

DISINTEGRATION OF LIQUID PHASES IN PRESSURIZED GASES – INFLUENCE OF MASS TRANSFER

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Abstract

The atomization of liquids in gases at atmospheric pressure is subject of a variety of experimental investigations, empiric and physical models. The atomization of liquid phases in pressurized gases has become increased interest. The aim of the present investigation is the enlightenment of the basic phenomena at the disintegration of liquid jets in pressurized gases up to 35 MPa at temperatures up to 393 K and the measurement of the influence of mass transfer between the liquid and the gas phase on the disintegration process.

Introduction

To elucidate the physical process of atomization and disintegration of liquids jets at high pressures, it is required to characterize the participating forces and to neglect the forces with a reduced influence on the disintegration, in order to minimize the number of variables to describe the problem. Additionally the disintegration is influenced by the geometric shape of the atomizer. The easiest shape of an atomizer is a long capillary tube.

CZERWONATIS et al. [1, 2, 3, 4] found out that the well known correlations and models at atmospheric pressure are not valid for high pressure conditions, since some forces had been neglected that cannot be neglected at high pressure conditions, when the supercritical (gas) phase density increases by a factor of up to 500 and the viscosity increases by a factor of up to 5 compared to atmospheric conditions. But these property data had not been considered in those models and correlations. Considering these property data, dimension analysis led to four dimensionless numbers that describe the ratios of the main participating forces. A new dimensionless number had been developed to describe the different regimes of jet disintegration at high pressure conditions. Those correlations had been developed using non saturated phases which leads to mass transfer during the injection if the participating phases are not inert. Even though mass transfer takes place between the non saturated phases, the thermodynamic property data of the pure compounds had been used since the mass transfer between the non saturated phases was estimated to be negligible. However, the interfacial tension had been measured in equilibrium.

It has not been proved yet that there is any influence of the mass transfer taking place between non saturated phases on the disintegration process. Additionally the influence of surface active additives on the disintegration process has not been investigated yet.

Measurement of thermodynamic properties of mixtures at elevated pressures

For the investigation of the disintegration process, the relevant thermodynamic properties depending on pressure and temperature are required to be known. As liquid phases vegetable oil (non polar phase) and diesel fuel (with or without surface active additives) have been used. As pressurized gas phase either carbon dioxide or nitrogen were chosen. Carbon dioxide is applied in a variety of processes in high pressure technology and nitrogen is as main component of air important in combustion processes. In the investigated range of pressure and temperature carbon dioxide is in the supercritical state ($T_{\text{crit}} = 31.05^\circ\text{C}$; $p_{\text{crit}} = 7.38 \text{ MPa}$). The thermodynamic properties of carbon dioxide as well as of nitrogen are known in a wide range of temperature and pressure and available as datasheet [5].

Vegetable oil - CO_2

Particularly the system consisting of carbon dioxide and vegetable oil show partial miscibility depending on pressure and temperature of the system. The phase equilibrium of this particular system has been taken from [7]. These data and the data of the system carbon dioxide and water taken from [6] for comparison are depicted in figure 1.

Figure 1 shows, that there is a high solubility of carbon dioxide in the liquid oil phase in contrast to the relative low solubility of vegetable oil in the supercritical phase. Caused by the high solubility of carbon dioxide in vege-

table oil up to 35% w/w in the investigated range of pressure, there is a significant change of the density and the viscosity of the liquid phase.

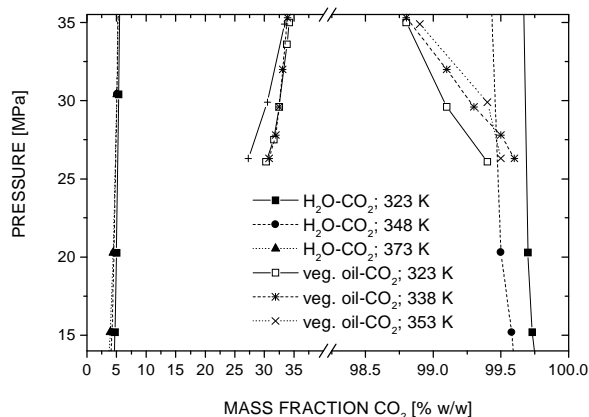


figure 1: Phase equilibrium of CO₂ – vegetable oil and CO₂ – H₂O

The mixture density of the saturated liquid phase, depicted in figure 2a, has been determined by a gravimetric measurement inside a pressure resistant view chamber connected to a high pressure magnetic coupled balance [8].

