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ATOMISATION OF SOLID SUSPENSIONS WITH DISSOLVED INERT GASES

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ABSTRACT
The dry substance content of coffee extract for spray drying is limited to 50 - 55 %, since high water contents allow low liquid viscosities, which are necessary for pumping and subsequently atomising the coffee extract. Carbon dioxide can be dissolved under elevated pressure in liquid coffee and has with high loading the effect to reduce the viscosity of the suspension. By adding and dissolving carbon dioxide, it would be possible to increase the dry substance content by metering carbon dioxide into the pressure line before the atomiser nozzle while maintaining the same viscosity value. Productivity of the plant and product quality could thus be increased. In the present work, liquid properties like surface tension, density and dynamic viscosity of coffee suspensions with and without dissolved carbon dioxide, solubility of carbon dioxide in coffee suspensions as well as the influence of dissolved carbon dioxide on atomisation were investigated.

INTRODUCTION

For spray drying of coffee extracts, the aqueous liquid, which is often quite viscous, may be charged with liquified inert gases before atomisation in order to alter bulk density and product colour [1]. Typically applied liquid gases are carbon dioxide (CO_2) and nitrogen (N_2), which are used in amounts of below 20 g per 1 kg of dry substance contained in the extract [2]. The dense gases are fed into the pressure pipe between feed pump and atomizer nozzle and are dispersed as homogeneously as possible in the fluid flow (see figure 1).
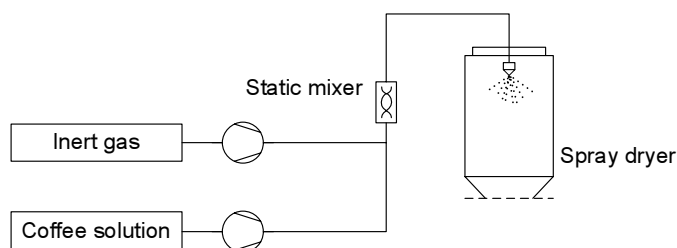

Fig. 1: Spray drying process with dense inert gas

Compared to nitrogen, carbon dioxide is soluble to a high extent in the liquid coffee under elevated pressure conditions before the nozzle. After atomisation, the coffee solution is expanded to ambient pressure in a supersaturated state so that dissolved gas desorbs and small bubbles are formed inside the droplets. Another effect of dissolved carbon dioxide is the viscosity reduction of the liquid, which is an important liquid property for atomisation. Hence it is possible to spray higher concentrated extracts by adding and dissolving carbon

dioxide. However, in order to use the viscosity reduction to full capacity, a certain amount of carbon dioxide, depending on temperature pressure and dry substance content, has to be dissolved until the coffee extract is saturated. Besides, single liquid phase flow with carbon dioxide completely dissolved in the liquid will guarantee a uniform spray. The available time for dissolving carbon dioxide is the residence time of the gas from point of addition to nozzle exit. Nitrogen has a negligible solubility in water and maintains in a gaseous state throughout the whole process. This results in two-phase flow, which causes a significantly higher pressure drop than a single-phase flow [3].

Due to bubbles that are formed before, during or after atomisation inside of the droplets the process is referred to as *foam-spray drying* in literature. Introduced 1922 by Heath and Washburn [4] for the production of milk powder, it was 1954 applied to instant coffee by Chase et al. [5] for the first time. Since then, the process has been used to instantize whole milk [6,7], skim milk [8] and cheese [9].

Until now, investigations on foam-spray drying have been experimentally limited to drying of single suspended drops, falling drops [10-12] and examination of morphology and physical data of spray-dried particles [6,13]. Besides, the viscosity reducing effect of carbon dioxide was not taken into account before and processes were limited to low pressure conditions.

The process investigated here is an atomisation process in which carbon dioxide is added to coffee suspensions under elevated pressures of up to 30 MPa. In contrast to former work on disintegration in pressurized gases [14,15], the spray will be injected into air at atmospheric conditions. The results presented here will cover liquid properties of coffee suspensions with and without dissolved carbon dioxide, the

dissolving behaviour of soluble inert gases in liquid coffee as well as the impact of inert gases on atomisation of coffee suspensions.

