Measurements of droplet vaporisation by means of light scattering

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Abstract

We describe how traditional elastic low-angle light scattering techniques (LAELS) can be employed to determine the evaporation rate of droplet arrays. This objective is achieved by measuring simultaneously the droplet size and spacing (from which the droplet speed can be derived) at different axial positions along the droplet path. The analysis shows that both quantities can be determined with good accuracy. The capability of the LAELS technique in performing reliable particle sizing has been tested by comparing experimental values with theoretical predictions. In addition, the method proved to be a valid tool for studying complex interaction phenomena between vaporising droplets, where competing effects between neighbouring droplets may result in a substantial decrease of droplet vaporisation rate when compared to the isolated droplet case. The temporal evolution of four different droplets arrays has been determined and the influence of interdroplet spacing on the vaporisation process correctly predicted.

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1. Introduction

This paper presents an alternative use of the low-angle elastic light scattering technique (LAELS) aimed at the simultaneous determination of both droplet size and spacing in the case of periodically generated droplets. Essentially, the droplet size is obtained by collecting the scattered light at small angle (as customarily done), while the droplet speed is deduced by exploiting the interference fringe pattern, associated to the droplet stream, as a way to determine the droplet distance and hence indirectly the speed. The technique is highly suitable for studying basic phenomena such as the evaporation or combustion of small droplets. In this study, we focus in particular on the evaporation of small droplets embedded in a high-pressure and/or high-temperature environment, as well as the mutual interaction between neighbouring droplets. Although very fundamentals in nature, these experiments find a direct technological application in modern propulsion systems, such as liquid-propellant rockets, gas turbines, and diesel engines. In all these systems, liquid fuels and/or oxidiser are usually injected into the combustion chamber as a spray of droplets, which undergoes a sequence of vaporisation, ignition and combustion processes in a high-temperature, high-pressure environment. For a clean and complete combustion, the spray performance and a good air-fuel mixing (before ignition) play a key role, thus requiring a thorough understanding of the different mechanisms for spray break-up, mixture formation and evaporation. Concerning the evaporation process in dense fuel sprays, there are two competing factors, which strongly influence the vaporisation process, namely droplet interaction and high-pressure/temperature effects. The importance of droplet interaction has not gone unrecognised in the literature. Leiroz and Rangel [1] performed a numerical simulation of the quasi-steady combustion of an infinite stream of droplets in a stagnant environment and showed the effects of droplets interaction on droplet mass vaporisation for different inter-droplet spacing. Due to the scarcity of experimental data, their results
were compared, for validation, to analytical solutions in the limit of isolated conditions. Following their findings, due to droplet interactions a decrease up to 40% in the evaporation rate can be expected for small spacings (i.e. interdroplet spacing of 2 diameters) when compared to isolated conditions. The only interaction experiments on droplets smaller than 2 mm in diameter, as those typically found in a fuel spray, were conducted by Sangiovanni and Labowsky[4]. Their results show that the classical “D2-law”, which states that the square of the diameter of an evaporating droplet decreases linearly with time, does not apply rigorously for interacting droplets. The increase in droplet lifetime due to interaction depends solely on droplet spacing and not on fuel type, droplet size, or ambient conditions. This paucity of experimental data is attributable to the difficulties in performing controlled experiments on free droplets of spray size, especially at high pressure. At these conditions, additional effects, such as gas solubility into the liquid, higher-order non-equilibrium effects, and ambient conditions near or above the critical point need to be taken into account. Their potential importance is so well recognised that numerous theoretical and experimental studies, aimed at improving the basic understanding of the physics, have been pursued[5–8]. Unfortunately, albeit several evaporation models have been proposed in recent years, which include higher-order non-equilibrium effects and more complicated equation of states, their full validation has been hampered by the absence of reliable experimental data. Aim of our research activities is to develop a new test rig for studying droplet vaporisation and interaction, at high-pressure/temperature conditions, through simplified and systematic experiments. To that aim, we have developed a diagnostic tool for determining the evaporation rate of single streams of monosized droplets by measuring simultaneously both the droplet size and speed. In this paper, we discuss in details the accuracy and reliability of this technique and, above all, its ability to accurately characterise vaporisation and interaction processes within droplet arrays.

