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Experimental Investigation of Droplet Injections in the Vicinity of the Critical Point: A comparison of different model approaches

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Abstract
The disintegration process of liquid fuel within combustion chambers is one of the most important parameters for efficient and stable combustion. Especially for high pressures exceeding the critical value of the injected fluids the mixing processes are not fully understood yet. Recently, different theoretical macroscopic models have been introduced to understand breakdown of the classical two phase regime and predict the transition from evaporation to a diffuse-mixing process. In order to gain deeper insight into the physical processes of this transition, a parametric study of free-falling n-pentane droplets in an inert nitrogen atmosphere is presented. Atmospheric conditions varied systematically from sub- to supercritical values with respect to the fluid properties. An overlay of a diffuse lighted image with a shadowgram directly in the optical setup (front lighted shadowgraphy) was applied to simultaneously detect the presence of a material surface of the droplet as well as changes in density gradients in the surrounding atmosphere. The experimental investigation illustrates, that the presence of a material surface cannot be shown by a direct shadowgram. However, reflections and refractions caused by diffuse ambient illumination are able to indicate the presence of a material surface. In case of the supercritical droplet injections in this study, front lighted shadowgraphy clearly revealed the presence of a material surface, even when the pre-heated droplets are released into a supercritical atmosphere. This detection of the droplet interface indicates, that the droplet remains subcritical in the region of interest, even though it is injected into a supercritical atmosphere. Based on the adiabatic mixing assumption recent Raman-scattering results in the wake of the droplet are re-evaluated to compute the temperature distribution. Presented experimental findings as well as the re-evaluation of recent Raman scattering results are compared to thermodynamic models to predict the onset of diffuse-mixing and supercritical disintegration of the droplet. Additionally, a one dimensional evaporation model is used to evaluate the validity of the adiabatic mixing assumption in the estimation of the droplet temperature. The presented findings contribute to the understanding of recent theoretical models for prediction of spray and droplet disintegration and the onset of diffuse-mixing processes.

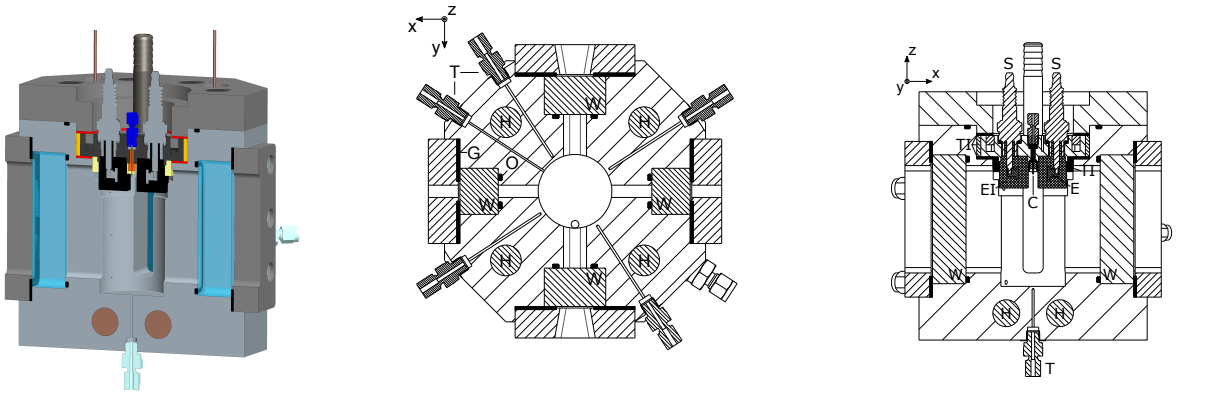
Keywords
Front lighted Shadowgraphy, Raman-scattering, Supercritical fluid injection

Introduction

The disintegration process of liquid droplets in a high pressure and high temperature environment is one of the most important parameters for stable and efficient combustion. Therefore, the behaviour of droplets in such an environment has long been a matter of analytical, numerical and experimental investigation. Especially the transition process from a two phase behaviour of the droplet or jet to a diffusion dominated mixture is not yet well understood. Recently, three different models predicting the onset of this diffuse-mixing have been introduced. The first model by Banuti and Hannemann [1] exploits the supercritical transition at the Widom-line of the fluid and introduces a new breakup model called pseudo-boiling. Another, second criterion, is proposed by Dahms and Oefelein [2] and later extended by Dahms [3]. Dahm's criterion compares the thickness of the interface layer with the characteristic length scale of heat transfer to calculate the probability of a supercritical breakup. Qiu and Reitz [4] proposed a third model, which extends classical thermodynamic conditions for a single component fluid to binary mixtures. The transition from sub- to supercritical fluid behaviour occurs, when the local temperature and pressure exceed the critical values of the mixture. Whereas the pressure of the mixture at the droplets interface is controlled in the experimental setup the temperature has to be estimated. Qiu and Reitz [4] suggested a adiabatic mixing assumption, which takes phase stability and phase separation into account, to calculate the local temperature based on the local concentration. In the following, we will focus on a discussion of the Qiu and Reitz [4] model using two different experimental techniques, namely front lighted shadowgraphy and Raman-scattering. Complementary, the evaporation model of Abramzon and Sirignano [5] is used to evaluate the validity of the adiabatic mixing assumption.