The viscosity of the saturated vegetable oil has been measured using a high pressure dropping ball viscometer. The saturation of the vegetable oil has been carried out using a gear pump that conveyed the saturated liquid from the saturation autoclave to the viscometer at almost isobaric conditions. The viscosity of the saturated liquid phase depending on pressure and temperature is depicted in figure 2b.

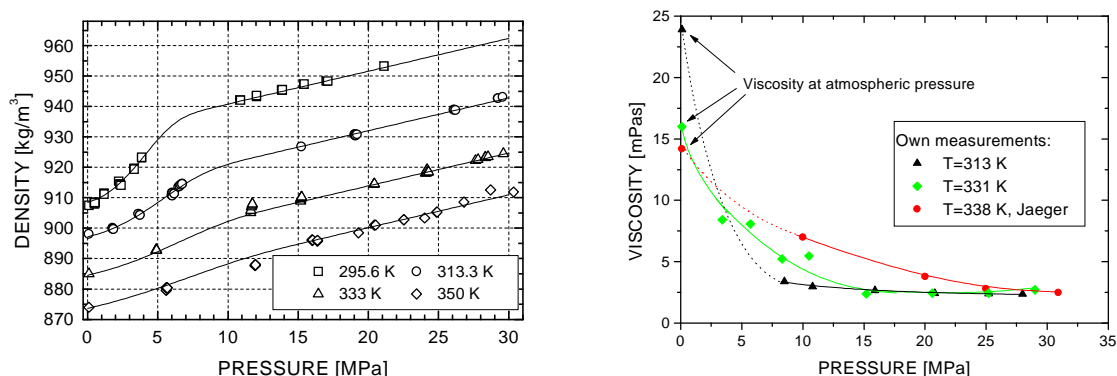


figure 2: Mixture data of the system CO₂ – vegetable oil

The increase of the density of the liquid phase in contact with carbon dioxide (fig. 2a [8]) is significant higher than the pressure induced increase which is linear to the system pressure in the investigated range of pressure[8] even though the density of carbon dioxide in this range of pressure and temperature is lower than the liquid density.

Figure 2b shows that for the investigated temperature range the viscosity is reduced significantly compared to the viscosity at atmospheric pressure. Since the viscosity increases with increasing pressure at absence of carbon dioxide, the carbon dioxide causes the drop of the viscosity. PETER [9] and LOCKEMANN [10] stated that the dissolved carbon dioxide reduces the viscosity, since the small carbon dioxide molecules between the long chains of fatty acids molecules reduce the internal friction of the liquid.

The determination of the interfacial tension at elevated pressures (figure 4) has been carried out using the pendant drop method. Inside a high pressure view chamber a liquid drop hanging at a capillary tube in carbon dioxide was photographed through high pressure resistant sapphire windows. If the ratio of the density between both phases is known the interfacial tension can be determined by numerical evaluation of the contour of the drop [11].

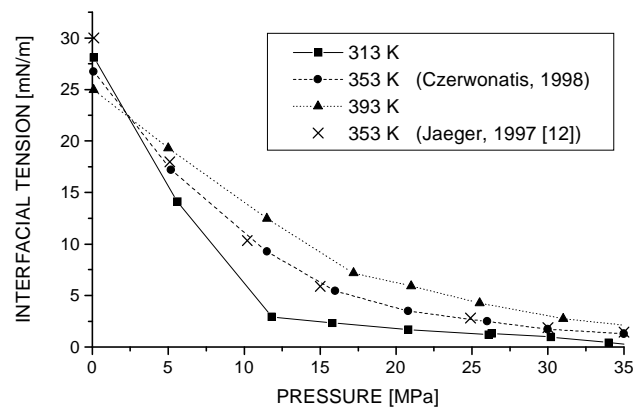


figure 3: pressure and temperature dependence of the interfacial tension of the system vegetable oil-CO₂

