Combustion and Flame 000 (2016) 1–15

Contents lists available at ScienceDirect

Combustion and Flame

journal homepage: www.elsevier.com/locate/combustflame

Time-resolved detection of diffusion limited temperature gradients inside single isolated burning droplets using Rainbow Refractometry

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ARTICLE INFO

Article history:
Received 2 July 2015
Revised 10 March 2016
Accepted 11 March 2016
Available online xxx

Keywords:
Droplet combustion
Temperature measurement
Temperature gradients
Rainbow Refractometry

ABSTRACT

Temporal and spatial resolved temperature measurements inside vaporizing and combusting droplets are needed to validate and improve numerical and simple approximated models. Elastic light scattering techniques such as Rainbow Refractometry are capable of probing unsuspended droplets below 100 μm. The high sensitivity of the rainbow structure to refractive index changes enabled the detection and resolution of time-varying temperature gradients inside combusting droplets. This was accomplished through comparison of different droplet combustion models as well as direct inversion using a Simulated Annealing algorithm. Models incorporating diffusion limited heat transport showed very good agreement with the experimental data. Accordingly, the direct inverted results showed also reasonable agreement and the feasibility of the direct inversion of the temperature. Finally, an analytical solution of the heat conduction equation for a constant droplet size with exponential increasing surface temperature described the process for half of the droplet burning time very well, enabling the incorporation in spray combustion models to effectively account for temperature variation within combusting droplets.

1. Introduction

A wide variety of today's industrial processes such as liquid fuel combustion in engines, comprise atomization and vaporization of liquid droplets. The enlarged surface area enhances heat and mass transfer between the gas and the liquid phase, which in turn determines droplet heating and vaporization [1]. In order to predict and design reliable systems, fundamental knowledge of the governing transport mechanisms is needed. However, the strong interaction between droplets within actual sprays hinders the experimental access and analysis, when simultaneous heat and mass transfer as well as chemical reactions occur.

To surmount these difficulties, idealized systems such as single burning droplet in an unbounded oxidizing atmosphere were established in the early 1950s [2–5] and extensively developed during the past decades [1,6–8]. These studies have been vital for the understanding of spray combustion processes. Researchers found that heat and mass transport rather than chemical reaction is the rate controlling mechanism during vaporization/combustion of droplets (termed d^2 law). Transient effects such as droplet heating were accounted for deviations between experiments and theory [9–11]. Wise and Ablow [12], Williams [13], Waldman [14] and Crespo and Liñan [15] considered a finite rate of liquid phase heating during combustion. They concluded that for a minimum heat of reaction, the assumption of a constant boiling point temperature is a very good approximation for burning liquid droplets. However, if a large fraction of the heat that reaches the droplet is used to raise the droplet temperature, rather than being used for vaporization, the temperature inside the liquid can be significantly lower than the boiling point temperature, resulting in temperature gradients. Similar conclusions were also stated by Law [16], who investigated the influence of a time-varying but uniform temperature (rapid-mixing model) inside burning droplets. He observed an initial unsteadiness of droplet radius and flame size due to changing liquid temperature. However, the rapid-mixing model is difficult to transfer to sprays, since internal convection due to shear-induced Hill vortices inside the droplets is not sufficient to attain a throughout mixing [17], leading to liquid heating models such as diffusion limited [18] and convective enhanced heat transport [19]. Although both models cannot be applied directly to spray calculations due to the considerable computational

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http://dx.doi.org/10.1016/j.combustflame.2016.03.007
0010-2180/

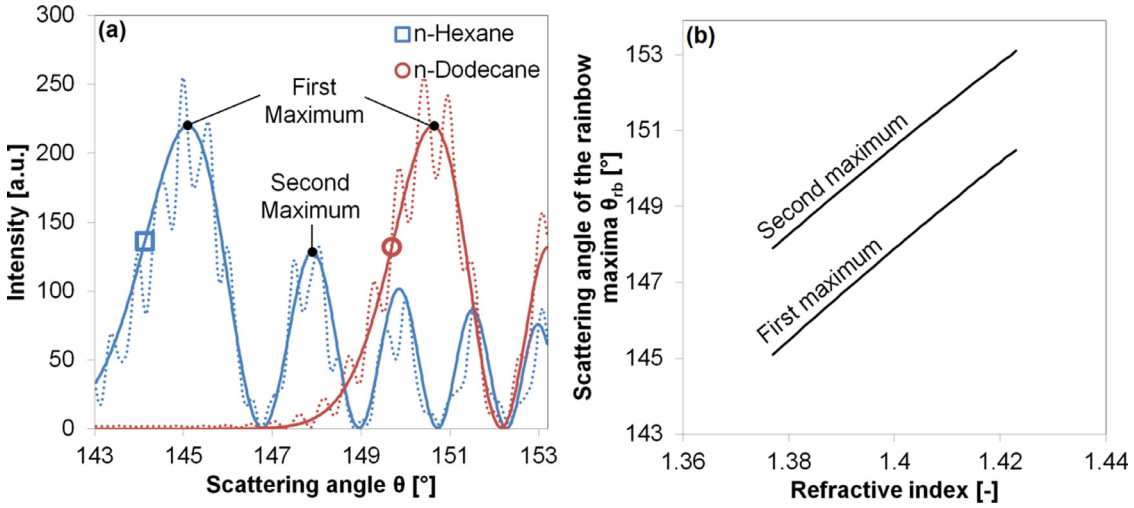

Fig. 1. (a) Rainbow scattering pattern simulated for 100 μm diameter *n*-hexane (□) and *n*-dodecane (○) droplets illuminated by a 532 nm plane wave. The dotted lines correspond to the calculation of the complete scattering pattern (LMT), while solid lines represent the low-frequency rainbow structure. (b) Correlation between the angular position of the first two rainbow maxima and the refractive index for an uniform 100 μm diameter liquid droplet. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

time needed, they are useful to validate and improve approximated models [20,21].