Experimental Setup

The experimental investigations described in this paper were done in two nearly identical heatable pressure chambers. The chambers are both designed for investigation of free falling droplets in near critical environments. In case of phenomenological investigations the droplets are generated in a closed chamber, detached and measured. For statistical investigations the chamber is used as a continuous flow chamber. The gas inside the chamber is a pressurized pure nitrogen atmosphere controlled by either solenoid valves mounted in the fluid supply and exhaust system or with a pneumatic valve at the system exhaust (Badger Meter). The pressure chamber can be operated at pressures up to 60 bar and temperatures up to 553 K. Pressure inside the chamber is measured at the fluid exhaust with a temperature compensated pressure transducer (Keller 35 X-HTC) with an uncertainty of ± 300 mbar. Several T-type thermocouples with an uncertainty rated at ± 1 K are installed in the chamber wall and the core to monitor the steel temperature as well as the temperature of the nitrogen atmosphere, respectively.



(a) 3D cross section of the chamber.

(b) Horizontal cross-section through chamber. G: Graphite gaskets; H: Heating cartridges; O: FFKM-O-Rings T: Type-T thermocouples; W: quartz windows.

(c) Vertical cross-section through chamber and droplet generator. C: Capillary; EI: Electric insulation; H: Heating cartridges; S: Spark plugs; T: Type-T thermocouples; TI: Thermal insulation; W: quartz windows.

Figure 1. Horizontal and vertical cut of the pressure chamber.

The pressure chamber is made out of temperature resistant stainless steel (EN-1.4571). The cylindrical core has a diameter of 48 mm and a height of 85 mm. Optical accessibility is granted through 4 UV-transparent quartz windows which are arranged in a 90° angle to each other. Six heating cartridges, four mounted vertically and two mounted horizontally, are inserted in the chamber body. The cartridges are controlled with type J-thermocouples in the heater cartridges as well as type T-thermocouples in the chamber. In figure 1 vertical and horizontal cross-sections of the chamber are depicted. Droplets are generated by an electric detachment system. The system utilizes the interactions between strong, pulsed electric fields and the fluid of the droplet. A detailed description of the droplet generator can be found in Weckenmann et al. [6] and Oldenhof et al. [7]. A analytical and numerical framework of the detachment process was done by Ouedraogo et al. [8]. For further information about the experimental setup the reader is referred to Bork et al. [9], Weckenmann et al. [6] and Oldenhof et al. [7].

Raman Scattering

The objective of the Stokes-shifted Raman scattering layout is the quantitative investigation of species concentrations in the wake of a free-falling droplet. A detailed description of the Raman scattering setup as well as the calibration and data evaluation can be found in Bork et al. [9]. Based on the concentration measurements a temperature estimation is performed to gain a deeper insight into the thermodynamic state of the fluid-mixture in the droplet wake. As a first approximation, the adiabatic mixing model is used for estimation of the temperature. Dahms and Oefelein [2], and later Dahms [3] used this assumption for their simulation study of supercritical jets. Qiu and Reitz [4] used an extended adiabatic mixing assumption, which includes phase instabilities and phase separation for their study on high pressure fuel injection.

Front Lighted Shadowgraphy

Front lighted shadowgraphy is an optical technique to indicate the thermodynamic state of the droplet and simultaneously visualize the changes in density gradients in its vicinity. It applies a direct overlay of a diffusive or scattered lighted image with a common shadowgram in the optical setup. Due to surface tension of the droplet, a strong density gradient at the interface occurs. Using this steep density gradient, the material surface of the droplet is detectable by refractions and reflections of the incoming light on the droplet surface. Changes in density gradients in the vicinity of the droplet are displayed by direct shadowgraphy in parallel light due to the Laplacian of the refractive index ($\partial^2 n/\partial x^2$) [10].

