Technique for Measurements of the Evaporation Rate of Single, Freely Falling, Droplets

by

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ABSTRACT

An experimental setup has been developed in order to determine evaporation rates of freely falling droplets. The droplets may consist of pure liquids, of a mixture of different liquids, or of a solution. After a droplet has been produced by a droplet on demand generator, it falls downwards along a vertical observation channel. Meanwhile a laser beam is directed vertically upwards in order to illuminate the droplet continuously. Shortly after the droplet has entered the observation channel the laser light scattered into the forward hemisphere is detected by a CCD camera. From the angular spacing of bright fringes in the intensity distribution of the scattered light the droplet size is determined. While the droplet is falling and evaporating the intensity of a part of the scattered light is detected by a photomultiplier. As the droplet is evaporating, its size is decreasing and morphology dependent resonances become visible in the temporal evolution of the intensity of the scattered light. From the time intervals between of the resonance peaks the evaporation rate is determined. On one side of the transparent observation channel a horizontal grid with transparent equidistant slits is posted. A second photomultiplier detects the scattered light, when the droplet passes the transparent slits. From the output signal of this photomultiplier the droplet velocity is determined. These measurement techniques allow to monitor droplet size and velocity during the evaporation process. It is important, that the measurements start shortly after droplet generation. Therefore this technique allows to study droplets consisting of liquids with higher volatility than it is possible with the optical levitation technique. A combination of this technique and the optical levitation technique allows to study mixture droplets consisting of higher and lower volatile components. Measurements of different n-alkanes have been performed. The results obtained have been compared with a simple analytical model and show a good agreement. First results for mixture droplets have been obtained.
1. INTRODUCTION

The evaporation rate of single droplets has been measured at the Institute of Aerospace Thermodynamics (ITLR) using an optical levitation technique described by Roth et al. (1995). With this technique evaporation rates of pure or binary mixture droplets of slowly evaporating hydrocarbons have been studied successfully (Gartung et al., 2000). With this technique it lasts at least 2-3 seconds until the droplet is levitated and stabilized before the measurements can start. For very low evaporation rates this initial phase of evaporation is not of particular interest. This is true for instance for liquid hydrocarbons with thirteen or more carbon atoms at room temperature. However, if the evaporation rate is high, the droplet may fully evaporate during these first seconds. This is true for more volatile hydrocarbons with less than ten carbon atoms or for higher ambient temperatures, as the evaporation rate is very sensitive on temperature (Wilms et al., 2001). For the case of mixture droplets containing components with a high volatility the initial phase is essential for the description of the whole evaporation process.

In order to overcome this lack of data during the initial phase of the evaporation process an experimental technique has been developed, which allows to determine the evaporation rate of droplets with a higher volatility. Then the important initial phase during the evaporation of mixture droplets containing high volatile components can be studied. The technique is mainly based on the detection of morphology dependent resonances of freely falling droplets.
2. EXPERIMENTAL SETUP AND MEASUREMENT TECHNIQUES

The droplets are produced by a droplet on demand generator. In changing the orifice of the generator the droplet diameter can be varied from approximately 20 µm to 100 µm. After generation, the droplets are falling vertically downwards along a squared channel made of glass with an intersection of 1.75 cm$^2$. The droplets are ejected horizontally into the channel by the droplet generator. Due to the drag of the droplets, they are decelerated and fall freely driven by the gravity force vertically downwards along the observation channel. As the observation channel can be purged by a slow air stream directed from top to bottom, the air stream velocity of only a few centimeters per second is superimposed to the free fall velocity of the droplets. A schematic view of the observation channel together with the measurement facilities is shown in Fig. 1.

A laser beam is directed vertically upwards within the channel in order to illuminate the falling droplet permanently. In contrast to the optical levitation technique the intensity of the laser beam is rather weak, minimizing its influence on the falling droplet. With this experimental setup the initial droplet size, the evaporation rate, and the droplet velocity, can be determined.

In order to explain the measurement techniques a n-decane droplet falling freely downwards along the center of the observation channel is used as an example in this section. The ambient temperature in the channel was 25°C in this case. Immediately after the droplet has been injected the channel, it falls freely downwards and reaches position 1 (Pos. 1). In this position the light scattered into the forward hemisphere is detected by the CCD camera. The camera is located exactly in the focal plane of the cylindrical lens in order to obtain a position insensitive intensity distribution of the scattered light. From the angular distance of the bright fringes in the intensity distribution, shown in Fig. 2, the initial droplet size can be obtained (König et al., 1986).

