Concentration Measurement of Dispersion Droplets using the Time-Shift Technique

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ABSTRACT

The characterization of droplets is important for a variety of applications, such as spray painting or spray drying. For example, in coating processes, small droplets lead to overspray, whereas large drops may blemish the surface. Additionally, for droplets of disperions, emulsions or suspensions, the concentration of the dispersed phase within the droplet is of high interest as it can directly influences the properties of the process, for example the solubility of spray dried powders.

The time-shift technique provides an efficient and accurate method to measure individual droplets in sprays. The present study explores the possibility of extending the time-shift technique to also measure the concentration of the dispersed phase in a droplet, through estimation of the light absorption/scattering intensity. The proposed method is validated using experiments, in particular, we show how the time-shift technique can distinguish between different aqueous milk concentrations.

1. Introduction

Spray characterization is important for a variety of applications, such as spray painting or spray drying. The efficiency and quality of these spray processes depend on atomization parameters like flow rate, injection pressure, airflow rate etc. that directly influence drop sizes and velocities. For instance, in coating processes, small droplets produce overspray; hence to wasted material. On the other hand large drops may lead to surface defects. Therefore, a detailed knowledge about drop size distributions is essential for process optimization. Another important parameter of dispersion droplets is the concentration of the dispersed phase.
For measuring the size and velocity of the droplets there are numerous methods and techniques; a good summary can be found in Tropea (2011) [14]. In the present study we focus on the time-shift (TS) technique since it provides an efficient and accurate method to measure size and velocity of individual droplets in sprays in real time [1], [2], [13]. One advantage of the TS technique is the possibility of characterizing not only transparent but also non-transparent particles and droplets [9], which often occur in industrial applications. Additionally, this technique operates in backscatter and does not require post-factory alignment, which is highly attractive for industrial applications.

The time-shift technique was first introduced by Semidetnov in 1985 [13] and was further developed by Damaschke et al. in 2002 [1], [2]. The TS technique is also known as the pulse displacement technique, where several variations have been discussed in [5],[6]. Most recent developments and validation of this technique can be found in [8] and [9]. The basic principle of this technique is the light scattering of droplets passing a shaped laser beam. At least two sensors, located at different scattering angles, detect the scattered light. The time shift between two acquired signals and the characteristics of the measured signals directly lead to an estimation of size and velocity.

So far, only size and velocity measurements with the TS technique have been studied in literature. The most recent advances concern the use of the technique for characterizing semi-transparent droplets, especially estimating the concentration of the dispersed phase leading to the non-transparency. This idea is not entirely new: proposals for measuring the concentration have been made in earlier studies of the time-shift technique; however neither a quantification of the effect, nor an implementation has been presented [10], [11]. Since the concentration of droplets directly influences the efficiency of the process that parameter becomes one the most important ones when characterizing sprays of suspended droplets.

The present study explores the possibility of measuring the concentration of the dispersed phase in a droplet, through estimation of the light absorption/scattering intensity. Based on experiments, we propose a first solution to estimate the concentration of droplets by using the time-shift technique. In particular, we show how the time-shift technique can distinguish between different milk concentrations.
2. The Time-Shift Technique

The measurement principle of the time-shift technique is only briefly summarized here, since adequate descriptions can be found elsewhere, e.g. Albrecht et al. (2003) [13]. The measurement principle of the time-shift technique is based on light scattering of a single particle from a shaped light beam. When a drop passes through the focused light beam, the scattered light is detected by photodetectors focused onto the scattering center. Each photodetector yields a time dependent signal (Fig. 1a), where the time delay between signals from different detectors is correlated to the droplet size and velocity [12].

An example time-shift signal originating from a water droplet passing through a Gaussian shaped beam and collected at a scattering angle of 150deg is illustrated in Figure 1 a), in which signal components are observable from the following scattering orders: surface wave long path, reflection (p=0), second-order refraction mode 1 (p=2.1), second-order refraction mode 2 (p=2.2) and surface wave short path [10]. The intensity of the scattering orders is described by the Debye series [5] expansion of the Mie [7] scattering functions, or by using a geometric optics approach [4],[15] to compute the scattered field.

In the case of non-transparent particles, e.g. droplets of emulsions, dispersions or suspensions, refracted light rays will no longer propagate undisturbed through the droplet due to absorption or secondary scattering from the dispersed phase, and these rays no longer contribute directly to the scattered light at a given detector position. Consequently, the time-shift signal includes only a reflection peak and no further peaks corresponding to refraction [11]. Nevertheless, light entering the suspension/emulsion particles can scatter from scattering centers of the dispersed phase within the particle and this may result in some scattered light intensity observed at the detector (Fig. 1b).
3. Experimental methods and observations

A droplet generator with three different apertures is used in order to produce droplets with different sizes (Fig. 2).

The sprayed material is milk, mixed with water. The milk – water ratio is varied in order to obtain three different concentrations, i.e. 100%, 50% and 15% milk. All generated droplets are measured in size and velocity by the time-shift technique (SpraySpy SSA15VW125 [16]) in the conventional manner.

