

Molecular tagging thermometry for transient temperature mapping within a water droplet

De Huang and Hui Hu*

Aerospace Engineering Department, Iowa State University, Ames, Iowa 50011, USA

*Corresponding author: huhui@iastate.edu

Received August 28, 2007; revised October 22, 2007; accepted November 16, 2007;
posted November 21, 2007 (Doc. ID 86931); published December 10, 2007

We present the progress made in developing a molecular tagging thermometry (MTT) technique for achieving spatially and temporally resolved temperature measurements within a small water droplet over a solid surface. For MTT measurement, a pulsed laser is used to tag phosphorescent 1-BrNp·M β -CD·ROH molecules premixed with water. Long-lived laser-induced phosphorescence is imaged at two successive times after the same laser excitation pulse. The temperature measurement is achieved by taking advantage of the temperature dependence of the phosphorescence lifetime, which is estimated from the intensity ratio of the acquired phosphorescence image pair. The measured transient temperature distributions can be used to quantify the unsteady heat transfer process inside convectively cooled water droplets over smooth or rough surfaces. © 2007 Optical Society of America

OCIS codes: 120.6780, 110.6820, 160.2540.

Aircraft icing is widely recognized as a significant hazard to aircraft operations. Advancing the technology for safe and efficient aircraft operation in atmospheric icing conditions requires a better understanding of the microphysical phenomena associated with the accretion and growth of ice. To elucidate underlying physics associated with aircraft icing, experimental techniques capable of providing detailed measurements to quantify important ice growth physical processes such as the dynamics of water droplets and the unsteady heat transfer process inside water droplets over smooth or rough surfaces are highly desirable. In this Letter, we report progress made in our recent effort to develop and implement a molecular tagging thermometry (MTT) technique to achieve temporally and spatially resolved temperature measurements within small water droplets to quantify the unsteady heat transfer process inside convectively cooled water droplets over smooth or rough surfaces for aircraft icing studies.

It is well known that both fluorescence and phosphorescence are molecular photoluminescence phenomena. Compared with fluorescence, which typically has a lifetime of the order of nanoseconds, phosphorescence can last as long as microseconds, even minutes. Since emission intensity is a function of the temperature for some substances, both fluorescence and phosphorescence of tracer molecules may be used for temperature measurements. Laser-induced fluorescence (LIF) techniques have been widely used for temperature measurements of liquid droplets for combustion applications [1–3]. Laser-induced phosphorescence (LIP) techniques have also been suggested recently to conduct temperature measurements of in-flight or levitated liquid droplets [4,5]. Compared with LIF techniques, the relatively long lifetime of LIP could be used to prevent interference from scattered or reflected light, and any fluorescence from other substances (such as from solid surfaces) that are present in the measurement area by simply putting a small time delay between the la-

ser excitation pulse and the starting time for phosphorescence image acquisitions. Furthermore, LIP was found to be three to four times more sensitive to temperature variation compared with LIF [4–7], which is favorable for accurate measurements of small temperature differences within small liquid droplets.

The MTT technique described at here is a LIP-based technique, which can be considered an extension of the molecular tagging velocimetry and thermometry technique developed by Hu and Koochesfahani [6]. For MTT measurement, a pulsed laser is used to tag a phosphor (e.g., phosphorescent dye) premixed in the working fluid. The long-lived LIP emission is imaged at two successive times after the same laser excitation pulse. The LIP emission lifetime distribution is estimated from the intensity ratio of the acquired phosphorescence image pair. The temperature distribution within a small water droplet can be derived by taking advantage of the temperature dependence of the phosphorescence lifetime. It should be noted that both the present MTT measurement and the work of Omrane *et al.* [4,5] are based on a similar idea of achieving temperature measurement by taking advantage of the temperature dependence of the phosphorescence lifetime. The work of Omrane *et al.* [4,5] is only a single-point feasibility study using photomultiplier-based instrumentation. The present work, to our knowledge, is the first planar temperature field measurement to achieve temporally and spatially resolved temperature measurement within a small water droplet based on direct imaging of phosphorescence lifetime with a conventional image-detecting CCD camera.

The technical basis of the MTT measurements is given briefly here. According to quantum theory, the intensity of phosphorescence emission decays exponentially. As described in [7], for a dilute solution and unsaturated laser excitation, the phosphorescence signal (S) collected by using a gated imaging detector with integration starting at a delay time t_o after the

laser pulse and a gate period of δt can be given by

$$S = AI_i C \varepsilon \Phi_p (1 - e^{-\delta t / \tau}) e^{-t_o / \tau}, \quad (1)$$

where A is a parameter representing the detection collection efficiency, I_i is the local incident laser intensity, C is the concentration of the phosphorescent dye (the tagged molecular tracer), ε is the absorption coefficient, and Φ_p is the phosphorescence quantum efficiency. The emission lifetime τ refers to the time at which the intensity drops to 37% (i.e., $1/e$) of the initial intensity.