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**Fig. 2:** Bright fringes in the intensity distribution of the light scattered in the forward hemisphere (top). Shown is a picture taken by the CCD camera. The diagram at the bottom corresponds to the intensity distribution across the horizontal centerline of the picture above.

Part of the light scattered by the droplet is focussed on photomultiplier I while the droplet is falling along the observation channel. The output of the photomultiplier is stored on a personal computer. The recording starts, when the droplet is produced by the droplet generator. When the droplet reaches position 1, the scattered light is
detected by the photomultiplier producing an output signal, which triggers the CCD camera. In this way the output of the photomultiplier and the size measurement is related precisely. The input window of the photomultiplier allows to detect the scattered light at every position along the observation channel.

![Graph showing output signal of photomultiplier I. In the enlarged view the peaks of the morphology dependent resonances can be found.](image)

**Fig. 3:** Output signal of photomultiplier I. In the enlarged view the peaks of the morphology dependent resonances can be found.

![Graph showing time interval Δt between neighbouring resonance peaks.](image)

**Fig. 4:** Time interval Δt between neighbouring resonance peaks.

As an example the light paths of the scattered light in position 2 (Pos. 2) of the droplet are sketched in Fig. 1. If the droplet evaporates its radius changes resulting in oscillations of the scattered light (Bohren & Huffman, 1983 and Popp, 1994). An output of the photomultiplier I can be seen in Fig. 3. As mentioned above, the time t = 0
corresponds to the time at which the droplet has been generated, the measurement starts a short time later, when the droplet passes position 1. The time delay until the measurement starts is much more smaller than in optical levitation techniques.

In evaluating these morphology dependent resonances the radius change $\Delta r$ within a time interval $\Delta t$ can be obtained. Here $\Delta t$ means the time interval between corresponding resonance peaks. The radius change $\Delta r$ within $\Delta t$ depends on the refractive index of the droplet liquid and is nearly independent on the droplet radius (Chylek, 1990). In Fig. 4 the results obtained from the data shown in Fig. 3 of an n-decane droplet are presented. In Fig. 4 the temporal evolution of the time interval $\Delta t$ is plotted during the evaporation process.

These results can be evaluated in different ways. In order to obtain the droplet radius $r$ as a function of time $t$, it is possible to start at time $t = t_1$ with the measured droplet radius $r_{1m}$ in position 1. For each time interval $\Delta t$ the radius decreases by $\Delta r$. This is an integration process. However, it is possible to obtain this result from the data shown in Fig. 4 alone without using the direct measured value $r_{1m}$ by assuming the validity of the $d^2$-law. From the $d^2$-law one obtains

$$ t = \frac{r_0^2 - r^2}{\beta} \quad (1) $$

with the evaporation rate $\beta$ and the radius $r_0$ at time $t = 0$. Differentiation of Eq. (1) with respect to $r$ gives

$$ \frac{dt}{dr} = \frac{-2r}{\beta} = \frac{-2}{\beta} \sqrt{r_0^2 - \beta t} \quad (2) $$

When this equation is squared linear behavior is obtained

$$ \left( \frac{dt}{dr} \right)^2 = \frac{4r_0^2}{\beta^2} \frac{4}{\beta} t \quad (3) $$

Fig. 5: Droplet diameter $D$ as a function of time $t$. The results are obtained by two different evaluation techniques. In addition the measured initial droplet diameter is shown.
This linear behavior can easily be compared with the measured values shown in Fig. 4, if the values of $\Delta r$ are divided by $\Delta t$ and if the result is squared. A linear regression $y = a + bx$ of these data can directly be compared with Eq. (3) and one obtains $\beta = 4a$ and $r_0 = \frac{1}{2} \beta b^{1/2}$. In order to obtain the value of $r = r_{1c}$ at position 1 without a direct measurement, the time $t_1$, when the measurement of the size has been triggered, has to be inserted in the equation for the $d^2$-law. A comparison between the results of both methods is shown in Fig. 5.