LIQUID PROPERTIES OF COFFEE EXTRACTS

Samples of liquid coffee, which were used for the following measurements, were prepared by redissolving spray dried coffee powder in warm distilled water. Commercially available instant coffee of arabica coffee beans was used for all experiments. The moisture content of the instant powder before preparing the solution was 5 – 6 %. The real dry substance content of every sample was determined by evaporating the water in a drying chamber, where the samples were left for 24 hours at 378 K. Food-grade carbon dioxide (99.95 %), utilized as inert gas, was provided by *Yara* and *Linde*.

Interfacial Tension

Interfacial tension is the work done in order to form a new interfacial area. It is thus an important liquid property for the atomisation process as well as for gas-liquid flow where interfaces are formed.

For spraying into the surrounding, the surface tension (interfacial tension against air) of a liquid at ambient pressure is a relevant quantity. Therefore, measurements of the surface tension of liquid coffee were accomplished by means of a tensiometer (provided by *Kruess*) at 293 K and dry substance contents between 20 and 60 %. The measuring principle of the tensiometer followed the *Du Noüy ring* method [16]. The results are shown in fig. 2 in combination with data from Jaeger [17] and Marckmann [18], who both used the pendant drop method for their measurments [19].
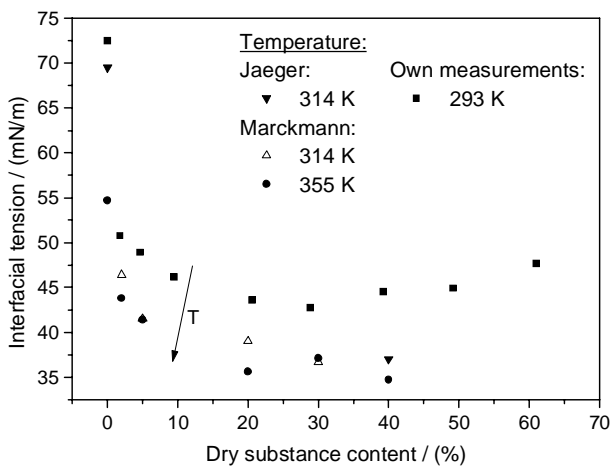

Fig. 2: Interfacial tension of liquid coffee at ambient pressure

As expected, the interfacial tension is lowered with rising temperature. With an increasing content of dry substance, first a decline of the surface tension between 0 and 10 % dry substance content is recognizable.

For gas-liquid flow in the pressure line, the interfacial tension against CO_2 under elevated pressure is relevant. Jaeger [17] and Marckmann [18] observed a reduction in interfacial tension to almost ¼ of the initial value with a pressure rise from 0 to 20 MPa.

Density

The density of liquid coffee was determined with a pycnometer at ambient pressure. Under high pressure conditions with dissolved carbon dioxide, liquid density increase slightly with increasing pressure [18,20]. However, the effect is so low that the densities at ambient conditions are sufficient for this work.

Fig. 3 shows the density of liquid coffee with different dry substance contents over temperature. The results are combined with data from Marckmann [18] who carried out measurements with coffee solution having dry substance contents of up to 40 % according to the same method. From the diagram, it can be seen that density decreases linearly with temperature. The gradient of the plot is independent from the dry substance content. The lines are hence shifted parallely to higher density data with increasing dry substance content.
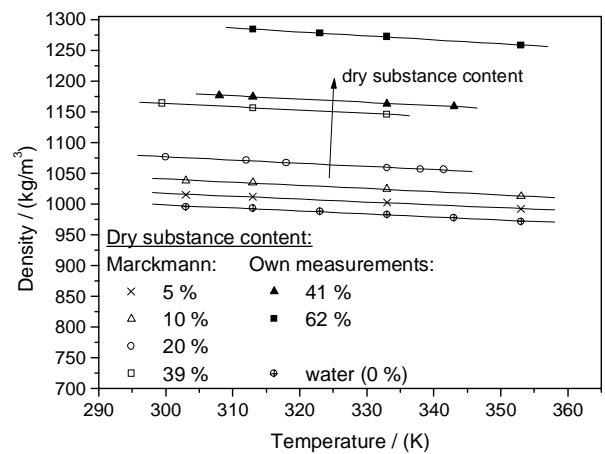

Fig. 3: Density of liquid coffee at ambient pressure

Viscosity

The dynamic viscosity of liquid coffee and liquid coffee saturated with carbon dioxide under elevated pressure was measured with a capillary viscosimeter, which was especially developed for high pressure applications (see figure 4).