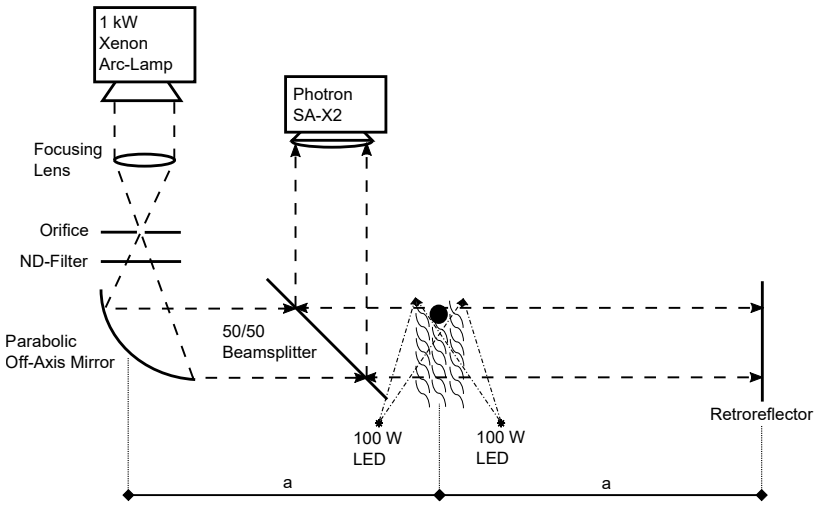


Figure 2. Front lighted shadowgraphy setup.

Figure 2 depicts the front lighted shadowgraphy setup. The retroreflector, which reflects the incoming light with the same incidence angle, on the right side of the chamber overlays a frontal illumination of the droplet with a backlighted shadowgraph. Both images are focused on the capillary and are aligned as well as overlayed by a $50/50$ beam splitter. Note, that optical distortions caused by the reflected shadowgram are not visible. This is due to the fact, that the focal depth of the used long distance microscope is at least one magnitude smaller than the distance between the chamber and the retroreflector. To increase reflection and refraction on the material surface of the droplet two LED arrays are located at the bottom of the pressure chamber illuminating the droplet detachment system. By removing the neutral density filter (ND-filter), located behind the orifice, the intensity of the frontal and back illumination can be adjusted. Without the ND-filter the intensity of the arc lamp overpowers the LED illumination leading to a common shadowgraph.

Experimental results: Characterization of the thermodynamic state during droplet evolution

A parametric study of free falling n-pentane droplets in a sub- to supercritical nitrogen atmosphere has been conducted using front lighted shadowgraphy. Experimental test conditions reach from subcritical chamber conditions to supercritical conditions. The injection temperature of injected fluid remains subcritical to anchor the droplet on the capillary. The reduced chamber temperature and pressure are referred to as $T_{r,ch} = T_{ch}/T_c$ and $p_{r,ch} = p_{ch}/p_c$, respectively. The reduced fluid injection temperature is $T_{r,f} = T_f/T_c$. Reduced values are normalized by the critical values of n-pentane ($p_c = 33.7$ bar, $T_c = 469.7$ K) [11].

(a) $T_{r,ch} = 0.81, T_{r,f} = 0.81$ (b) $T_{r,ch} = 0.95, T_{r,f} = 0.95$ (c) $T_{r,ch} = 1.11, T_{r,f} = 0.95$

Figure 3. Front lighted shadowgraphs at four different positions for supercritical chamber pressures ($p_{r,ch} = 1.78$) and sub- to supercritical temperatures. Left: shadowgraph without ND-filter; Right: front lighted shadowgraph with ND-filter.

The temporal evolution of the droplet under sub- to supercritical ambient temperature and supercritical chamber pressure ($p_{r,ch} = 1.78$) is depicted in figure 3. The right side of each image displays the front lighted shadowgram, whereas on the left hand side the common shadowgraph is depicted. Every image is a compilation of snapshots recorded during the same experiment at four different locations. The shadowgrams and front lighted shadowgrams were recorded directly after each other and care was taken to insure similar chamber conditions during the experiment. As can be seen from figure 3, the mounting of the ND-filter increases the reflections and refractions on the droplet surface by reducing the intensity of the arc lamp. The bright spot at the top of the droplet in the upmost location is a result of reflections caused by the illuminated droplet detachment system, whereas the glare point in the centre of the droplet is a result of the refraction of the laser beam used for triggering. Without ND-filtering the reflection caused by the illuminated droplet detachment system is just slightly visible. This shows the overpowering of the LED arrays by the arc lamp. Changes in density gradients in the vicinity of the droplet are visible in both images.