As the method, which needs the initial diameter measured, is based on integration a rather smooth curve is obtained. However, the integration method can be applied to the calculated initial diameter too and a similar smooth curve is obtained. For the present example the measured diameter was $D_{1m} = 42.5 \mu m$ and the calculated initial diameter was $D_{1c} = 41.9 \mu m$.

The droplet velocity has been determined by photomultiplier II in a simple way. Photomultiplier II is arranged in the same way as photomultiplier I. The observation channel has, however, on the side where the scattered light is observed by photomultiplier II, a grid with horizontal equidistant slits, which are transparent. Therefore, only when the droplet passes a transparent slit, a signal can be detected. An outcome for the example discussed above is shown in Fig. 6.

![Image of Fig. 6](image)

*Fig. 6: Output signal of photomultiplier II. The intensity maxima correspond to the times the droplet passes the transparent slits in the window the droplet is observed through.*

As the distance $\Delta s^*\$ between the transparent slits is known the mean velocity $v = \Delta s / \Delta t$, of the droplet can be calculated from the time interval $\Delta t$ between two neighbouring intensity maxima. The droplets are assumed to fall in the center of the observation channel. The distance $\Delta s$ differs from the distance $\Delta s^*$. As the geometrical arrangement is known, $\Delta s$ can be calculated from $\Delta s^*$. The result of the evaluation is shown in Fig. 7.

As the falling velocity is a function of droplet size the falling velocity decreases as the droplet evaporates. In this example the observation channel has been purged before the measurement and not during the measurement.
3. RESULTS

In this section the influence of purging the observation channel during the measurements and different examples of evaporating droplets are shown.

Fig. 7: Mean droplet velocity $v$ as function of time $t$.

Fig. 8: Mean droplet velocity $v$ as function of time $t$ for two different volume rates 1.78 ml/s and 3.5 ml/s. The circular symbols indicate the result with no purging. The higher the volume rate the higher the droplet velocity.
Purging the observation channel is absolutely necessary in order to avoid vapor accumulation within the chamber, which may reduce the evaporation rates measured. Purging should be performed in any case before the measurement starts. However, the channel can be purged during the measurements. Then the droplets pass position 1 at an earlier time after generation, as can be seen from Fig. 8.

This has an advantage for investigations of volatile droplets. As the droplets are rather small it is assumed in a first approach, that the droplet velocity is obtained by superposition of the velocity of the gas in the center of the channel, where the droplets are falling, and the free fall velocity of the droplets. In Fig. 8 the droplet velocity is plotted against time for different volume rates through the observation channel. The circular symbols represent the data from Fig. 7 without purging the chamber. As expected, the droplet velocity is increased together with the volume rate. The volume rate through the observation channel can be controlled by a valve and measured by a rotameter.

Different n-alkanes have been studied. In Fig. 9 the output signals of photomultiplier I for a n-decane and a n-undecane droplet are shown. The signal of the n-undecane droplet with thirteen resonance peaks is caused by a decrease of the droplet diameter of approximately 1.4 μm. This shows clearly that this technique is able to detect very small changes in droplet size.

![Fig. 9: Output signals $U_{pI}$ of the photomultiplier I as a function of time $t$ from a n-decane and a n-undecane droplet.](image)

A comparison with an analytical model of the evaporation rates of n-nonane, n-decane and n-undecane is given in Fig. 10. The difference between the analytical model and the n-decane droplet may be caused by inaccuracies in the temperature measurement in the experiment.
4. CONCLUSIONS

The technique described above allows to measure evaporation rates of droplets with higher volatility. The setup is suitable to study mixture droplets and solutions, when the initial phase of the evaporation process is important. This means that mixture droplets with high volatile components in addition with less volatile components can be studied. In combination with optical levitation techniques the whole droplet history can be investigated. The technique closes the gap in observation time from shortly after the droplet production to the moment at which optical levitation techniques can be applied. Results of the combination of this technique and the optical levitation technique will be presented on the "19th Int. Conf. on Liquid Atomization and Spray Systems (Europe)" in Nottingham (Wilms et al., 2004).

REFERENCES


Wilms, J., Roth, N., Arndt, S., & Weigand, B. 2004 Evaporation of ternary mixture droplets. In *Proc. 19th Int. Conf. on Liquid Atomization and Spray Systems*, ILASS (Europe), accepted for publication.