In general, the absorption coefficient ε , quantum yield Φ_p , and the emission lifetime τ are temperature dependent, resulting in a temperature-dependent phosphorescence signal (S). Thus, in principle, the collected phosphorescence signal (S) may be used to measure fluid temperature if the incident laser intensity and the concentration of the phosphorescent dye remain constant (or are known) in the region of interest. It should be noted that the collected phosphorescence signal (S) is also the function of incident laser intensity (I_i) and the concentration of the phosphorescent dye (C). Therefore, the spatial and temporal variations of the incident laser intensity and the non-uniformity of the phosphorescent dye in the region of interest would have to be corrected separately to derive quantitative temperature data from the acquired phosphorescence images. In practice, however, it is very difficult, if not impossible, to ensure a nonvarying incident laser intensity distribution, especially for unsteady thermal phenomena with a varying index of refraction. This may cause significant error in the temperature measurements. To overcome this problem, a lifetime-based thermometry [6] was developed to eliminate the effects of incident laser intensity and concentration of phosphorescent dye on temperature measurements.

The lifetime-based thermometry works as follows: as illustrated in Fig. 1, laser-induced phosphorescence emission is interrogated at two successive times after the same laser excitation pulse. The first image is detected at time $t = t_o$ after laser excitation for a gate period δt to accumulate the phosphorescence intensity S_1 , while the second image is detected at time $t = t_o + \Delta t$ for the same gate period to accumulate the phosphorescence intensity S_2 . It is easily shown, by using Eq. (1), that the ratio of these two phosphorescence signals (R) is given by

$$R = S_2 / S_1 = e^{-\Delta t / \tau}. \quad (2)$$

In other words, the intensity ratio of the two successive phosphorescence images (R) is a function of only the phosphorescence lifetime τ and the time delay Δt between the image pair, which is a controllable parameter. This ratiometric approach eliminates the effects of any temporal and spatial variations in the incident laser intensity and nonuniformity of the dye concentration (e.g., due to bleaching). For a given molecular tracer and fixed Δt value, Eq. (2) defines a unique relation between phosphorescence intensity ratio R and fluid temperature T , which can be used for thermometry.

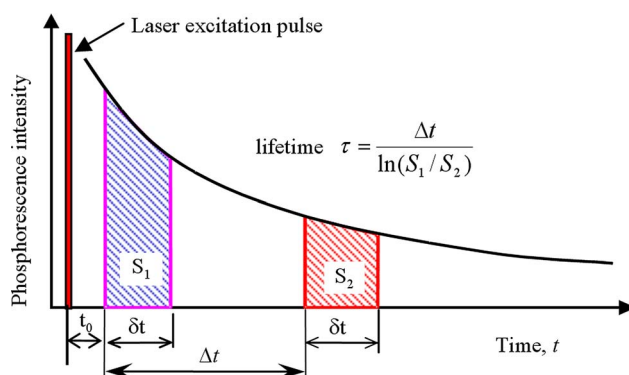


Fig. 1. (Color online) Timing chart for MTT measurement.

A demonstration experiment was conducted to implement the MTT technique described above. In the present study, the phosphorescent triplex (1-BrNp·M β -CD·ROH) was used as the molecular tracer for the MTT measurements. The phosphorescent triplex (1-BrNp·M β -CD·ROH) is actually the mixture compound of three different chemicals [8], which are lumophores (indicated collectively by 1-BrNp), maltosyl- β -cyclodextrin (indicated by M β -CD), and alcohols (indicated collectively by ROH). Figure 2 shows the measured phosphorescence lifetime of 1-BrNp·M β -CD·ROH molecules versus temperature with the laser excitation wavelength of 266 nm (quadrupled wavelength of the Nd:YAG laser). It can be seen clearly that the phosphorescence lifetime varies significantly with increasing temperature, decreasing from about 3.4 to 1.5 ms as the temperature changes from 22°C to 36°C. The relative temperature sensitivity of the phosphorescence lifetime is about 5.7% per degree Celsius, which is much higher than those of fluorescent dyes [1–3] (such as Rhodamine B, which is only about 2.0% per degree Celsius).

A water droplet with a size of about 10 μ m and initial temperature of 32.5°C was placed on a test plate. The temperature of the test plate was set as the same as that of ambient air, which was 23.5°C. The water droplet was convectively cooled after it was placed on the test plate. A laser sheet (1.0 mm in thickness) from a pulsed Nd:YAG laser at a quadrupled wavelength of 266 nm was used to tag the premixed

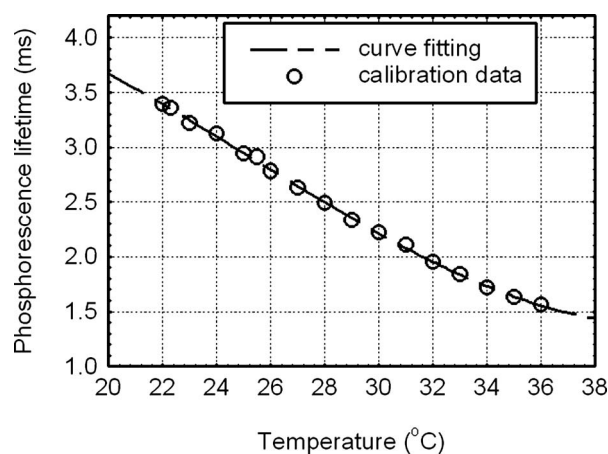


Fig. 2. Phosphorescence lifetime versus temperature.