A visual comparison of reflections and refractions in the front lighted shadowgrams depicted in figure 3 reveals similar behaviour for all conditions. Changes in density gradients are displayed by both common and front lighted shadowgraphy. The material interface of the droplet, however, can only by revealed by reflections and refraction caused by the diffuse illumination in the front lighted shadowgrams. Additionally, presence of a droplet surface is clearly indicated at all conditions, even when injected into supercritical atmospheres. At the transition from sub- to supercritical fluid states the phase boundary of the droplet should be vanishing, due to vanishing surface tension. Weckenmann et al. [12] illustrated, that a diffuse lighted image is sensitive to phase boundaries. Reflections and refractions on the droplet surface would concordantly vanish or show a different optical behaviour. A supercritical droplet, to be precise, a supercritical gas mixture, will therefore show no or different signs of reflective spots or refractive glare points compared to subcritical droplets. By the virtue of that fact, detection of the material surface and similar optical behaviour compared to subcritical temperatures indicates, that the droplet remains subcritical in the field of view although it is injected into a supercritical atmosphere. One possible explanation for this fluid behaviour are cooling effects due to evaporation of the droplet, which mitigate the heat fluxes from the supercritical atmosphere into the droplet. A more detailed explanation will be given in the next section by both experimental and analytical investigations of the droplet wake and the evaporation of the droplet, respectively.

Assessment of thermodynamic models

In this section experimental investigations regarding the thermodynamic state of the droplet are affiliated with thermodynamic analysis of the interfacial temperature as well as the thermodynamic state of the droplet. Our thermodynamic model is based on the true adiabatic mixing temperature calculation by Qiu and Reitz [4], which takes phase stability into account and uses the PC-SAFT equation of state (EOS). The PC-SAFT EOS is formulated as a fundamental equation in the Helmholtz energy utilizing results from statistical physics [13]. PC-SAFT uses chains of tangentially bonded segments of hard spheres as molecular model coarse graining the real molecular structure considering hard-sphere repulsion, chain formation, van der Waals attraction association and polar interactions. One-fluid treatment allows easy transfer from pure components to mixtures. Consideration of the molecular structure allows for good qualitative and quantitative representation of thermodynamic properties with a minimum number of parameters. Since Qiu and Reitz [4] used the Peng-Robinson EOS, figure 4 shows a direct comparison of the theoretical adiabatic mixing temperature calculated by Qiu and Reitz and the PC-SAFT EOS. The fluid system consist of a dodecane-nitrogen mixture at 60 bar and 900 K ambient pressure and temperature, respectively. The fluid temperature is 363 K. As can been seen in figure 4 both calculation coincide very well.

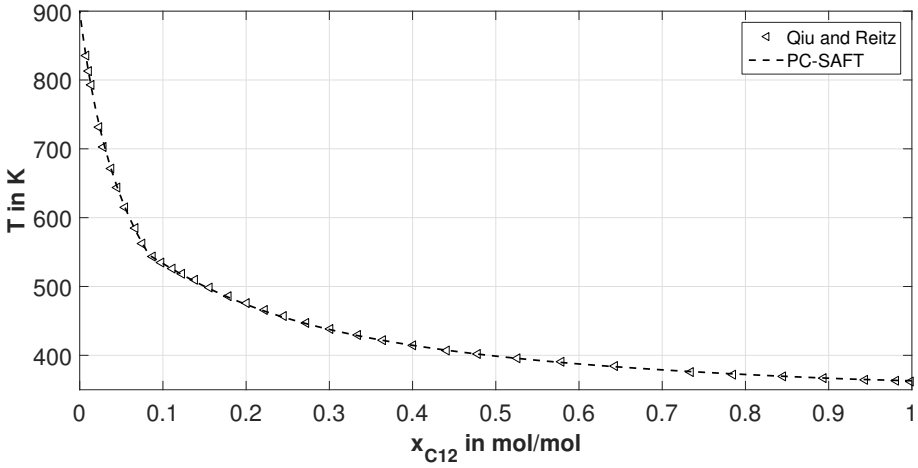


Figure 4. Comparison of the theoretical adiabatic mixing temperature calculations for a n-dodecane-nitrogen (C12-N_2) mixture.

Characterization of the droplet wake

A comprehensive Raman test campaign was conducted to analyse the concentration field in the droplet wake for a variety of test fluids injected into a nitrogen atmosphere. In this paper a re-evaluation of the results for the n-heptane/nitrogen binary mixture done by Bork et. al [9] is discussed. A similar re-evaluation of the experimental results using the thermodynamic model of Dahms [3] is currently in development but not yet published. In this paper we will focus on the thermodynamic model presented by Qiu and Reitz [4]. The measured concentration field in the droplet wake is complemented with an estimation of the temperature field to provide insight into the role of evaporation on the droplet's surface temperature evolution. The temperature field is estimated using the concentration data applying the theoretical adiabatic mixing assumption by Qiu and Reitz [4]. The data is afterwards fitted by an iteration of the initial droplet temperature using the adiabatic mixing assumption at constant chamber conditions. Whilst the adiabatic mixing assumption has not been validated by independent measurements of temperature and concentration, we will apply this assumption to the experimental results to enable a direct comparison with the predictions from the theoretical models of Qiu and Reitz [4].

