Laser-based temperature imaging close to surfaces with toluene and NO-LIF

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Abstract. Two novel techniques based on Laser-Induced Fluorescence (LIF) were applied to measure gas-phase temperature distributions in boundary layers close to wall surfaces. Single-line toluene-LIF thermometry was used to image temperature in a nitrogen gas flow above a heated wall. The nitrogen gas flow was doped with evaporated toluene. When excited at 266 nm, the toluene LIF-signal shows an exponential dependence on temperature. This behavior was used to calculate absolute temperatures from LIF images after calibration at known conditions. The second technique, multi-line NO-LIF thermometry was applied to image temperature in the quenching boundary layer close to a metal wall located on a flat flame burner. A small amount of nitric oxide was mixed into the air/methane mixture. NO molecules were excited in the A-X (0,0)-band at 225 nm. NO-LIF excitation spectra were acquired by tuning the excimer laser wavelength and recording the NO LIF-signal with an ICCD camera. Absolute temperatures were calculated for every pixel by fitting simulated excitation spectra to the experimental data. Temperature distributions close to the wall surface were measured at two different flow-rate conditions. A high nominal spatial resolution of 0.016 mm/pixel in direction perpendicular to the wall was reached. Wall surface temperatures were recorded simultaneously by embedded thermocouples and compared with gas-phase temperature near the wall surface.

1. Introduction

A detailed understanding of transport phenomena in boundary layers is essential for the quantitative description of reactive flows, as appear in internal combustion engines, power plants, etc. Within the recent years, laser diagnostics have been frequently applied to combustion analysis, e.g. in internal combustion engines [1,2]. However, most of these techniques have been used to measure ‘bulk flow/combustion’. Diagnostics for wall/flow interactions in boundary layers have not yet been developed sufficiently.
Temperature measurement in boundary layers seemed to be more important than velocity and species concentration, for the following reasons. First, the temperature distribution close to surfaces in a cylinder of reciprocating engines affects directly flame propagation, knocking, and quenching. These phenomena dominate engine performance and exhaust emissions. Second, temperature information is very useful for LIF measurement of species, such as OH and fuel compounds. The temperature dependence of their LIF signals hampers the application of LIF techniques to conditions with spatial temperature gradients. If the temperature distribution is known, however, the LIF signal can be corrected for its temperature dependence.

Gas phase temperatures can be measured based on Rayleigh scattering off gas molecules [3]. In the vicinity of surfaces scattering generates a prohibitive background. This technique is therefore not applicable unless filtered Rayleigh techniques [4] are used. Another option is to take advantage of the temperature dependence of the LIF signal from molecular tracers. Such methods have the advantage that the signal from the excited molecules is shifted with regard to the excitation wavelength. This allows the discrimination against scattered light with appropriate filters.

Popular tracers are 3-pentanone and acetone [5,6]. Recently, toluene was discovered as promising tracer for thermometry. All these substances show broad temperature dependent LIF spectra [7].

In single-line techniques (one laser wavelength and one detection band) the change of the total fluorescence intensity is measured and temperature is calculated after calibration. They can only be applied if the tracer concentration is invariant in the observed field. Two-line techniques (i.e. either two excitation wavelengths or two detection bands) and multi-line techniques gather information about either the emission or the excitation spectrum of the tracer. From its temperature dependent shape temperatures can be calculated. These methods allow measurements also in systems with inhomogeneous distribution of the tracer.

In this work, two novel techniques based on LIF were chosen and applied to measure gas-phase temperature distributions in boundary layers. First, single-line toluene-LIF thermometry was applied to visualize a temperature gradient in a nitrogen gas flow above a heated wall. Second, multi-line NO-LIF thermometry was applied to image temperature in the quenching boundary layer close to a metal wall located on a flat flame burner.

### 2. Temperature imaging with toluene-LIF thermometry

#### 2.1. Single-line toluene thermometry

Fluorescence quantum yield and temperature sensitivity are important criteria for the choice of a tracer for thermometry. Here toluene has a clear advantage compared to the widely used tracers 3-pentanone or acetone (a factor ~500 higher quantum yield). Together with the measurements published by Luong et al. [8], this work is the first application of toluene as a tracer for thermometry.

The technique is based on measurements of the temperature dependant absorption cross section $\sigma_{abs}$ and fluorescence quantum yield $\phi$ of toluene in nitrogen bath gas [7]. They were conducted with 266 nm excitation wavelength in a temperature regime from 300 K to 930 K. When calculating the LIF-signal per volume $S_f$ at constant pressure and constant tracer mole fraction $x_{tr}$, the change of number density with temperature has to be accounted for as well:

$$S_f \propto x_{tr} \frac{1}{T} \sigma_{abs}(\lambda, T) \phi_f(\lambda, T)$$

The resulting plot shows a logarithmic dependence of total LIF signal on temperature and is displayed in figure 1. An exponential fit to this plot gives the following equation:

$$T = T_{ref} - 107.53 \ln \left( \frac{S_f(T)}{S_f(T_{ref})} \right)$$

with $S_f(T_{ref})$ being the fluorescence signal at the known temperature $T_{ref}$.