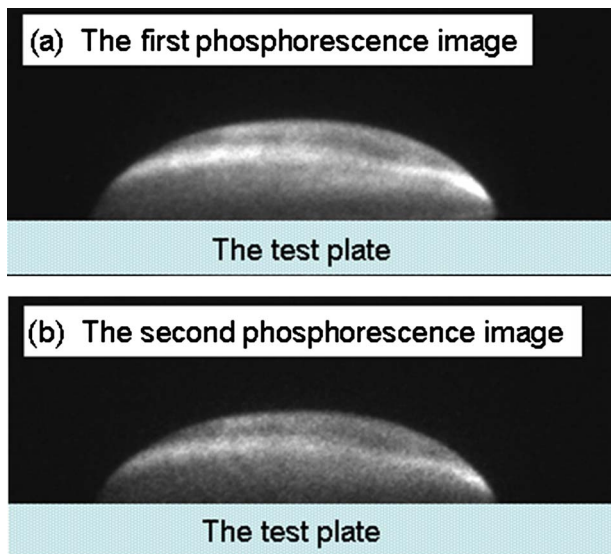


Fig. 3. (Color online) Typical phosphorescence image pair.

1-BrNp·M β -CD·ROH molecules along the mid plane of the water droplet. A 12 bit gated intensified CCD camera (PCO DiCam-Pro) with a fast decay phosphor (P46) was used to capture the phosphorescence emission. The camera was operated in the dual-frame mode, where two full-frame images of phosphorescence were acquired in quick succession after the same laser excitation pulse. Figure 3 shows a typical phosphorescence image pair, which was acquired about 45 s after the water droplet was placed on the test plate. The first image was acquired at 0.4 ms after the laser pulse, and the second image at 4.9 ms after the same laser pulse with the exposure time of 1.5 ms for the two image acquisitions. Since the time delays between the laser excitation pulse and the phosphorescence image acquisitions eliminated scattered or reflected light and any fluorescence from other substances (such as from solid surfaces) effectively, the phosphorescence images of the water droplet are quite clean. Figure 4 shows the instantaneous temperature distribution within the water droplet derived from the phosphorescent image pair shown in Fig. 3. Because of the relatively high temperature sensitivity of the present MTT technique, the small temperature difference within the water droplet could be revealed clearly from the MTT measurement. As it is expected, the surface temperature of the water droplet was found to decrease rapidly to

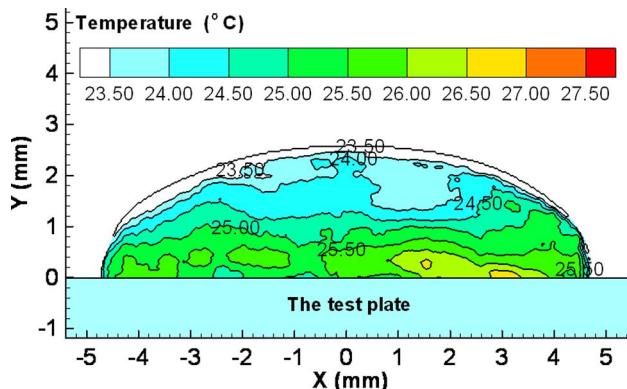


Fig. 4. (Color online) Instantaneous temperature distribution.

ambient air temperature. The bottom temperature of the water droplet was found to be slightly higher than ambient air temperature owing to the low thermal conductivity of the plastic test plate. Based on the time sequence of the measured transient temperature distributions within the water droplet, the unsteady heat transfer process inside the convectively cooled water droplets can be revealed quantitatively. Such information is highly desirable to improve our understanding of microphysical phenomena associated with the accretion and growth of ice for aircraft icing studies.

The authors thank Manoochehr Koochesfahani of Michigan State University for providing chemicals used for the present study. The support of National Science Foundation CAREER program under award number CTS-0545918 is gratefully acknowledged.

References

1. Q. Lu and A. Melton, *AIAA J.* **38**, 95 (2000).
2. S. Escobar, J. E. Gonzalez, and L. A. Rivera, *Exp. Heat Transfer* **14**, 119 (2001).
3. M. Wolff, A. Delconte, F. Schmidt, P. Gucher, and F. Lemoine, *Meas. Sci. Technol.* **18**, 697 (2007).
4. A. Omrane, G. Juhlin, F. Ossler, and M. Alden, *Appl. Opt.* **43**, 3523 (2004).
5. A. Omrane, S. Santesson, M. Alden, and S. Nilsson, *Lab. Chip* **4**, 287 (2004).
6. H. Hu and M. Koochesfahani, *Meas. Sci. Technol.* **17**, 1269 (2006).
7. H. Hu, C. Lum, and M. Koochesfahani, *Exp. Fluids* **40**, 753 (2006).
8. A. Ponce, P. A. Wong, J. J. Way, and D. G. Nocera, *J. Phys. Chem.* **97**, 11137 (1993).