The interfacial tension of the system consisting of vegetable oil and carbon dioxide measured in equilibrium declines with increasing pressure. At supercritical conditions the interfacial tension declines with decreasing temperature. A reduced interfacial tension leads to smaller drops since the energy demand to create interfacial area is reduced. Small drops are desired in the most engineering processes because of the improved heat and mass transfer.

Diesel fuel(and additives) - Nitrogen

In contrast to the system consisting of vegetable oil and carbon dioxide the solubility of nitrogen in diesel fuel is negligible. Therefore the thermodynamic property data of the pure substances may be used to characterize the forces involved within the disintegration process. The viscosity of diesel fuel increases slightly with increasing pressure. The system of diesel fuel and a detergent concentration of 1% w/w in contact with nitrogen have similar property data compared to the system without additives except for the interfacial tension depicted in figure 4. The interfacial tension decreases with increasing pressure and temperature. In presence of additives the interfacial tension is generally reduced caused by molecules moving to the interfacial area. The higher the temperature the lower is the reduction of the interfacial tension caused by the presence of detergents.

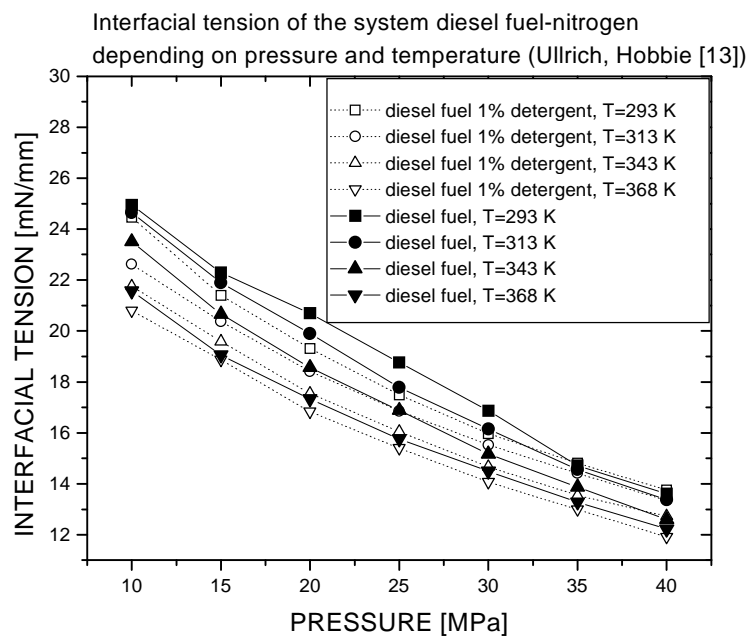
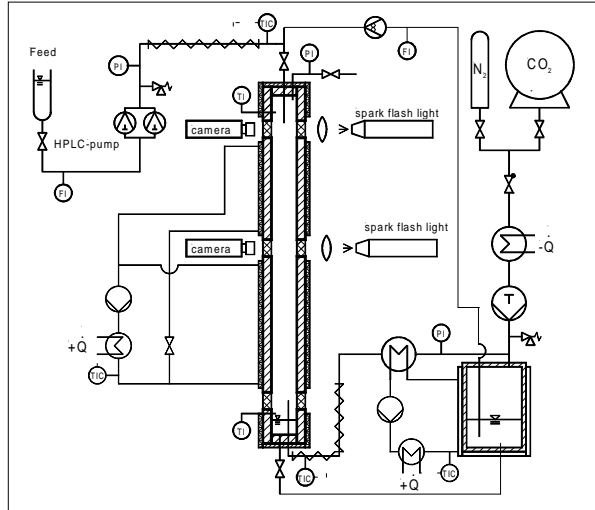