2. Experimental setup

2.1. Droplet generator

In the experiments presented here, the droplets are produced by a vibrating-orifice droplet generator. Its main components are a piezoelectric ceramic and an orifice plate. Upon exciting the piezoelectric element with a suitable frequency, a monodisperse chain of uniformly spaced droplets is obtained. The regularity of the droplet stream is maintained over a certain distance along the path of the droplets, if no external disturbance and/or traverse flow are used to disperse the droplets. By an appropriate choice of the orifice diameter, the excitation frequency and the liquid feed rate, the initial size for the droplet can be adjusted very precisely. A detailed description of the droplet generator can be found in [9]. Fig. 1 shows a schematic view of the droplet generator, its peripherals, and the undisturbed droplet stream. The initial droplet temperature can be regulated by a heating/cooling device, and is controlled by a thermocouple. The droplets are falling vertically downwards along the generator axis and are immersed in stagnant air. The generator is mounted on a x–y translator, so that the droplet size can be measured at different axial positions.

2.2. Optical setup

The general layout of the setup follows the configuration originally proposed by Ferri[10] and is sketched in Fig. 2. As light source, a He–Ne laser is used. The laser power can be reduced up to a factor 106 by means of a λ/2 retardation plate and a Glan-Thompson polariser (GT). The latter is also used to set the polarisation of the electrical field in the horizontal direction. The beam is spatially filtered, expanded to a diameter of 9 mm by the system of lenses L1, L2 and then shined onto the sample. The power impinging on the sample is monitored by collecting the reflection from the beamsplitter (BS) onto the photodiode PD1. The light scattered by the sample is collected by lens L3 and forwarded on the CCD camera. Lens L4 conjugates the sensor plane with the focal plane of lens L3, therefore realising a one-to-one mapping between scattering angles and pixel positions with a magnification factor $M_{CCD} = 5.2$. A mirror, placed in the focal plane of lens L3, deviates the transmitted beam upward onto the photodiode PD2. The mirror is positioned in such a way...
that the focused transmitted beam hits its tip very close to the upper edge, allowing light scattered at very low angle to pass clear. This feature made it possible to use common CCDs as detectors, thus resulting in an increased angular resolution and sensitivity. During an experimental run, a number of standard precautions were observed in order to increase the accuracy of the measurements. In order to minimise the stray light falling onto the CCD, all optical elements were coated with an antireflection (AR) coating. Furthermore, care was taken in aligning the optics in such a way to prevent beam reflections from falling onto the CCD.

3. Low-angle light scattering technique

3.1. Droplet sizing

The droplet size history is measured by means of low-angle elastic light scattering (LAELS). The technique has been successfully applied by Ferri et al. [12] to the characterisation of large (0.7–80 μm) polystyrene spheres and consists in measuring the light scattered by an ensemble of particles at various angles in the forward direction. A typical diffraction pattern generated by the monodisperse droplet stream is shown in Fig. 3a. Here Young’s fringes, produced by the interference between the scattered wavefronts, are clearly visible. The images are processed by dividing the pixel matrix in concentric rings, centred on the optical axis, so that each ring corresponds to a specific scattering angle \( \theta \), as shown in Fig. 3b. For the present experimental configuration, we used 200 contiguous rings equally spaced and linearly scaled. By averaging the pixel intensity over the entire ring, we obtain the scattered angular intensity distribution \( I_{\text{exp}}(\theta) \). As customarily done, it is possible to express the scattered intensity as function of the wavevector \( \tilde{q} \), defined as the difference between \( \tilde{k} \) (scattered wavevector) and \( \tilde{k}_0 \) (incident wavevector), i.e. \( \tilde{q} = \tilde{k} - \tilde{k}_0 \) (see Fig. 3b for reference). The magnitude of \( \tilde{q} \), from now on indicated simply as \( q \), is related to the scattering angle by the relation

\[
q = \frac{4\pi n}{\lambda} \sin \left( \frac{\theta}{2} \right),
\]

where \( \lambda \) is the wavelength of light in vacuum and \( n \) the refractive index of the medium. When the sample is constituted by an ensemble of non-interacting droplets, the average scattered intensity is given by

\[
I_{\text{exp}}(q) = \int I_{\text{Mie}}(q, r) M(r)^{-1} W(r) \, dr,
\]