The designed capillary viscosimeter possesses a feed container, in which the liquid coffee can be exposed to carbon dioxide. Therefore, carbon dioxide is pumped by means of a double piston pump (type *MSF 111* manufactured by *Maximator*) into the feed container after filling it with liquid coffee. The time needed for establishing equilibrium depends primarily on temperature and dry substance content of the coffee solution. In order to accelerate the mass transfer from the gaseous phase into the liquid phase a high pressure circulation pump (manufactured by *Micropump*) is attached to the feed container. The circulation pump sucks carbon dioxide from the upper part of the container and reinjects it at the lower part, which leads to an intensive mixing of both phases. The pressure remains constant when the system has reached equilibrium. Afterwards, excess carbon dioxide is carefully discharged from the system in order to prevent two-phase flow in the capillary, since that would lead to false results.

A constant flow is maintained by means of a high pressure syringe pump (type *260 D* manufactured by *Isco*), which contains a precise flow control. The pressure loss along the capillary is measured with a differential pressure sensor (series *LD301* manufactured by *Smar*). The pressure of the system is kept constant by a back pressure regulator (series *44-1100* manufactured by *Tescom*). Since the working medium of the high pressure syringe pump is water, a piston inside the feed container transmits the flow of the water from the syringe pump to the liquid coffee. Although the piston was greased for easier sliding, the measured pressure loss exhibited fluctuations within the range of 10 kPa. The application of a capillary viscosimeter for viscosity measurements is limited to Newtonian liquids. This requirement was checked by choosing different flow rates, since the viscosity has to be independent of the flow rate for a Newtonian liquid.
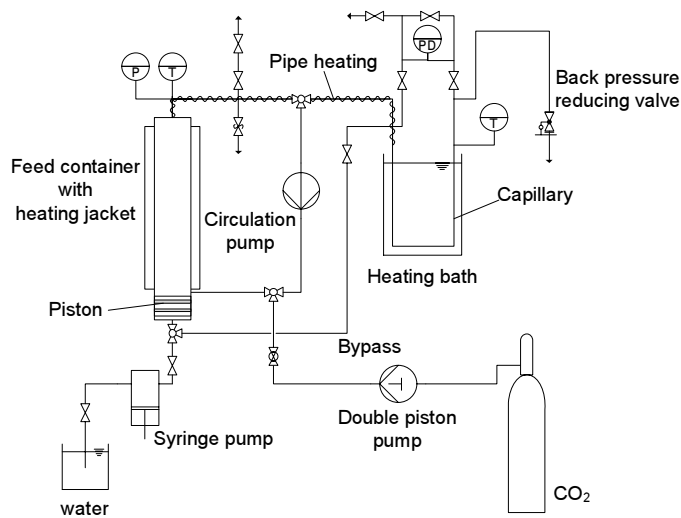

Fig. 4: The capillary viscosimeter

Figure 5, 6 and 7 show the results for the dynamic viscosity of liquid coffee with and without dissolved carbon dioxide. In figure 5 the viscosity is plotted as a function of temperature. As expected, the viscosity decreases with higher temperatures. However, the differences in viscosity for liquid coffee with and without dissolved carbon dioxide become less with higher temperature.
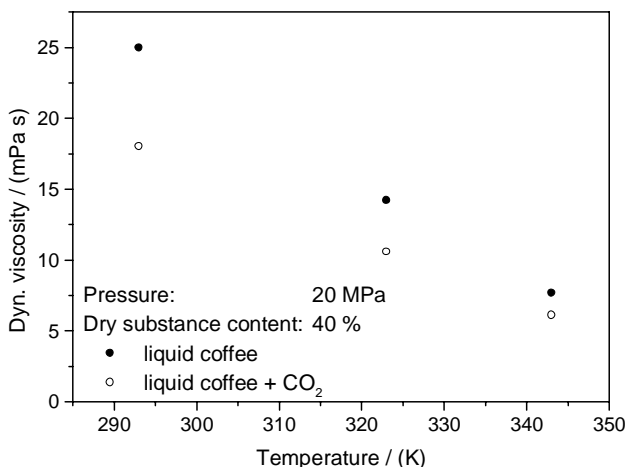