The major focus of this contribution is the dispersed phase concentration of individual droplets. To find a first solution for this problem, signals of different concentrations are compared. Fig. 3 exemplarily shows two typical measurement signals, generated by two different concentrations.
Fig. 3: Received time-shift signal from a milk drop with water-milk ratio of 15% milk (a) and 100% milk (b)

Both signals show the reflection peak and the internal scattering as introduced in Section 2. However, the main difference is observed in the size of the baseline signal and the peak arising from the reflective glare point observed from the wetted drop surface. The key idea is to use that information to distinguish between different concentrations within a droplet. As absolute intensities often lead to inaccurate interpretation due to different gain factors, intensity ratios are preferred. We suggest using the ratio between the amplitude of the baseline $A_{\text{baseline}}$ and the amplitude of the reflection peak $A_{\text{refl}}$.

In order to define the internal scattering intensity and the reflection peak properly, a physical model is essential. The intensity model of the reflection peak $I_R(t)$ (i.e. order $p = 0$) is well known [11] and is given by

$$I_R(t) = A_{p=0}(m, \theta_S) \exp \left( -\frac{2(t-t_0^{(p=0)})^2}{\sigma^2} \right), \quad (1)$$

where $A_{p=0}(m, \theta_S)$ is the amplitude depending on refractive index $m$ and scattering angle $\theta_S$, $t_0^{(p=0)}$ is the peak position and $\sigma = \frac{w_0}{v}$, where $w_0$ is the laser beam width and $v$ is the drop velocity. In the following we denote $A_{\text{refl}} = A_{p=0}(m, \theta_S)$ as the amplitude of reflection.

The model of the formulated baseline is non-trivial as it has to capture the internal scattering when a shaped beam focusses a suspension. To begin with, we use the model as proposed in [11]. Here, the droplet is irradiated slice wise that causes the internal scattering. The scattered intensity, called the baseline intensity, $I_B(t)$ is given by

$$I_B(t) = C_0 w_0 (d^2 - 4v^2 (t - t_0)^2), \quad (2)$$

where $d$ is the drop size, $t_0$ is the baseline position in and $C_0$ includes the information of the optical properties and the concentration of scattering centers and incident intensity of the light.
beam. Here we identify $A_{\text{baseline}} = C_0 w_0 d^2$.

In the next step we estimate the parameters $A_{\text{baseline}}$ and $A_{\text{refl}}$ using a maximum likelihood approach. Therefore we define the complete suspension model

$$I_S(t, \boldsymbol{\theta}) = I_B(t) + I_R(t), \quad (3)$$

where the parameter vector $\boldsymbol{\theta}$ contains all unknown parameters such as the baseline amplitude $A_{\text{baseline}}$ and the reflection amplitude $A_{\text{refl}}$. Since we want to estimate $A_{\text{baseline}}$ and $A_{\text{refl}}$ to build the mentioned ratio, we use a maximum likelihood estimation defined as

$$\hat{\boldsymbol{\theta}} = \arg\min_{\boldsymbol{\theta}} \| r(t) - I_S(t, \boldsymbol{\theta}) \|_2, \quad (4)$$

where $r(t)$ is the received measured data over time, $\hat{\boldsymbol{\theta}}$ is the estimated parameter vector and $\| \cdot \|_2$ is the $L_2$ norm that is minimized. Fig. 4 illustrates the example, where the model parameters are estimated from the measured data.

![Figure 4](image)

**Fig. 4**: Received signal from a milk droplet. (Solid line) The original measured signal of a milk droplet. (Dashed line) Model estimation of the signal.

Fig. 5 shows the results, where the estimated characteristic parameter $A_{\text{baseline}}/A_{\text{refl}}$ is plotted for the three different concentrations and for three different drop sizes.
We observe that $A_{\text{baseline}}/A_{\text{refl}}$ is directly correlated to the concentration of the measured droplets, i.e. the parameter increases when the concentration of the drop increases. A similar correlation is observed for different drop sizes that we produced by using different apertures. The size dependence can be explained by the following. The amplitude of the reflection peak in the signal is not expected to be strongly size dependent, since the receiving aperture will result in an effectively small glare point being observed on the droplet surface, i.e. the surface curvature will not play a large role. On the other hand the baseline portion of the signal, due to the internal scattering and absorption, will decrease as the path length of light through the particle increases, as would be the case with larger particles. Furthermore, this size dependence would then be expected to be stronger for high concentrations of the dispersed phase (higher absorption), which is also observed in Fig. 5. The size dependence exhibited for 100% milk is significantly higher than for the 15% milk solution. In conclusion, the amplitude ration can be used to directly estimate the concentration of a droplet, once an appropriate calibration curve, such as shown in Fig. 5, is available.

4. Conclusion and Outlook

In this study we illustrate how to measure the concentration of a dispersed phase within a drop using the time-shift technique. Proof of concept is demonstrated by performing measurements on aqueous milk solutions of different concentration and showing the dependence on the signal features.

Future work will focus on developing a physics based model describing the scattered light of the dispersion droplet using a radiative transfer ansatz. This should yield a better model for parameter estimation from the signals.
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