where \( I_{\text{Mie}} \) is the intensity scattered by a droplet of radius \( r \) at the wavevector \( q, M(r) = (4/3)\pi \rho r^3 \) the droplet mass (being \( \rho \) the density), and \( W(r) \) the mass of droplets with radii between \( r \) and \( (r + \, dr) \). Eq. (2) is a first-kind Fredholm integral equation in which \( I_{\text{exp}}(q) \) is provided by the experiment, \( I_{\text{Mie}}(q, r) M(r)^{-1} \) represents the kernel, computed according to Mie theory, and \( W(r) \) the unknown distribution to be retrieved. The inverse problem of reconstructing the function \( W(r) \) from the scattering data involves the inversion of Eq. (2). Because the information content in a set of scattering measurements is limited, inverse problem are usually ill posed and do not possess a unique solution. In literature, a copious number of algorithms have been proposed for the inversion of Eq. (2) [13–15]. Here, a modified version of the Chahine algorithm, introduced for the first time by Ferri et al., is used. A detailed description of the algorithm and its application to the inversion of low-angle scattering data can be found in [11,12].

It should be pointed out that once the size distribution \( W(r) \) has been retrieved, it is possible to calculate the two overall sample concentrations, specifically the volume fraction \( \phi \) and...
the droplet number density \( n_d \). The first is a non-dimensional quantity and is defined as the volume occupied by the liquid sample (i.e. the droplets) divided the total measurement volume \( V_{tot} \):

\[
\phi = \frac{1}{V_{tot}} \int W(r) \rho \, dr.
\] (3)

The second concentration \( n_d \) is defined as the total number of droplets per unit volume and has the dimensions of \( \text{cm}^{-3} \):

\[
\frac{1}{V_{tot}} \int W(r) \frac{M(r)}{\rho} \, dr.
\] (4)

Both concentrations are used in Section 4.2 to verify the consistency of all our predictions, obtained with the inversion method.

As an example of the potential of the method in correctly retrieving the droplet size distribution, a typical solution is discussed in some details. Fig. 4a shows the measured scattered intensities (filled circles) together with the intensities reconstructed on the basis of the recovered size distribution (solid line) as function of the wavevector amplitude \( q \). Note that the reconstructed intensities \( I(q) \) match quite well the experimental data \( I_{exp} (q) \), with deviations of only a few percents (r.m.s.). The droplet size distribution is shown in Fig. 4b as weight distribution. It is noteworthy noticing that the retrieved distribution is peaked in correspondence of the expected nominal radius, as expected for a monodisperse droplet stream. Similarly, the accuracy on the recovered average radius is of the order of a few percents. For the particular experiment shown in Fig. 4, the nominal \( (D_0 = 55.2 \, \mu m) \) and recovered \( (D_{\text{avg}} = 56.4 \, \mu m) \) diameters differ of roughly 2%.

### 3.2. Determination of droplet speed

As mentioned in the previous section, a system of Young’s fringes is superimposed to the diffraction pattern produced by the monodisperse droplet stream, as clearly visible in Fig. 3a. The fringes are a direct consequence of the particular geometrical configuration of the scatterers which, due to their regular spacing, can actually act as a grid. In our experiments, the interdroplet distance ranges from approximately 20 to 700 \( \mu m \) for droplets having diameters between 10 and 150 \( \mu m \). Fig. 5 shows the Young’s fringes extract from the diffraction pattern of Fig. 3a, in correspondence of the first maxima (i.e. 1st bright ring). These measurements were performed at a distance of 10 mm from the generator exit. The regularity in droplet spacing is represented by the constant spacing of the fringes. This regularity is maintained over a certain distance along the droplet path. Then the coherent structure of the stream tends to disappear due to external disturbances. If minor differences in the droplet spacing occur, Young’s fringes can still be observed. However, only the fringes close to the optical axis show a significant modulation. The fringe spacing is directly related to the interdroplet distance and, therefore, represents an indirect measure of the droplet speed. Indicating the fringe spacing with \( s_f \) and the droplet spacing with \( s_d \), it is easy to show that the two quantities are inversely proportional and are related to each other according to the following relation:

\[
s_d = \frac{\lambda f_l}{M_{\text{CCD}} s_f},
\] (5)

where \( \lambda \) is the wavelength of light, \( f_l \) the focal distance of lens \( L_3 \), and \( M_{\text{CCD}} \) the magnification factor of the optical system \( (M_{\text{CCD}} = 5.2) \). Recalling that the droplets reach the probe volume at a frequency \( f_G \), which corresponds to the excitation frequency of the generator, the droplet speed can...
then be calculated from
\[ v_{\text{drop}} = \frac{s}{d} f_G. \] (6)