Fig. 5: Dynamic viscosity of liquid coffee against temperature

In figure 6, the dynamic viscosity is plotted against dry substance content of the coffee solution. It shows an exponential increase at higher dry substance contents. The effect of viscosity reduction by saturation with carbon dioxide is intensified at higher dry substance contents. For a dry substance content of 53 % and higher, the viscosity can be reduced by 30 % or more when the coffee solution is saturated with carbon dioxide. On the other hand, for a dry substance content of 20 % there is no effect noticeable. For pure water, even a slight viscosity increase can be observed when carbon dioxide is added [18].
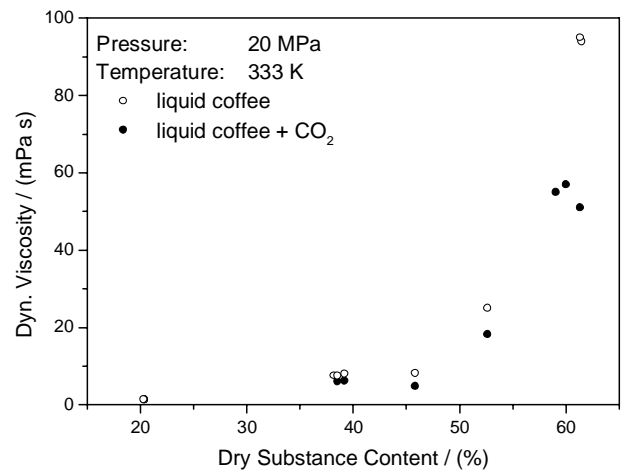

Fig. 6: Viscosity of liquid coffee against dry substance content

Figure 7 illustrates the pressure dependence of the viscosity. It has to be considered that a pressure change alters the carbon dioxide loading, since solubility depends greatly on pressure conditions. An influence of pressure on the viscosity of the saturated solution was expected, since a relation between viscosity and the quantity of dissolved carbon dioxide was assumed. On the contrary, the results presented in figure 7 shows that the viscosity increases with pressure both for liquid coffee and for liquid coffee, which is saturated with carbon dioxide. The content of dissolved carbon dioxide at 3 MPa is only one third of the carbon dioxide content at 25 MPa. However, the quantity of the viscosity reduction is almost the same. That implies that the viscosity could be reduced already at relatively low pressures of 5 MPa with dissolved carbon dioxide.

Solubility

The maximum loading of a liquid with dissolved gas is generally referred to as solubility. It depends on temperature and pressure conditions and – in case of coffee extract – on the content of dry substance. Determination of the solubility of carbon dioxide in the liquid extract is extremely relevant for the industrial application of spraying gas-saturated solutions in order to avoid an excess of carbon dioxide in the liquid flow, which can not be dissolved. A heterogeneous two-phase flow, e.g. stratified flow, can lead to a collapsing spray cone.

The solubility of carbon dioxide in liquid coffee under elevated pressures is measured according to the static method [21]. In order to saturate the liquid phase with carbon
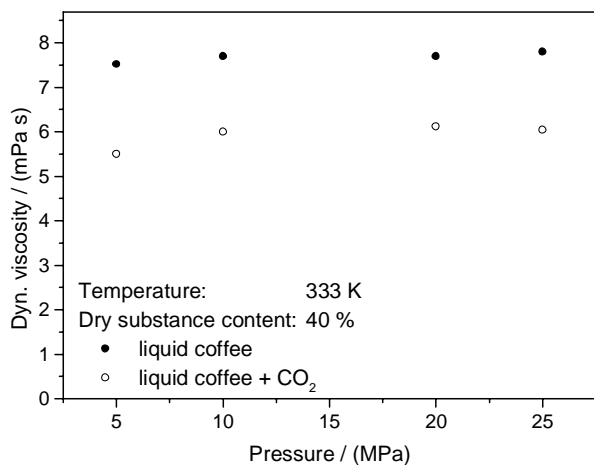
Fig. 7: Viscosity of liquid coffee against pressure

