are detected. For verification, a pure, gelatinized corn starch suspension – containing no oil drops at all – was measured and it depicts for gelatinization times between 5 and 120 min nearly identical size distributions. This indicates some remaining unsolved starch granules in the suspension but can not be explained definitely in this work.

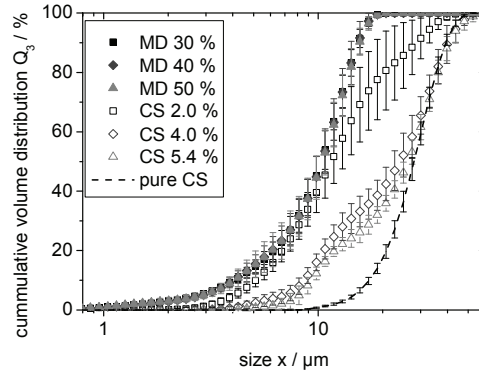


Fig. 4. Size distributions in untreated emulsions prior to atomization (symbols) and pure, gelatinized corn starch suspension (dashed line).

With increasing *ALR* a higher shear rate in atomization is assumed and therefore more drop breakup is expected. In figure 5 (left) the oil drop size distributions of an emulsion containing 30 % maltodextrin is displayed prior to and after effervescent atomization at two different *ALR* values. In atomization oil drops are disrupted. As expected with increasing *ALR* the drop sizes decrease further.

As Grace stated an optimum in drop breakup in laminar shear flow is expected for a viscosity ratio between 0.5 and 1. In figure 5 (right) the oil drop size distributions of maltodextrin containing emulsions with viscosity ratios between 0.09 and 1.35 after atomization at constant *ALR* and the averaged oil drop size distribution prior to atomization are shown. With decreasing viscosity ratio drop breakup can be reduced. Based on the viscosity ratio no significant difference between 30 % ( $\lambda = 1.35$ ) and 40 % ( $\lambda = 0.35$ ) is expected, but is found in the experimental results.

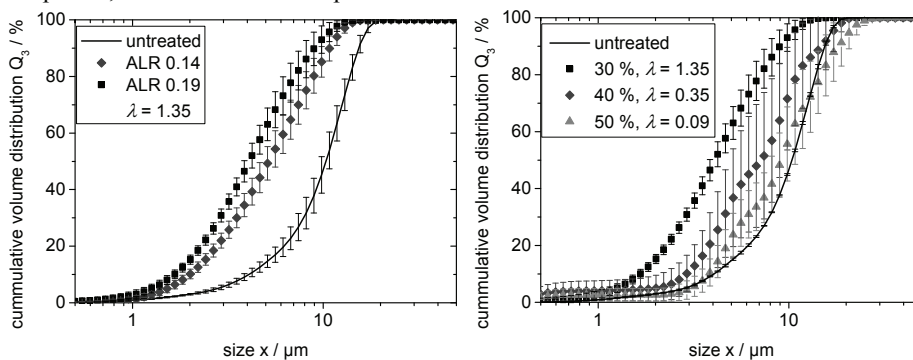


Fig. 5. Oil drop size distributions prior to and after effervescent atomization of emulsions containing 30 % maltodextrin ( $\lambda = 1.35$ ) at varied *ALR* (left) and varied maltodextrin concentrations at *ALR* 0.19 (right)

Due to the high viscosity ratio of emulsions containing 4.0 % ( $\lambda = 4.32$ ) and 2.0 % ( $\lambda = 13.41$ ) corn starch no further drop breakup is expected in effervescent atomization. Only at a corn starch concentration of 5.4 % ( $\lambda = 1.95$ ) a drop breakup is still expected. In figure 6 oil drop size distributions prior to and after atomization are shown for  $\lambda = 1.95$  (left) and  $\lambda = 13.41$  (right). The emulsions with high viscosity ratios depict no further drop breakup as expected by theory. However, an increase in drop size is found which may be due to coalescence of drops. There is no influence of  $ALR$  found. The emulsions with a viscosity ratio of 1.95 depict a slight decrease in drop size after atomization. Due to high standard deviations of the mean values this decrease can not be valued as significant. Overall the corn starch data have to be considered with care since there is an overlay effect of the pure corn starch particles on the measured oil drop size distribution as shown above which can not be eliminated so far. This also obviates the comparison of emulsions with different corn starch content directly as influence of starch content and atomization can not be distinguished.

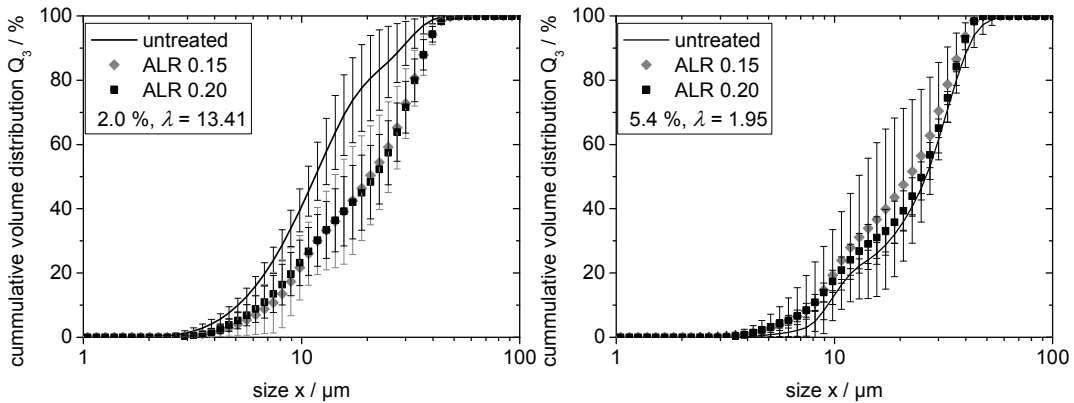


Fig. 6. Oil size distributions prior to and after effervescent atomization of corn starch emulsions at varied  $ALR$ ,  $\lambda = 1.95$  (left) and  $\lambda = 13.41$  (right).

Possible reasons for the slight differences between the model of Grace and the observed trends can be the following. Likely no ideal laminar shear flow is present in the nozzle orifice which is assumed in the model of Grace. The model of Grace assumes also Newtonian behavior of the fluid. Furthermore, the shear rates inside the nozzle orifice are expected to be up to  $10^6$  1/s and viscosities at  $10^3$  1/s are used for calculation of the viscosity ratios. The last could be avoided in future investigations by using measurement data from capillary rheometry.

#### 4. Conclusions & Outlook

The invariance of the spray drop size distribution on minor changes of fluid properties like viscosity is a favorable characteristic in food processing where such changes are common due to natural variations of the raw materials. The  $ALR$  has the largest influence on the resulting spray drop size distribution in effervescent atomization.

It can be stated that the viscosity ratio proves to be an essential parameter for the oil drop breakup in an emulsion being atomized by an effervescent atomizer. This allows to control the influence of the atomization on the oil drop size distribution in the spray. The viscosity ratio values Grace stated as optimal fit good but not exactly to the observed trends but the differences in flow regime and rheological properties from model to experiment have to be considered.

In future work the shear rates in the nozzle orifice will be determined more precisely for further verification of the applicability of Grace's theory on the oil drop breakup in effervescent atomization.

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