This equation allows the calculation of the temperature in a nitrogen/toluene gas flow based on the change of the LIF signal in respect to a reference signal. This is subject to the condition that the concentration of toluene is kept constant and that the distribution of laser energy does not change and is far from saturation limits. The absorption of laser light along its path through a gas depends on the
absorption cross-section and the number density of the absorbing molecules. For toluene the product of both is normalized and plotted against temperature in figure 2. Between 300 K and 500 K more light will be absorbed with increasing temperature. Therefore the distribution of laser energy along the light sheet is temperature dependant. To reduce this error source it is important to keep the toluene concentration low.

Figure 1. Temperature-dependence of toluene LIF.

Figure 2. Product of absorption cross section and number density as a function of temperature. Normalized to the value at 300 K.

2.2. Experimental

A sketch of the experimental setup is shown in figure 3. A 6 l/min nitrogen gas flow was seeded with 18 g/h toluene using an evaporator and mixer and then further diluted in a stream of 24 l/min nitrogen. Gas and liquid flows were controlled by Bronkhorst flow controllers. The gas mixture was introduced into an optical accessible flow channel (5×60 mm² cross section, 258 mm length) to form a laminar flow of 1.7 m/s average velocity with a toluene concentration of 2300 ppm. The bottom wall of the flow channel was made of aluminum (3 mm thickness) with a wall heater unit (Watlow NM180X50, 180 mm length) attached to it. A thermocouple was embedded into the aluminum wall 204 mm behind the inlet to monitor the wall temperature at this location.

A quadrupled Nd:YAG laser (Thomson CSF, 5000 Series) provided laser light at 266 nm. The beam was expanded in vertical direction by a pair of cylindrical lenses (f₁ = −40 mm, f₂ = 80 mm). A section was cut out by an aperture and formed to a light sheet using another cylindrical lens (f = 1000 mm). Via a periscope stage the light sheet was aligned under a small angle of incidence onto the heated bottom wall. In order to avoid saturation effects the laser energy was reduced by changing the Q-switch timing. The laser beam had an energy of ~0.5 mJ per pulse at the entrance window of the flow channel.

Images were acquired using a LaVision NanoStar ICCD camera with a Nikon UV lens (f = 105 mm, fₐ = 4.5) and averaged over 50 laser shots. One pixel geometrically represents ~50 μm in the observed area. A Schott WG 280 filter reduced elastically scattered laser light.

The remaining background from elastically scattered laser light (less than 1% signal strength compared to the toluene LIF) was recorded in pure nitrogen. With toluene seeded into the flow, LIF images were acquired at room temperature and at two different wall temperatures. These were measured by the thermocouple to be 409 K and 447 K.

The background images were subtracted from all LIF images. Then, temperature distributions over the heated wall were calculated pixel-by-pixel according to equation (1) with the image taken at room temperature serving as reference (Tᵣₑᵥ = 296 K).
2.3. Results and discussion
The temperature distribution in a laminar channel flow with an average velocity of 1.7 m/s over a heated surface was imaged at two different wall temperatures. The results are shown in figure 4. The heated surface was considered to be at the position of highest temperature and the images were cut off along this line. At three positions, at the left and right edge of the observed area and at the location of the thermocouple, temperature profiles were extracted from the images. They show a temperature boundary layer with an almost linear temperature gradient towards the wall. In the middle of the imaged area gas-phase temperatures at the wall are about 15 K below the value measured inside the wall by the thermocouple.

The peak temperature decreases by ~16 K in flow direction which is due to an inhomogeneous heat flux from the heater unit. The absorption through the imaged area was calculated to increase by 2% if the wall is heated, resulting in an error of max. 2 K. Thus a change in the laser energy distribution due to this effect should play a minor role. Further experiments will include monitoring the temperature in the heated wall at several locations.

The standard deviation of temperatures was calculated from the root mean square of the LIF images to be ~10 K (2.5%) for all pixels. This originates from oscillations of the laser energy and can significantly be reduced by recording the laser energy shot-per-shot.
3. Temperature imaging with multi-line NO-LIF thermometry

3.1. Multi-line NO-LIF thermometry

Multi-line NO-LIF temperature imaging was recently introduced by Bessler and Schulz [9]. Accuracy and versatility were significantly improved compared to conventional two-line techniques. The multi-line technique was applied to temperature measurements in sooting flames [9], high-pressure flames [10] as well as in spray flames [11].