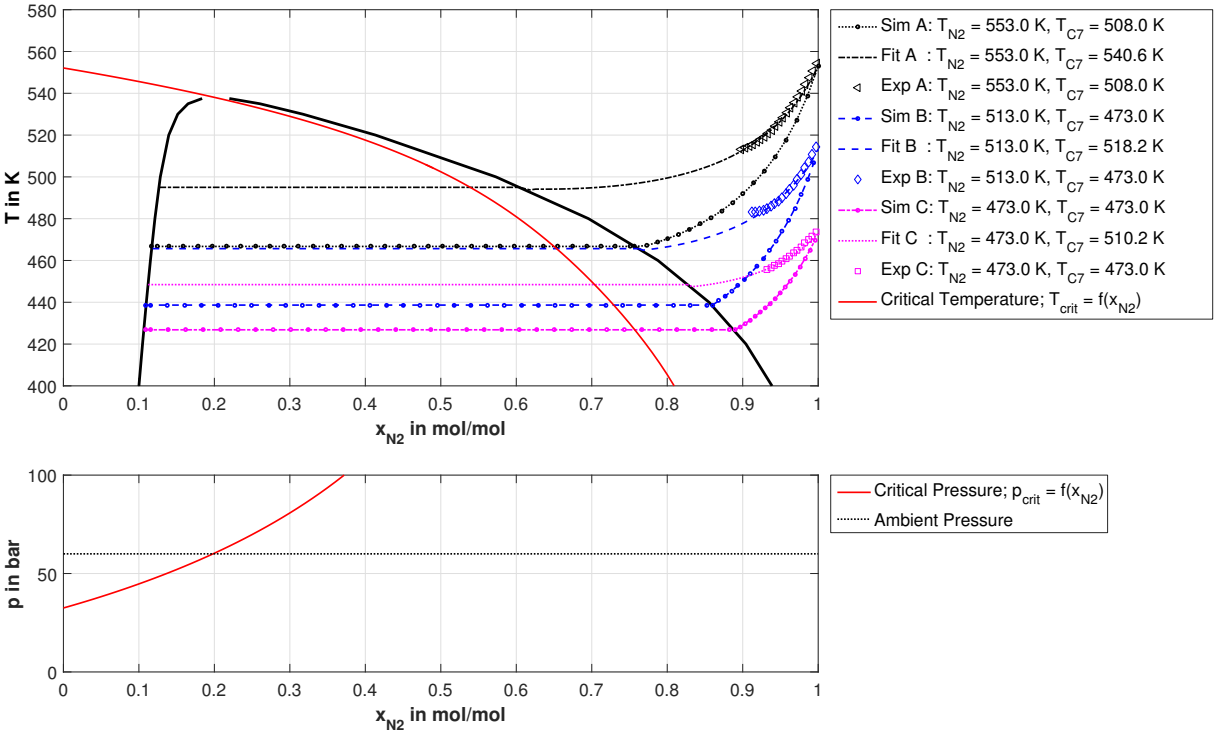


Figure 5. Correlation between measured mole fraction in the droplet wake and mixing temperature plotted with the vapour liquid equilibrium for a n-heptane-nitrogen (C7-N_2) mixture at 60 bar and the critical locus. The temperature in the vapour phase is estimated assuming adiabatic mixing using the PC-SAFT EOS.

Figure 5 shows adiabatic mixing lines fitted onto the experimental data (curves without markers), plotted together with the adiabatic mixing lines, derived according to the Qiu and Reitz [4] model (curves with markers). The latter are calculated for the experimental test conditions of the Raman scattering investigations. In contradiction to Qiu and Reitz [4], we assume a homogeneous droplet temperature, which is equal to the estimated surface temperature. Hence, the temperature inside the two phase region is constant. The different colours distinguish between the experimental cases A, B and C of the Raman scattering investigations. The critical temperature as well as the critical pressure of the mixture are plotted as well. Note, that critical temperature and pressure are solely depending on the composition of the mixture. Comparing the theoretical adiabatic mixing lines derived from the Qiu and Reitz [4] model (curves with marker) with the fitted experimental data (curves without markers) the results deviate systematically from each other. Especially the resulting interfacial temperature of the theoretical adiabatic mixing line, which is the intersection of the mixing line with the dew point line, is significantly lower. Additionally the necessary initial droplet temperature to fit the experimental data is higher for all cases. The same discrepancies can be observed using the adiabatic mixing assumption from Dahms [3]. Note, that this similar behaviour is to be expected, since adiabatic mixing by Dahms [3], and Qiu and Reitz [4] is similar as long as the phase stability limit is not reached. The observed discrepancy can be explained by an evaluation of the inherent assumptions of the two models. Both models consider the system in stable thermodynamic equilibrium for the initial conditions and couple the temperature in the vapour phase with the mole fraction assuming adiabatic mixing. However, the injection time and dwell time of the droplet or jet is in the range of seconds or milliseconds, respectively. Hence, it is highly questionable that thermal equilibrium is reached within this short residence time. The next section will focus on this by applying the evaporation model of Abramzon and Sirignano [5] to the discussed experimental cases.