dioxide, a stirrer vessel (manufactured by *Nova Werke AG*) with a maximum operating pressure of 30 MPa is partly filled with coffee solution and afterwards charged with carbon dioxide up to the desired pressure (see figure 8). The vessel is equipped with a magnetically coupled stirrer, in order to intensify the phase contact. The pressure is kept constant by means of a syringe pump (manufactured by *Thar*), which is featured with a pressure controller. The delivered carbon dioxide passes a buffer vessel in order to avoid a disturbance of equilibrium. After reaching phase equilibrium, a sample is sucked off from the liquid phase and caught in a trap to allow the sample to degas under ambient pressure. The mass of the desorbing gas results from the displaced volume of water in a burette.
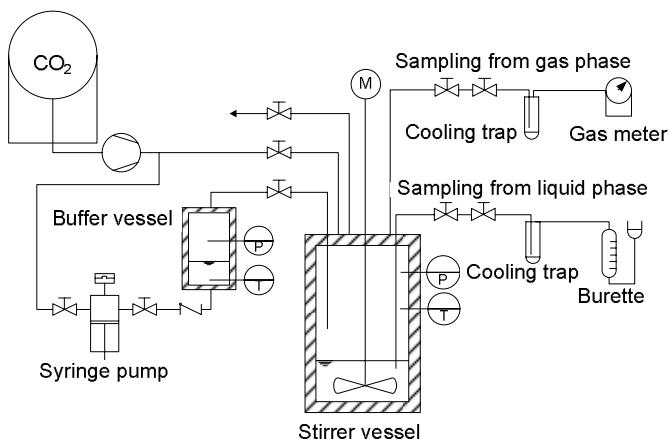

Fig. 8: Equilibration apparatus

The solubility of carbon dioxide in liquid coffee was measured for a dry substance content of 71 % at 353 K and 20 MPa. Fig. 9 gives the solubility as amount of dissolved carbon dioxide per amount of water contained in the sample. It comprises data for pure water [22], data for liquid coffee from Marckmann [18] as well as results from own measurements.

For a dry substance content of 70 %, the solubility is about 15 % increased compared to pure water. In contrast to that, the values of Marckmann show a tendency to lower solubilities with higher dry substance contents. These contradictory gradients can be explained by our own observation that the release of carbon dioxide from the liquid phase sample happens to be slowed down strongly in the

presence of dry substance. For own measurements with 70 % dry substance content the sample was slightly warmed up, in order to achieve completely degassing of carbon dioxide.

Previously, it was assumed that the solubility is independent of the dry substance content [18]. However, the results prove that the dry substance promotes the dissolution of carbon dioxide. It is possible that carbon dioxide adsorbs to undissolved sediments, whose content increases with higher dry substance contents. This assumption is also supported by the decelerated release of carbon dioxide from liquid coffee, which was observed during the experiments. The absolute solubility related to the amount of entire liquid phase decreases however with rising dry substance content due to the smaller amounts of water present in the system.
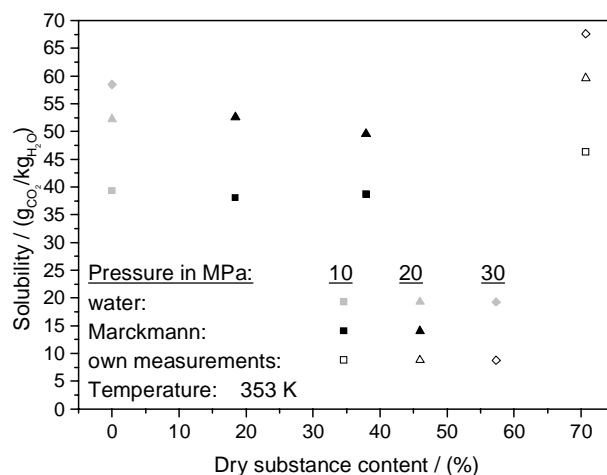

Fig. 9: Solubility of CO_2 in liquid coffee against dry substance content

ATOMISATION OF GAS-SATURATED COFFEE SUSPENSIONS

The droplet size distribution determines the duration and the quality of the drying process in the spray dryer. Knowledge about the influence of carbon dioxide on the droplet size distribution allows therefore conclusions about the uniformity of the drying process.