Temporal and spatial temperature measurements inside burning droplets are challenging, making a validation of liquid heating models difficult. Moreover, temperature measurements are often carried out with thermocouples [22,23], resulting in additional heating and catalytic effects and therefore altering the vaporization/combustion rates [24,25]. In contrast, laser-based measurement techniques offer the opportunity of non-invasive temperature probing. Elastic light scattering techniques can be applied to a wide variety of processes [26]. Since most liquid droplets in sprays are spherical due to the acting surface tension force [27], the analysis of the light scattering measurements can be readily described by the Lorenz–Mie-Theory (LMT) [28]. One example is the Rainbow Refractometry (RRF), where the shift of the so-called rainbow in the light scattering pattern of a spherical droplet is used for determination of the refractive index of the liquid. Since the refractive index changes with temperature for pure liquids, the temperature can be determined. This technique was first applied by Roth et al., Massoli et al., Sankar et al. and van Beeck et al. [29–32], in order to determine spatially uniform droplet temperatures. However, measuring a spatially distributed refractive index caused by gradients inside the droplet remains a challenge. Especially combustion with high vaporization rates leads to temperature differences between the droplet surface and its core that can sustain throughout the entire droplet lifetime [18]. Schneider et al. [33] and Kai et al. [34] showed in light scattering investigations a shift of the rainbow in case of temperature gradients in droplets. Although the temperature gradients inside liquid droplets have been studied [35–39], to the best of our knowledge experimental evidence and inversion of the diffusion limited heat transport inside burning droplets has not been reported until now.

Here, we report the detection and evaluation of transient temperature gradients due to diffusion limited heat transport inside micron sized droplets with RRF. We will present the limitation of the Rainbow Refractometry for gradients, followed by a brief representation of the LMT for multilayered spheres, which is used to evaluate the scattering patterns of burning droplets and extraction of the spatial and time-resolved temperature gradients. This will be accomplished both by comparison of simulation results with different combustion models and the direct inversion with Simulated Annealing (SA).

1.1. Limitations of Rainbow Refractometry

A uniform temperature throughout a droplet results in linear rainbow pattern shifts to larger or smaller scattering angles with increasing or decreasing refractive index (increasing or decreasing temperature), respectively. Figure 1a shows scattering patterns for 100 μm diameter *n*-hexane (blue) and *n*-dodecane (red) droplets simulated with the LMT (dotted) as well as with the second order refraction (solid) of the Debye decomposition algorithm [40]. For a fast data analysis, the low-frequency rainbow pattern (solid) is often preferred, because the angular positions of the maxima is almost linear correlated with the refractive index of the liquid droplets (Fig. 1b). The functional relation between the temperature and the refractive index is linear for most liquids, making temperature measurements possible through detecting the linear shift of the rainbow.

However, for temperature gradients inside the droplets, a simple rainbow analysis is often misleading. Figure 2a shows the simulated angles of the first two rainbow maxima for a 100 μm diameter *n*-hexadecane droplet having a time variant temperature gradient (Fig. 2b). The temperature profiles were calculated according to the analytical solution for the time-varying heat conduction of a sphere with constant surface temperature and radius [41]:

$$\frac{T(r,t)-T_0}{T_s-T_0} = 1 + \frac{d_p}{\pi\,\Delta r}\sum_{n=1}^{\infty}\frac{(-1)^n}{n}\sin\left(\frac{n\pi\,\Delta r}{0.5d_p}\right)exp\left(\frac{-an^2\pi^2\Delta t}{(0.5d_p)^2}\right) \quad (1)$$

where T, d_p, a, Δr, Δt and n are the temperature, droplet diameter, thermal diffusivity, incremental radius, time and running number of the infinite series, respectively. The subscripts 0 and s designate initial and surface conditions, respectively. The thermal diffusivity was taken according to Table 1. For clarity, only a few time steps were selected to illustrate the temperature gradients. Simply applying the linear correlation from Fig. 1b, which holds for a uniform temperature without gradients, would result in an initial wrong decreasing temperature and subsequent increasing temperature as the rainbow scattering angle in Fig. 2a suggests. In reality, the droplet is heating up instead (Fig. 2b).

The angular shifts of the rainbow maxima towards higher scattering angles are caused by curved trajectories of the rainbow rays [34] for radial temperature gradients (Fig. 2c). If the temperature

Please cite this article as: C.D. Rosebrock et al., Time-resolved detection of diffusion limited temperature gradients inside single isolated burning droplets using Rainbow Refractometry, Combustion and Flame (2016), http://dx.doi.org/10.1016/j.combustflame.2016.03.007