Figure 4: Influence of additives on the surface tension of the system nitrogen and diesel fuel

Experimental Setup



Maximum pressure:	35MPa
Maximum temperature:	393 K
Inner column diameter:	35 mm
Inner column height:	2m
Capillary tubes	0.12, 0.25, 0.5 mm (d/L) < 0.001
pumps:	piston pump HPLC-pump Gear pump
Windows:	Sapphire Windows 18 mm diameter

Figure 5: Experimental set-up for the investigation of the disintegration of liquid jets in pressurized gases up to 35 MPa

The apparatus consisting of a high pressure column with pressure resistant windows for optical observation has been filled with carbon dioxide or nitrogen from the reservoir. The pressure of the gas has been adjusted with a piston pump. The temperature can be adjusted with heat mantles. The conveyance of the liquid is carried out using a proportioning HPLC-pump. If the saturation of the liquid is required, the liquid is conveyed from the saturation vessel to the column at constant pressure within a cycle. After a certain time the equilibrium has been obtained, depending on the mass transfer kinetic. In the high pressure column the liquid jet emerges from the orifice of a capillary tube into the pressurized gas. Capillary tubes with different inner diameters between 0.5 down to 0.12 mm have been used. With the used optical high-speed measurement system it was possible to get sharp images of fast moving disintegrating jets. A sparkflash lamp (NANOLITE) with exposure times down to 18 ns has been used to visualize the phenomena in counterlight technique.

Results

The different kinds and the phenomena of disintegration of liquid jets in gases at elevated pressures are equal to those in gases at atmospheric pressure [4]. With increasing jet velocity three distinct regimes of breakup can be detected: Rayleigh breakup > sinuous wave breakup > atomization.

Depending on the ratio of the inertia force and the viscosity force, which the Reynolds number consists of, laminar jets are being formed at long capillary tube at moderate outlet velocities. The formation of drops from laminar jets is called Rayleigh breakup. To the rotational axis radially symmetric waves are formed by the interaction of primary disturbances in the liquid and surface tension force. Drops are detached by the radially symmetric wave. The oscillation is amplified by the forces caused from the continuous phase [14]. If the outlet velocity is increased, a helical oscillation of the jet surround its own axis will be formed, which is called sinuous wave breakup. The jet disintegrates in short liquid threads which are transformed into spherical drops caused by the surface tension force. A further increase of the outlet velocity leads to atomization of the liquid jet.

Characterization of the disintegration of liquid jets

The complete theoretical description of the disintegration was not possible yet. A method to declare the different regimes of the disintegration at high pressure condition based on two dimensionless numbers had been developed by CZERWONATIS et al. [2]. The new dimensionless number Z^{**} ,

$$Z^{**} = Oh \cdot \sqrt{We_g} \cdot \sqrt{\frac{\eta_l}{\eta_g}} = \frac{\eta_l \cdot v}{\sigma} \cdot \sqrt{\frac{\rho_g}{\rho_l}} \cdot \sqrt{\frac{\eta_l}{\eta_g}} \quad (1)$$

consists of three different dimensionless numbers which represent ratios of the main forces causing the disintegration (Figure 6). Experiments using the system diesel fuel (and additives) – nitrogen have been carried out to confirm this method for other systems. The results are depicted in Figure 7.