Experimental setup

For the measurement of the droplet size distribution, a laboratory plant was used for atomisation (see fig. 10). In contrast to the industrial process, the laboratory plant allows a batchwise saturation of the liquid medium with carbon dioxide. Since the plant was constructed by using parts of the capillary viscosimeter, the operation of the plant is similar to that of the viscosimeter. However, instead of allowing the liquid to flow through the capillary, the liquid is atomised by means of a hollow cone swirl nozzle. The nozzle (axial inlet, type *SK* manufactured by *Spraying Systems Co.*) used is typically applied to spray drying of coffee extracts in industry. It consists of an internal swirl body (no. 16) and an orifice insert (no. 80) with an orifice diameter of 0.3 mm. Liquid coffee samples were prepared in the same manner as

for the measurements of the liquid properties. Besides, used materials were identical.
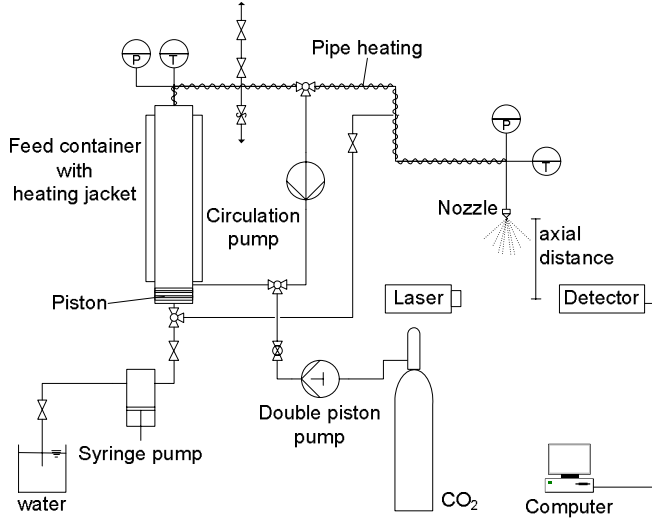

Fig. 10: Laboratory plant for spraying of gas-saturated coffee suspensions

The droplet size distribution was measured with a laser diffraction particle sizer (*Malvern Particle Sizer 2600c*). The chosen specifications were:

Laser:	He-Ne-Laser
Wavelength:	633 nm
Focal length:	300 mm
Measuring length:	30 mm
Number of sweeps:	1000
Correction:	Liquid droplets in air (lds)
Analysis:	Model independent

Using this configuration, droplets in a diameter range of 5.8 – 564 µm could be measured. The results were given as a frequency distribution in mass m_i of droplets within a given diameter range Δd_i, divided by total mass m:

$$Q_3\left(\Delta d_i\right) = \frac{m_i}{m} \quad . \tag{1}$$

From this, an incremental frequency distribution was constructed according to the following equation:

$$q_3\left(\Delta d_i\right) = \frac{\Delta Q_3\left(\Delta d_i\right)}{\Delta d_i} \quad . \tag{2}$$

Results

Atomisation was done at a pressure of 3 MPa and a temperature of 333 K. Both water and coffee suspension (38 % dry substance content) were used as liquids. The volume flow of the liquid was in the range of 151 – 161 ml/min for water and 200 ml/min for liquid coffee. Droplet size distributions were measured near the nozzle in a distance of 20 mm, 40 mm and additionally 80 mm for liquid coffee.

