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One-dimensional Raman spectroscopy and shadowgraphy for the analysis of the evaporation behavior of acetone/water drops

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ABSTRACT

This paper reports – for ambient conditions – experimentally determined surface regression rates of acoustically levitated acetone/water drops as a function of the mixture composition at the drop boundary. The fluid levitated as a drop was a mixture of acetone and water with vapor pressures of 24.5 kPa and 3.17 kPa, respectively, at ambient conditions. As expected the acetone evaporated faster from the acetone/water drop than water. Already small quantities of acetone in the mixture significantly increased the surface regression rate of the drop.

Temporally and spatially resolved composition profiles were measured along a line through the drops using one-dimensional Raman spectroscopy. Shadowgraphs of the evaporating drops were acquired, from which the drop shrinkage was derived. Due to the good spatial (120 μm) and temporal (1 s) resolution of the one-dimensional Raman experiment the evolution of the radial composition profiles through the drop could be followed showing that the diffusion-process inside the drop plays an important role in the binary evaporation process.

1. Introduction

The fractional evaporation of bi-component liquids dispersed as a spray has a wide impact in various fields of engineering. In this context "fractional evaporation" means that, due to the preferential evaporation of the more volatile compound, the composition of the remaining liquid phase gets richer in the less volatile compound due to different vapor pressures of the components. Applications of multi-component dispersed liquids range from energy engineering (spray combustion or direct injection internal combustion engines) to particle technology (spray drying or spray polymerization). In particles-from-spray processes, for example, often mixtures of solvents are used to tune the solvation characteristics of the solvent-mixture to the requirements of the involved process [1]. Here, due to fractional evaporation of the dispersed liquid to the bulk gas phase, the composition of the liquid solvent mixture can change and with this the solvation characteristics, which influence the particle formation process. Next to the fact that fractional evaporation can change the overall composition of multi-component drops – exceptions are azeotropic mixtures – it also influences the radial composition profiles of drops [2]. Radial composition profiles develop if the resistance of mass transport $R_{i,k}$ inside (index i) the drop is different for various compounds k, and if these resistances $R_{i,k}$ are not negligibly small compared to the mass transport resistance $R_{o,k}$ of the same compound k outside (index o) the drop. Here $R_{i,k}$ and $R_{o,k}$ are the mass transport resistances of the compound k in the liquid and the gas phase, respectively. The developing radial composition gradients have to be considered as they control the overall evaporation rate of the drop as well as other processes and properties inside the drop, such as heat and mass transport processes, phase transition processes and the mutual solubility of the compounds inside the drop. Therefore, the provision of straightforward measurement techniques for the determination of radial composition profiles during the fractional evaporation of bi-component liquids is of particular importance for spray modeling approaches as existing models can be verified or developed further.

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While it is difficult to investigate into individual drops in a spray, it is possible to study single drops by means of acoustic levitation. The non-intrusive, time-resolved and contactless Raman spectroscopy has been successfully employed in acoustically levitated drops for the analysis of phase equilibria [3] and (re)-crystallization processes [4–6]. It has also been applied to track the simultaneous evaporation and polymerization process [7], dehydration of particles [8], formation and aggregation of nanoparticles [9], and even the dynamics of red blood cells and detection of hemozoin in malaria-infected cells [10]. As the investigations mentioned above collected the Raman spectra as an integral signal from the entire drop, it was not possible to resolve the mechanisms taking place inside the drop spatially, and thus radial profiles have not yet been provided on a single-shot-measurement basis. Therefore in this study, we demonstrate a Raman-based one-dimensional imaging technique for spatially- and temporally-resolved *in situ* composition measurements along a line through the levitated drops, which makes the radial composition profiles in fractionally evaporating bi-component drops accessible. From the evolution of the radial composition profiles it was derived that the diffusion plays an important role in the binary vaporization process. As we correlated the Raman measurements with shadowgraphy measurements, we – for ambient conditions and when solely modifying the initial composition of two drops – were additionally able to quantify the surface regression rate as a function of the composition of the drops.

2. Experimental

In Fig. 1 we present the experimental setup including the acoustic levitator and the self-assembled Raman sensor, which provides line composition profiles through levitated drops. Acetone, which was acquired from Merck with 99.9% purity, and the deionized water were used without further purification. For the formation of evaporating drops, liquid mixtures of water and acetone (before delivery at 295 K) were delivered manually through a capillary into one of the node points of the acoustic levitator. As the levitator was not housed, the levitated drops were subject to the convection in the laboratory, which on the one hand caused a little movement/oscillation of the levitated drops within the pressure node of the acoustic levitator and on the other hand promoted the transportation of already evaporated substance away from the drop. The levitation itself also causes non-negligible forces on the drop's surface which, according to Brenn et al. [11], can induce convection inside the drop.