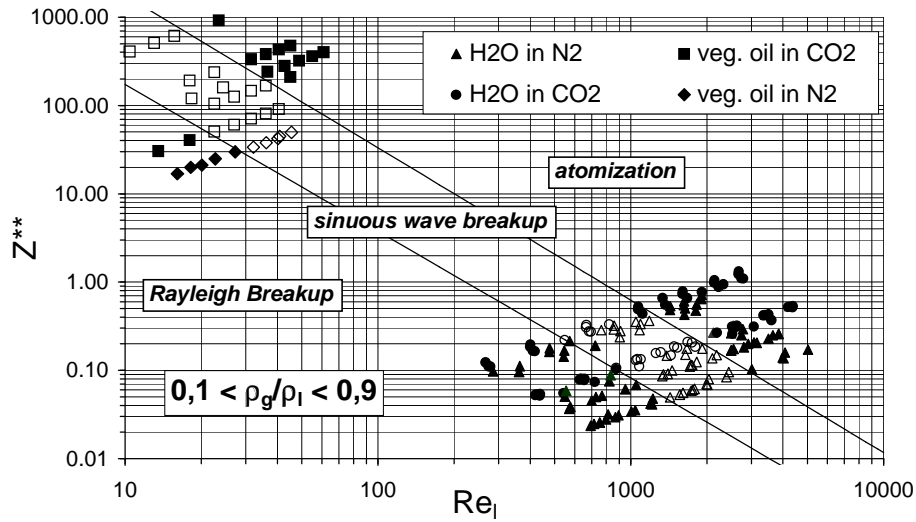


Figure 6: Regimes of jet disintegration at high pressure conditions

According to Figure 7 the measurements using diesel fuel and nitrogen confirmed the method that had been used on other systems before, at least concerning the boundary between the Rayleigh breakup and the sinuous wave breakup regime. Additional measurements are necessary to confirm the boundary between sinuous wave breakup and atomization. It has been confirmed even by a system with surface active detergents that was added to the liquid phase with a certain concentration, although the movement of surface active molecules take a certain time.

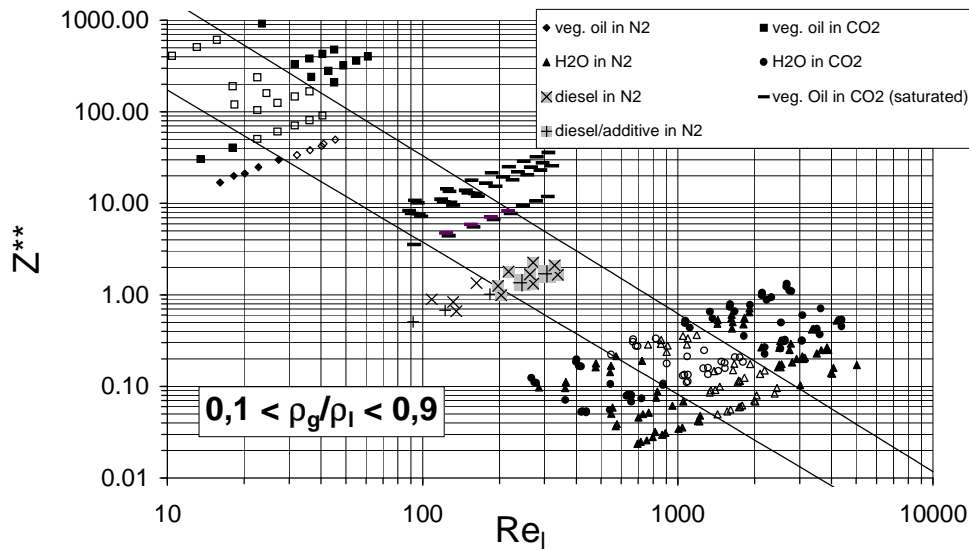


Figure 7: Characterization of the disintegration of different systems

Further on figure 7 shows that also the disintegration of pre-saturated vegetable oil in carbon dioxide can be described by the developed dimensionless numbers. Caused by the saturation of the liquid phase, the Reynolds number increases but the dimensionless number Z^{**} decreases. The boundaries between Rayleigh Breakup and sinuous wave breakup could both be confirmed by these measurements.

Summary

The measurements that have been carried out confirmed the correlation developed by CZWERWONATIS et al.: The saturation of the system vegetable oil and carbon dioxide have not limited the validity of this correlation as well as the presence of surface active additives in the case of the system consisting of diesel fuel and nitrogen. Thus the influence of mass transfer taking place within the measurements of CZERWONATIS et al. related to the used system of vegetable oil and carbon dioxide on the disintegration process is negligible.

Nomenclature

η	dynamic viscosity
ν	kinematic viscosity
ρ	density
v	velocity
d	diameter of the orifice

Subscripts

g	gas
l	liquid

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