Anders et al. [16] showed that there is no appreciable difference in accuracy between this technique and more standard methods, such as laser Doppler anemometry (LDA). As a matter of fact, the accuracy of this technique is limited by the spatial resolution of the employed CCD. In the present case, the CCD pixel size is equal to 6.7 × 6.7 μm, which leads to an inaccuracy in the determination of the average droplet speed of roughly 2%. For the experiments considered here, the droplet speed remains essentially constant along the droplet path, with typical average values ranging from 6 to 20 m/s. Note that, for a given fluid, orifice size and initial fluid temperature, the droplet speed depends solely on the pressure difference applied to the droplet generator. Fig. 6 shows the droplet velocity \( v_{\text{drop}} \) as function of the driving pressure difference \( \Delta p \) for two different fluids (heptane and isopropanol, respectively). The curves are derived empirically from liquid flow rate measurements and by applying a volumetric balance to the liquid column. The average droplet speed, derived from the fringe spacing method, is also plotted on the same graph (filled symbols). As it can be immediately seen, the measured velocities fit the empirical curves reasonably well. Discrepancies are due to evaporation effects which lead to an underestimation of the liquid flow rate.

4. Results

This section examines critically the ability of the proposed technique to correctly characterise droplet evaporation and interaction phenomena. To that aim, the performances of the LAELS technique are evaluated from different perspectives. First, we check the accuracy of the instrument in measuring the angular intensity distributions. The latter represents a key prerequisite for performing accurate measurements. This aspect is discussed in Section 4.1. Then, we ascertain the capability of the LAELS technique in performing reliable particle sizing. Finally we investigate its suitability for studying complex interaction phenomena between vaporising droplets embedded in a convective flow. The latter aspect is discussed in Section 4.2.

4.1. Overall performances

A detailed analysis of the instrument properties concerning its accuracy, sensitivity, dynamic range, and linearity can be found in [10]. Here, we discuss briefly its overall performances. In particular, we show the ability of the instrument to measure angular intensity distributions over the entire angular range. By selecting pinholes of different sizes (from 5 to 200 μm in diameter), we measure the diffraction pattern by placing the pinhole in the cell plane and centring it on the optical axis. According to Fraunhofer diffraction theory, the intensity angular distribution, diffracted by a pinhole of radius \( r \), depends solely on the product \( qr \) and its amplitude increases as \( r^4 \). Therefore, by properly rescaling the diffraction patterns of different pinholes, it is possible to make all data collapse to a single curve. In Fig. 7a, the diffracted intensities of 5, 30, 50, and 100 μm pinholes, normalised by the factor \( r^4 \) (i.e. the corresponding nominal radius), are plotted as function of \( qr \). The data match with high accuracy the angular position of the minima and maxima, but are somewhat less accurate on the intensity scale. This is most probably due to the \( r^4 \) normalisation, which strongly amplifies even small percentage error on pinhole size. Fig. 7b shows the asymptotic value \( I(q = 0) \) as function of pinhole size on a log–log scale. The data exhibit a power law behaviour with
4.2. Droplet evaporation

The accuracy of the LAELS technique in correctly recovering the droplet size is checked by comparing the experimental predictions, measured at the generator outlet, with the nominal value derived from theoretical considerations. Under the hypothesis of regular disintegration and negligible evaporation, the nominal droplet radius can be calculated from the conservation of mass:

\[ R_{\text{drop}} = \left[ \frac{3V_L}{4\pi f_G} \right]^{1/3}, \]  

\[ (7) \]

where \( V_L \) is the flow rate through the orifice. 1 Fig. 8 shows the theoretical curves, derived from Eq. (7) for three different flow rates, and the experimentally determined droplet radius as function of the generator excitation frequency \( f_G \). The recovered droplet size matches quite well the theoretical curve throughout the whole experimental range, with deviation of a few percents (r.m.s.), thus providing justified confidence on the reliability and accuracy of the LAELS technique. The suitability of the technique for studying complex interaction phenomena between vaporising droplets is investigated by assessing the influence of droplet spacing on the vaporisation process and checking the consistency between experimental and theoretical predictions. To that aim, two different experimental situations are considered. In the first one (named Case A), the evaporation rates of two heptane droplet chains are compared. The two droplet arrays differ in the initial droplet temperature \( T_d \), but are characterised by the same interdroplet distance \( \Delta \). The latter is defined as the distance between the centres of two neighbouring droplets. In the second case (named case B), the opposite situation is analysed. We consider two isopropanol droplet chains at the same temperature, but with different droplet spacing. All relevant parameters for each experiment are summarised in Table 1.