Besides the mixing lines, figure 5 shows critical temperature and critical pressure of the mixture. In compliance with the transition criterion predicting the onset of diffuse-mixing by Qiu and Reitz [4], the droplet remains subcritical when the droplet's temperature and pressure remain subcritical. Hence, they are below the critical values of the mixture. In order to reveal the thermodynamic state of the droplet, the mole fraction in the liquid phase has to be taken into account. The latter can be extracted from the bubble point line assuming a homogeneous droplet temperature. The corresponding temperature is therefore the estimated interface temperature. Following this criterion n-heptane droplets remain subcritical for all our experimental conditions, since the droplet temperature is below the critical temperature of the liquid compositions. In the vapour phase in the vicinity of the droplet the critical pressure of the mixture exceeds the ambient pressure leading again to a subcritical thermodynamic state. In figure 6, the n-pentane-nitrogen (C5-N_2) fluid system of the front lighted shadowgraphy investigations are depicted. Applying the transition criterion by Qiu and Reitz [4] to the n-pentane cases, a subcritical droplet as well as a subcritical vapour phase at the boundary layer of the droplet is observed. Hence, for both fluid systems our experimental observations agree with the transition criterion proposed by Qiu and Reitz [4].

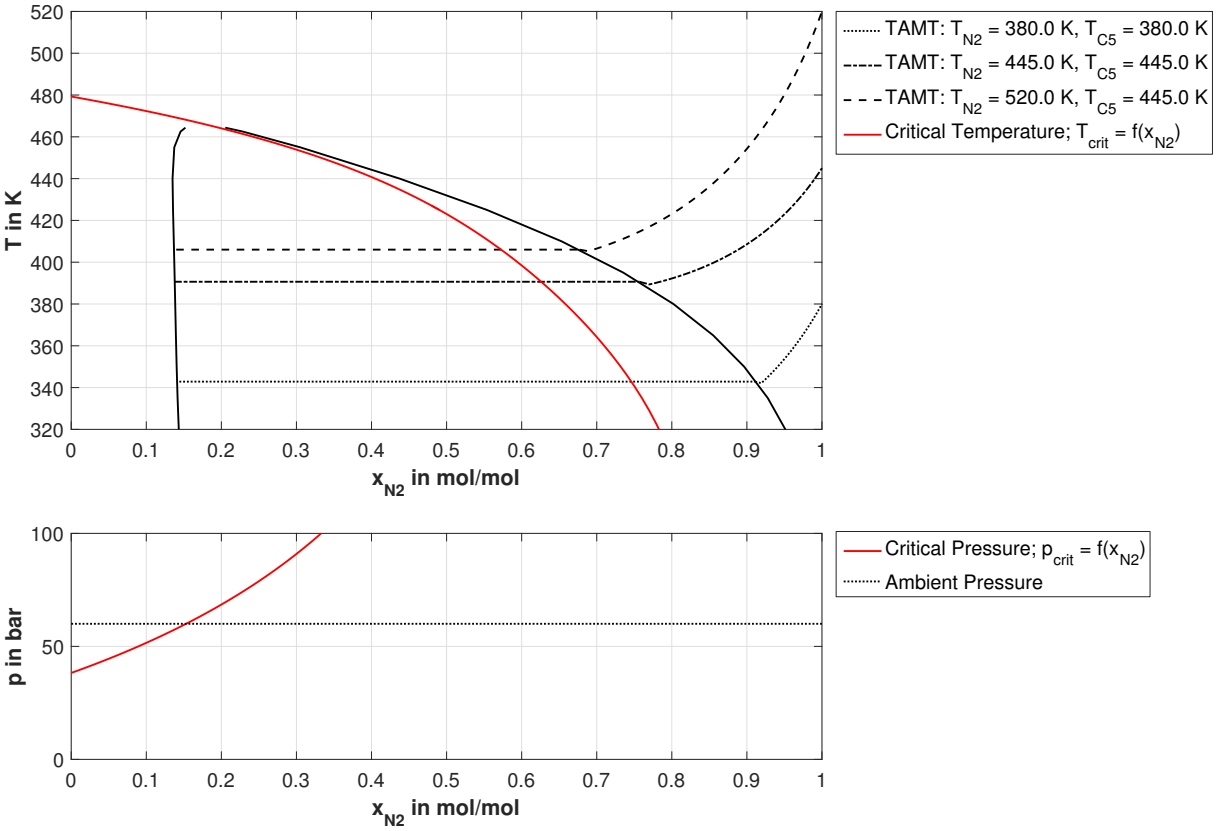
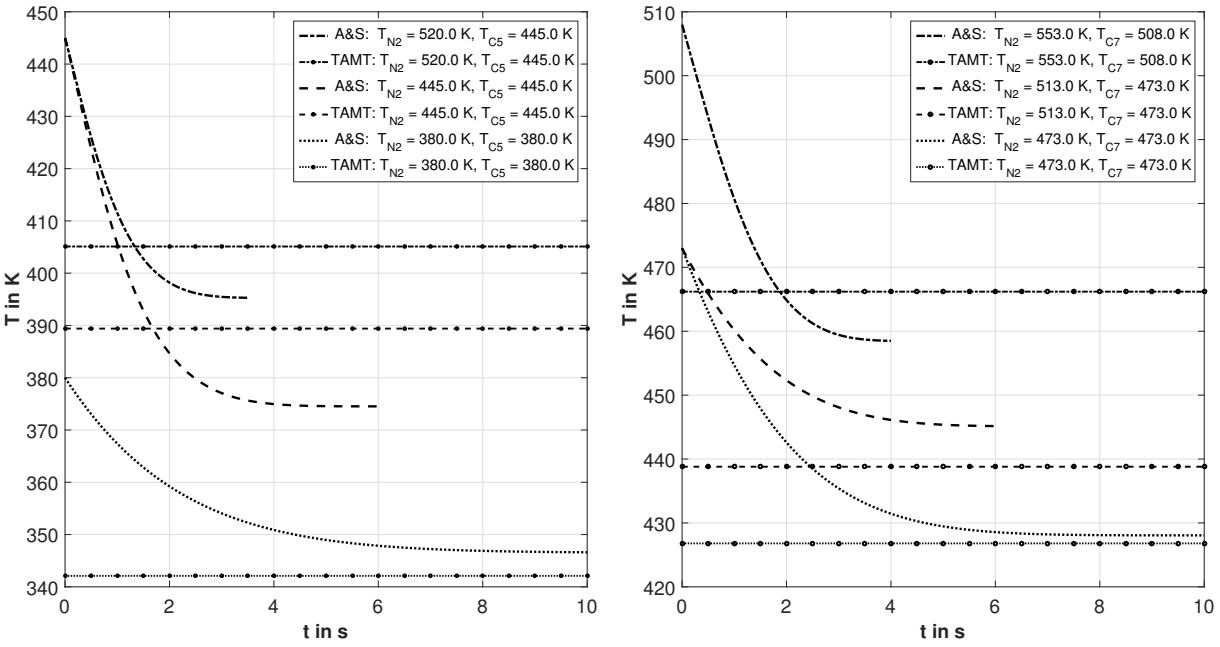