The evaporation of two drops was analyzed at room temperature (295 K) and at ambient pressure with the Raman sensor, for which the initial compositions of the fed acetone/water mixtures are provided in Table 1. Due to the continuous evaporation of the levitated drops and due to the time delay between the positioning of the drops and the first measurement event, the compositions measured inside the drop during the first measurement event deviate from the composition of the fed mixture, as it can be seen in Table 1. Acoustic levitation allowed for a contactless positioning of a sample near a pressure node of a standing acoustic wave (here 42 kHz) emitted by a sonotrode fixed diametrically opposite to the concave reflector. While detailed descriptions on acoustic levitation are provided in references [12,13], our acoustic levitator is specified in reference [14].

2.1. Shadowgraphy

Images of levitated drops were acquired using a shadowgraphy setup. While – for a clear presentation of the Raman experiment in Fig. 1 – the light source required for the shadowgraphy experiment and the path of the light aligned for the shadowgraphy measurements are not shown in Fig. 1(b), the detector used for the shadowgraphy measurements is shown and labeled as CCD 1. The shadowgraphy measurements were carried out decoupled from the Raman measurements in a different set of experiments as the Raman excitation laser would have interfered with the shadowgraphy measurements. Exemplary shadowgraphs of one evaporating levitated drop are provided in Fig. 2(c), from which also the size and the shape of the drops can be extracted. It can be seen that with decreasing drop size its initially oblate-like deformed shape transforms more and more into a spherical one [15,16]. From the shadowgraphs the temporal evolution of the contour of the evaporating non-spherical drops was extracted, from which their volume-equivalent diameter was computed.

2.2. The Raman experiment

We used a frequency-doubled continuous wave Nd:YVO_4 laser as a Raman-excitation source, operating at 532 nm with an output power of 2.0 W. After passing through a Galilean telescope and a converging lens, the laser beam was focused into the drop, the

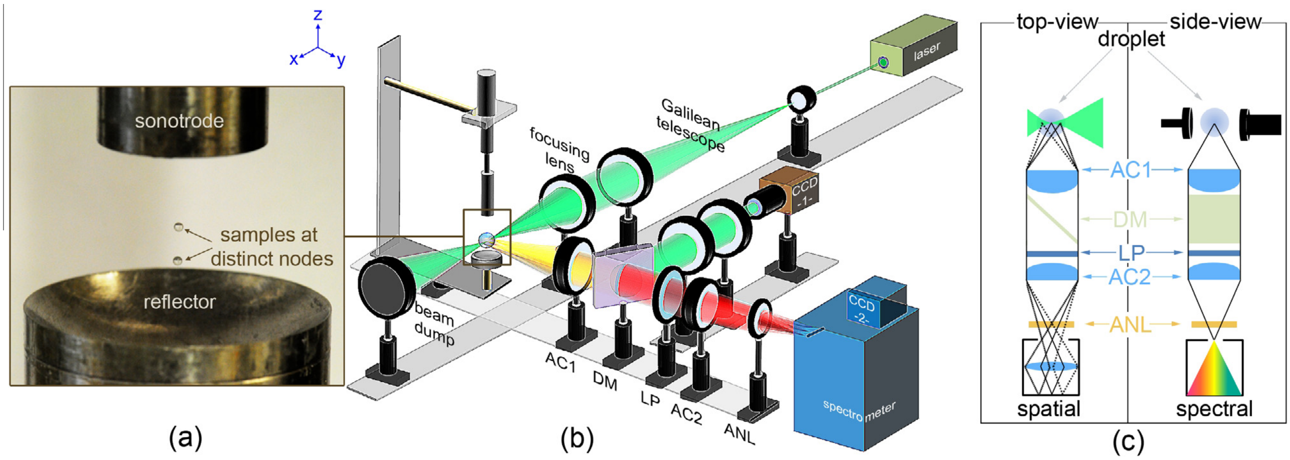

Fig. 1. (a) Image of drops levitated between a sonotrode and a reflector in two node points; (b) sketch of the experimental one-dimensional Raman experiment; (c) top- and side-views of the Raman detection part of the setup consisting of the achromatic lenses (AC1 and AC2), dichroic mirror (DM), long-pass filter (LP) and a polarization analyzer (ANL). Visualization of the drops via shadowgraphy and control of the position of the laser beam waist inside the drop via elastic light scattering are realized using charge-coupled device (CCD) 1. Detection of the Raman signals is realized using CCD 2.