2D-LIF excitation scans were performed over a spectral range in the NO A-X(0,0) band while acquiring two-dimensional intensity maps with an ICCD camera. For each individual pixel, LIF intensities were extracted versus excitation wavelengths, yielding experimental excitation spectra. A spectral simulation code [12] was then used to fit simulated spectra to the experimental spectra for each pixel using a non-linear least-square fitting method. The fitting technique yields absolute temperature without the necessity of calibration.

Figure 6. Experimental setup: overall figure (left) and the inside of the burner housing (right).

3.2. Experimental

A schematic of the optical setup is shown in figure 6. The beam from a tunable, narrow-band ($\Delta \nu = 0.6$ cm$^{-1}$) KrF excimer laser (Lambda Physik, EMG 150 TMSC) was used to pump a H$_2$-Raman-cell. Behind the cell a Pellin-Broca prism and a slit aperture separated the first anti-Stokes frequency from the fundamental and the other Raman-shifted frequencies. A cylindrical lens formed the laser beam into a horizontal light sheet. Laser pulse energies were approximately 1 mJ, which were detected by a photodiode on a single-shot basis to correct the LIF signal for laser energy fluctuations.

Inside the burner housing, premixed methane/air flat flames, shown in figure 7, were stabilized on a flat-flame burner [13,14]. The flame folder is a honeycomb disk with 805 holes, each 1 mm in diameter. It was made of brass, which has high thermal conductivity, to uniform the temperature distribution. The surface was coated with nickel plating to prevent corrosion due to the seeded nitric oxide. The burner had a co-flow nozzle to stabilize the flat flame. The flow rate of the co-flow was adjusted to the velocity of the unburned mixture flow.

0.5 mm above the flame folder, an edged metal wall was located. It was made of stainless steel, with a cooling-water channel inside. The cooling-water temperature was kept to 60°C. The object surface of the wall was polished. Four thermocouples are embedded close to the surface.

The horizontal laser sheet passed over the flame-holder. The cross section of the light sheet was ~20 x 0.2 mm$^2$ at the burner center. It had a small depression angle to the metal wall surface, $\theta$ in
figure 6, to prevent blind spot close to the surface. For the same reason, a 45° mirror was shifted slightly towards the CCD camera to have a small depression angle to the wall surface. Therefore, the field of view included not only fluorescence images on the horizontal cross-section, but also reflection images on the wall surface.

Excitation scans were performed by recording 2D-images at a given laser wavelength and then tuning the laser to the next wavelength. The scans covered the 44407-44418 cm$^{-1}$ range with 0.2 cm$^{-1}$ spectral resolution. At each wavelength, LIF signals of 50 images were averaged to improve signal/noise ratio. A band pass filter separated the signal light from elastically scattered light and other interference. LIF signals were focused (Halle, $f = 200$ mm achromatic UV lens) onto a LaVision Flowmaster ICCD camera with 100 ns exposure time. The camera chip was binned x8 on the y-axis to enhance the signal level, yielding raw images of 160 × 1024 pixels. A high nominal spatial resolution of 0.016 mm/pixel in direction perpendicular to the wall (x-axis) was achieved.

Figure 7. Structure of the flat flame burner and the metal wall.  
Figure 8. An example of measured temperature images and procedure of data evaluation.

3.3. Data evaluation

Figure 8 shows the procedure of data evaluation. The multi-line spectra fitting was performed by the LIFSim program package [12]. Simultaneous fit of background signal and LIF strength makes this technique robust against both broadband interference and laser and signal attenuation. It yields absolute, quantitative temperature measurements without the necessity of external calibration. However, while accurate temperatures were evaluated at most pixels on the 2D images, the spectra fitting did not converge at some pixels, as indicated by the black areas in the images. Therefore, these error pixels were corrected by interpolation. Then, the center parts of the 2D(\textit{xy})-images, located between the pair of thermo-couples at the same height, were extracted and averaged on the y-axis. These one-dimensional temperature distributions on the $x$-axis at different heights were combined to generate a 2D-image in the $xz$-plane.

The detection of the wall position is important to achieve high spatial accuracy. The authors considered the position of the lowest temperature as the wall position, because the temperature must be the lowest on the wall. The dispersion of the lowest temperature position was ± 3 pixels (± 0.05 mm). Finally, the 2D-(\textit{xz})-images were trimmed at wall position.
3.4. Results and discussion

Temperature distributions close to the wall surface were measured at two different flow-rate conditions; 7.5 l/min (Case-1) and 15 l/min (Case-2), yielding mean flow velocities of 6 m/s and 12 m/s respectively. The equivalence ratio of the lean flames was 0.9 in both cases. The concentrations of seeded nitric oxide were adjusted to 5000 ppm and 4000 ppm, respectively, to yield sufficient LIF signal levels.