The measured droplet size distributions at an axial distance of 40 mm are given in figure 11 and figure 12. They show that the distribution of liquid coffee is clearly broader

than that from water. This is due to the higher viscosity of liquid coffee on the one hand and contained sediments on the other hand. A higher viscosity leads to bigger droplets, while the drop diameter decreases in the presence of sediments, which cause hole formation in the liquid sheet emanating the orifice and promote the disintegration into droplets.
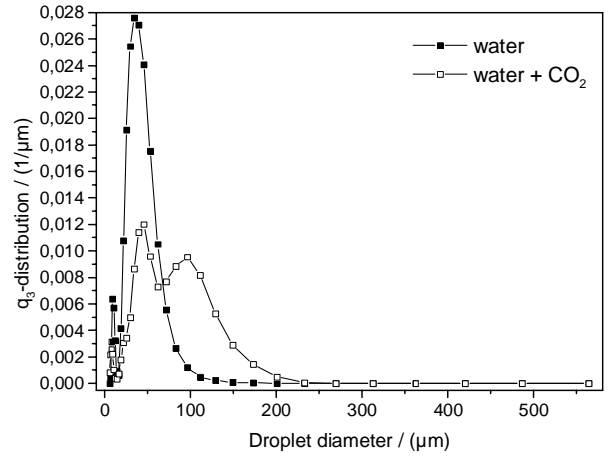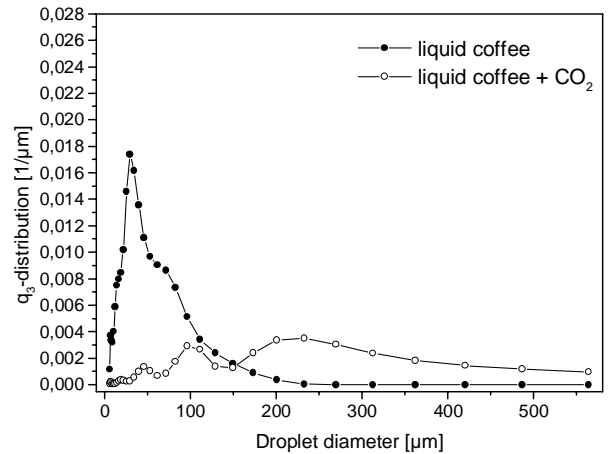

Fig. 11: Droplet size distribution for water

Fig. 12: Droplet size distribution for liquid coffee

For both, water and liquid coffee, it is shown that the droplet size distribution of the spray shifts with carbon dioxide saturation to larger droplet diameters and becomes broader at the same time. Carbon dioxide has a stronger effect on the droplet size distribution of liquid coffee than on that of water. Since the pressure in the droplets is regarded as equal to ambient presure, the liquid with the dissolved gas is present in a supersaturated state. Hence, carbon dioxide is desorbing in the droplets. On the one hand, a certain amount of dissolved carbon dioxide will desorb from the droplets surface. On the other hand bubble nuclei will be formed by carbon dioxide, which is located far from the surface in the middle of the droplets. Subsequently, bubbles will grow inside the droplets and blow them up. Bubble formation and growth as well as desorption from the surface are determined by the transport of carbon dioxide in the fluid phase. Sediments in the coffee suspensions could act as nuclei for bubbles. This could be a reason for a stronger effect of droplet enlargement of coffee solution.

According to product quality, enlargement of droplets is desired for the following reasons:

- Bulk density is lowered by inert gas inclusions.
- The droplet surface, which is the mass transferring surface for the drying process, is increased.

However, the observed broadening of the distribution is undesired, since this would lead to a non-uniform drying.

It is possible, that the formation and growth of bubbles lead to droplet break-up far from the nozzle, especially when higher pressure differences are applied. The droplet break-up by flashing of dissolved gas for aerosol formation was investigated several times, e.g. for fuel sprays [23,24]. However, the droplet break-up in the spray drying process of coffee extract will lead to undesired aroma loss. Future work will thus deal with droplet size measurements far from the nozzle and for higher pressure differences.

ACKNOWLEDGMENT

NOMENCLATURE

Symbol	Quantity	SI Unit
d	Droplet diameter	µm
m	Mass	g
Q_3	Frequency distribution (mass)	--
q_3	Incremental frequency distribution (mass)	1/µm
T	Temperature	K

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