Figure 6. Theoretical adiabatic mixing temperature over mole fraction plotted with the vapour liquid equilibrium for a n-pentane-nitrogen (C5-N_2) mixture at 60 bar and the critical locus. The temperature in the vapour phase is estimated using the PC-SAFT EOS.

Characterization of the droplet surface temperature

The observations in the previous section reveal, that investigations of the surface temperature of the droplet are highly important to confirm the thermodynamic state of the droplet and account for evaporative cooling of the droplet. Since experimental investigations regarding the droplets surface temperature in the discussed setup have not been done yet, we will focus on an analytical approach to estimate the temperature history of the droplet. As a first strategy the equilibrium evaporation model of Abramzon and Sirignano [5] is used. Thermodynamic properties of the pure fluids are extracted from the NIST Database [11], whereas properties of the mixture are calculated according to Amagats law for gas mixtures. Diffusion coefficients are calculated using the method of Wilke and Lee [14] with Lennard Jones parameter extracted from Hirschfelder et al. [15] for n-pentane and nitrogen. For n-heptane the Lennard Jones parameter are listed in Chea and Violi [16]. Furthermore, the saturation pressure of the liquid in the vapour phase is corrected by the binary mixture composition in equilibrium using an enhancement factor as explained by Luijten [17]. The temporal evolution of the droplet temperature is shown in figure 7 for both n-pentane and n-heptane droplets. The initial droplet diameter is $D_0 = 1$ mm and the atmospheric pressure is $p_{amb} = 60$ bar.