Figure 9 shows the resulting temperature distribution. The pairs of narrow black lines show the position and thickness of laser light sheets. The temperature distributions between these pairs of lines were interpolated. In each case, the flat flame was bent upward by the metal wall, and the left edge of the flame was lifted to approximately 2 mm above the flame holder. From this height, the quenching layer thickness increased as it ascended. The layer thickness at $z = 13$ mm in case-1 is around 6 mm, which reduces by 50% in case-2.

![Figure 9](image)

**Figure 9.** Temperature distribution images of methane/air flat flames close to a metal wall with equivalence ratio 0.9: (left) 7.5 l/min, 6 m/sec, (right) 15 l/min, 12 m/sec.

![Figure 10](image)

**Figure 10.** 3D-graphs of gas-phase temperature distribution of methane/air flat flames and solid-phase temperature distribution on the wall surface: (left) 7.5 l/min, 6 m/sec, (right) 15 l/min, 12 m/sec.

To compare gas-phase and solid-phase temperatures on the wall, 3D graphs were plotted, shown in Figure 10. The solid lines show the temperature distributions on the wall ($x = 0$). Each small circle shows the average of temperature measured by a pair of thermocouples at the same height. Temperature distributions between thermocouples at different heights were estimated, shown as dotted
lines. Wall temperatures measured by the upper thermocouples were close to the coolant water temperature (333 K) in both cases, because of the short distance between the thermocouples and the water channel. The distance between the dotted line and the solid line shows a temperature drop from gas-phase to solid-phase on the wall surface. This temperature drop increases in case-2 with higher flow rate.

Note that the thermocouple measurements do not show the true wall-temperature, because the contact points of the thermocouples were located inside the wall, ~0.2 mm away from the wall surface, resulting in a temperature drop within the solid-phase.

4. Summary and future plan
Two novel techniques based on LIF were applied to measure gas-phase temperature distributions in boundary layers close to wall surfaces;-
- Single-line toluene-LIF thermometry was used to image temperature in a nitrogen gas flow above a heated wall.
- Multi-line NO-LIF thermometry was applied to image gas-phase temperature in the quenching boundary layer close to a metal wall located on a flat flame burner.
Results of multi-line NO-LIF thermometry will be used to correct LIF signals of species, such as OH, O₂, etc., for their temperature dependence. These measurements will be conducted in the same quenching boundary layers. The authors are planning to apply these techniques to in-cylinder temperature measurement in internal combustion engines.

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References
[1] N.M. Laurendeau: Prog. Energy Combust. Sci. 14, 147 (1988)
[2] E.W. Rothe, P. Anresen: Appl. Optics 36, 3971 (1997)
[3] R.W. Dibble, R.E. Hollenbach: Proc. Combust. Inst. 18, 1489 (1981)
[4] R.B. Miles, W.R. Lempert, J.N. Forkey: AIAA paper 91-0357 (1991)
[5] F. Großmann, P.B. Monkhouse, M. Ridder, V. Sick, J. Wolfrum: Appl. Phys. B 62, 249 (1996)
[6] M.C. Thurber, F. Grisch, R.K. Hanson: Opt. Lett. 22, 251 (1997)
[7] W. Koban, J.D. Koch, R.K. Hanson, C. Schulz Phys. Chem. Chem. Phys. 6, 2940 (2004)
[8] M. Luong, W. Koban, C. Schulz: Novel strategies for imaging temperature distribution using Toluene LIF (ICOLAD, London, 2005)
[9] W.G. Bessler, C. Schulz Appl. Phys. B 78, 519 (2004)
[10] W.G. Bessler, C. Schulz, T. Lee, D.I. Shin, M. Hofmann, J.B. Jeffries, J. Wolfrum, R.K. Hanson: Appl. Phys. B 75, 97 (2002)
[11] H. Kronemayer, I. Düwel, C. Schulz: Temperature imaging in spray flames (European Combustion Meeting (ECM), Louvain-la-Neuve, 2005)
[12] W.G. Bessler, C. Schulz, V. Sick, J.W. Daily: A versatile modeling tool for nitric oxide LIF spectra (http://www.pci.uni-heidelberg.de/pci/lifsim) (3rd Joint meeting of the US sections of The Combustion Institute, Chicago, 2003, PI1)
[13] K. Wakai, et al: Journal of gas turbine society (Japan) 7, 33 (1979 [http://www.gifu-u.ac.jp/~wakailab/index_e.html])
[14] M. Saffman: Combustion and flame 55, 141 (1984)