Fig. 9a shows the droplet diameter as function of time for the heptane experiments (Case A). As it can be immediately seen, the evaporation rate \( (dr/dt) \) is basically the same for both experiments, which is in clear contradiction with theoretical predictions for an isolated droplet. In this latter case, one would expect a higher evaporation rate for Exp. No. 5

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1 Note that \( V_L \) represents the generator nominal flow rate (i.e. devoid of evaporation effects) and is derived independently from liquid flow rate measurements.
Table 1

<table>
<thead>
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<th>Exp. no.</th>
<th>P_{amb} (bar)</th>
<th>T_{amb} (°C)</th>
<th>Δp (bar)</th>
<th>f (kHz)</th>
<th>T_d (°C)</th>
<th>Δ/D_b</th>
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<tr>
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<tr>
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</table>

The orifice diameter is 30 μm. The above-mentioned parameters represent in the order: experimental run, ambient pressure and temperature, pressure difference at the generator outlet, generator excitation frequency, initial droplet temperature, non-dimensional droplet spacing (D_b being the initial droplet diameter).

due to the higher degree of superheating (T_d = 73.5 °C). This discrepancy can be only explained in light of droplet interaction effects, which strongly hamper the evaporation process. Sangiovanni and Labowsky [4] presented an extensive compilation of data (2 alcohols, 2 pure hydrocarbons, 3 binary hydrocarbon mixtures, 2 fuel oil, 100–300 μm droplets, 4 ambient conditions) and demonstrated that the increase in droplet lifetime depends solely on droplet spacing and not on fuel type, droplet size, or ambient conditions. The results shown in Fig. 9a represent an additional confirmation of their findings. Fig. 9b shows the droplet diameter as function of time for the isopropanol experiments (Case B). Note that, for droplet spacing of roughly 2 diameters, the vaporisation process is radically quenched. Leiroz and Rangel [1]
studied numerically the interaction effects during droplet-stream combustion and derived a correlation between the total droplet mass vaporisation rate and the interdroplet spacing. Following their correlation, an increase in vaporisation rate of 14% is predicted as the interdroplet distance increases from 2.5 to 4.1 diameters. From the result shown in Fig. 9b, we obtained an increase of 20% in vaporisation rate, which is in reasonable agreement with the theoretical predictions of Leiroz and Rangel. As additional check, we compare the generator nominal flow rate $V_L$ with the corresponding value derived from the inversion method. In fact, recalling the definitions of liquid volume fraction $\phi$ and droplet number density $n_d$ (see Eqs. (3) and (4) for reference), the liquid flow rate can be expressed as

$$V_L = \frac{\phi n_d}{\Delta}$$

(8)

The results are shown in Fig. 9c and d for all experiments considered here. The temporal decrease in liquid flow rate is self-explanatory and a direct result of the vaporisation process. Obviously, in correspondence of the generator outlet, the derived value for $V_L$ has to match the generator nominal flow rate. As already mentioned, the latter is obtained separately from empirical correlations, expressing the nominal flow rate as function of the driving pressure difference applied at the generator. For all experiments, the difference between recovered and nominal liquid flow rate amounts to ±4%, thus providing an indirect global verification of the correctness and consistency of the analysis.

5. Conclusions

Low-angle elastic light scattering (LAELS) can be used to study droplet interaction effects in vaporising arrays in some detail. By exploiting the interference pattern generated by the scattered wavefronts, it is possible to measure simultaneously both droplet size and speed. This provides an indirect way to obtain quantitative information on the evaporation rate of the droplet stream. Although LAELS cannot rival more innovative techniques, such as partial wave resonance[17], in the study droplet interaction effects in vaporising streams have also been successfully studied. Our preliminary results are consistent with the findings from previous numerical/theoretical works. Even though we have used considerably smaller droplets (droplet diameter below 100 µm) and only pure substances, the effect of droplet spacing on the reduction of the vaporisation rate does not differ substantially from those observed on much bigger droplets and hydrocarbons mixtures.

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