(a) n-pentane droplet in nitrogen atmosphere.

(b) n-heptane droplet in nitrogen atmosphere.

Figure 7. Droplet surface temperature evolution over time compared to the theoretical adiabatic mixing temperature (TAMT) at the surface of the droplet. The latter is the intersection of the TAMT with the dew point line at vapour liquid equilibrium.

In figure 7, a comparison between the estimated droplet surface temperature with the theoretical value TAMT (true adiabatic mixing temperature) at the droplet's interface is depicted. The latter is the intersection between the theoretical adiabatic mixing line calculated as described by Qiu and Reitz [4] and the dew point line of the binary mixture. The fluid systems and experimental conditions are the same for figure 5 and 6. The comparison of the surface temperature shows, that for low initial droplet temperatures the TAMT at the interface sightly underestimates the wet bulb temperature. However, the agreement at high dwell times is good, due to the attainment of local thermodynamic equilibrium. Indeed, for lower initial droplet temperatures the equilibrium vapour mole fraction at the drop interface is rather low as shown in figure 5 and 6. Consequently the wet bulb temperature coincides with the TAMT. For higher initial droplet temperatures the equilibrium vapour mole fraction increases significantly. Hence, the vapour boundary layer at the droplet interface remains undersaturated, leading to a sustained evaporation rate. Concordantly, the surface temperature drops below the TAMT values. This discrepancy increases with decreasing latent heat as can be seen by comparing n-pentane (low latent heat) and n-heptane (high latent heat) in figure 7. This leads to the following conclusion: The TAMT can be applied to estimate the wet bulb temperature only for low initial droplet temperatures. However, one must consider that the latter is reached after a characteristic dwell time, which depends on the initial droplet size. In the described experiments the dwell time is approximately 0.5 s for the n-heptane cases and circa 2 s for n-pentane. Hence, using the TAMT to calculate the surface temperature would lead to an underestimation of up to 40 K of the droplet temperature and therefore seems, as discussed in the previous section, not applicable to calculate the temperature of the droplet surface. For high temperature cases models based on the assumption of global thermodynamic equilibrium between the droplet and the gaseous phase are not suited to characterise the droplet temperature at any time.

Conclusions

An experimental and analytical study of drop dynamics at high pressure and temperature conditions has been conducted. We employed front lighted shadowgraphy to investigate the thermodynamic state of the droplet by detecting the material surface of the droplet as well as Raman scattering to investigate the concentration field in the droplet wake. Both investigations were carried out on falling alkane droplets in a nitrogen atmosphere. These experiments were complemented with an analysis of a transitional criterion to predict the onset of diffuse-mixing. Additionally, an evaporation model was applied to study the evolution of the droplet temperature compared to the adiabatic mixing assumption. Our main conclusions are the following:

- The front lighted shadowgraphy setup was able to reveal the presence of a material surface by means of reflections and refractions on the material surface of the droplet. In all experimental cases the presence of a material surface was observed, even when the droplet was injected into a supercritical atmosphere. This observation can be explained by the evaporative cooling effects. Evaporation prevents the droplet from gasification by mitigating the heating of the droplet interface.

- The temperature field in the droplet wake, calculated from the concentration field assuming adiabatic mixing, further confirm the role of evaporation for the evolution of the droplet.

- From the presented experimental studies the Qiu and Reitz model [4] predicting the onset of diffuse-mixing shows good agreement. Hence, the critical locus of the mixture seems to be a suitable criterion to determine the thermodynamic state of a fluid injection. However, since the critical locus has not been reached yet, further experimental investigations at higher temperature levels are necessary.
- The comparison of the evaporation model of Abramzon and Sirignano [5] with the adiabatic mixing assumption of Qiu and Reitz [4] or Dahms [3], corroborates the role of evaporative cooling in the droplet evolution. Furthermore, the time scales of the fluid injection and dwell time have to be taken into account when calculating the temperature distribution in high pressure injection process.
- By applying the critical locus as the onset criterion of diffuse-mixing a global thermodynamic equilibrium is assumed. Evaporative cooling effects, however, lead to strong departure from global thermodynamic equilibrium and therefore may deduce erroneous results. This is especially true, for fast evaporation processes and short injection times. Hence, a detailed evaluation of the prediction of the onset of diffuse-mixing requires further investigations in the vicinity of the critical locus and eminently above the critical line.

Acknowledgements
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