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# Quantitative planar laser-induced fluorescence of naphthalenes as fuel tracers

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## Abstract

In planar laser-induced fluorescence (PLIF), naphthalenes are commonly used as tracers for diesel-like fuels and fuel surrogates. Previously, such a technique was used to visualize the air–fuel mixing in a meso-scale ( $\sim 10 \text{ cm}^3$  device volume) burner prototype running on dodecane as a single-component JP-8 surrogate. For quantification of this technique, the influence of temperature, oxygen quenching, and the presence of the fuel surrogate was investigated through spectrally resolved LIF of naphthalenes in a heated jet as well as through broadband imaging. For excitation at 266 nm, naphthalene and 1-methylnaphthalene show very similar fluorescence behavior. Fluorescence is not influenced by the presence of dodecane but is strongly quenched by oxygen. Due to this strong oxygen quenching, fluorescence intensity was found to be linear with equivalence ratio at all temperatures. Both increased temperature and increased oxygen concentrations lead to a spectral broadening and red-shift. This effect is minor for oxygen addition and saturates at about 10% oxygen, but is significant for temperature increase and continuous throughout the temperature range investigated. This red-shift was exploited to simultaneously image temperature and equivalence ratio. A dichroic beamsplitter was used to obtain a long-wavelength and a short-wavelength image whose ratio represents temperature. Calibrations were performed on the heated jet, and subsequently this two-color PLIF method was applied to the mesoscale burner. After averaging the images from 20 processed single-shot measurements, the equivalence ratio and temperature images obtained had signal/noise ratios of 25 and 11, respectively. Temperature correction of the equivalence ratio resulted in excessive noise. Likely inaccuracies in temperature were identified in very lean regions where the signal is low and sensitive to the background corrections employed.

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## 1. Introduction

A mesoscale burner for the chemical-to-thermal conversion of  $\sim 10 \text{ ml/h}$  of JP-8 to 100 W thermal power is being developed at Yale. This burner

will be interfaced to a thermal-to-electric conversion scheme, which is being developed elsewhere, to yield  $\sim 20 \text{ W}$  of electric power, therefore providing an alternative to conventional batteries with the potential advantages of high energy density and quick refueling. Detailed descriptions of concept, prototypes, and performance of the burner can be found in [1]. The general concept is one of lean, premixed catalytic combustion. Fuel–air mixing is achieved by multiple electrospays

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dispersing the fuel into preheated air. Mixing takes place in a laminar, but not necessarily steady, environment in a volume of about  $10 \text{ cm}^3$ . The mixture is then converted in a stack of several catalytically coated grids. There is no “flame” in the sense of a homogeneous gas-phase reaction. Due to the lean conditions, reaction takes place at relatively low temperatures around 1200 K. Currently, the air is preheated to  $\sim 470 \text{ K}$  to simulate the anticipated effects of a recuperation scheme.

One of the central optimization problems in this device is the minimization of space necessary for fuel–air mixing. Previously, planar laser-induced fluorescence (PLIF) of a fuel tracer has been used by our group to image the mixing process in a modified, optically accessible version of the prototype burner. To obtain qualitative images of the instantaneous and time-averaged equivalence ratio, dodecane as a single-component fuel surrogate was doped with 5% 1-methylnaphthalene (1-MN). Fluorescence was excited at 266 nm and detected broadband by an intensified CCD camera in the standard PLIF arrangement featuring a pulsed UV-laser sheet and perpendicular imaging train. A more detailed description of these imaging experiments and results was presented in [2]. For several aromatic tracers, it has been shown that under suitable conditions the fluorescence signal can be interpreted as the local equivalence ratio [3–7]. For linear fluorescence (signal proportional to laser energy) of a non-reacting tracer with oxygen as the only significant quencher, and when quenching is fast compared to other de-excitation paths, the general time-integrated proportionality for the tracer fluorescence intensity  $S_{\text{n}}^{\text{T}}$

$$S_{\text{n}}^{\text{T}} \sim n_{\text{T}} \frac{k_{\text{n}}^{\text{T}}}{k_{\text{f}}^{\text{T}} + k_{\text{q}}^{\text{T}} n_{\text{O}_2}} \quad \text{simplifies to } S_{\text{n}}^{\text{T}} \sim \frac{n_{\text{T}}}{n_{\text{O}_2}} \sim \phi, \quad (1)$$

where  $n_{\text{T}}$  and  $n_{\text{O}_2}$  are the number densities of tracer and oxygen, respectively.  $k_{\text{n}}^{\text{T}}$ ,  $k_{\text{f}}^{\text{T}}$ , and  $k_{\text{q}}^{\text{T}}$  are the rate coefficients for spontaneous emission, collision-independent de-excitation, and quenching by oxygen, respectively. Proportionality of fluorescence to equivalence ratio is therefore a good assumption if the Stern–Volmer coefficient  $k_{\text{SV}} = k_{\text{q}}/k_{\text{f}}$  is large. Toluene and tetrahydrothiophene, for example, show a highly linear dependence of fluorescence on equivalence ratio  $\phi$  [6,7].

The above is valid for a constant temperature. To quantify PLIF in an inhomogeneous temperature field, the temperature dependence of the fluorescence process needs to be known, and the local temperature needs to be obtained. Melton’s group has used time-based gating for imaging of temperature and fuel concentration in an oxygen-free environment with naphthalene as a tracer [8] as well as for imaging fuel and oxygen concentrations in an isothermal environment using the tracer flu-

oranthene [9]. The objective of the work presented here was to investigate parameters influencing the naphthalene fluorescence, establish a method for simultaneously measuring temperature and equivalence ratio under the given conditions, and test this method in the actual burner prototype.

## 2. Spectrally resolved measurements

### 2.1. Experimental details

For obtaining fluorescence spectra, a heated jet with known and controllable temperature and gas composition was used. The experimental set-up is shown in Fig. 1. Fuel and, if applicable, tracer are metered by a syringe pump and atomized in a flow-assisted nozzle in a flow of heated nitrogen. The mixture is heated further, and oxygen is added 20 cm upstream of the exit nozzle, which is about 8 mm in diameter. A closed-loop controller regulates the exit temperature on the input from a K-type thermocouple just before the nozzle exit. All parts of the flow system are heavily insulated and heated to avoid condensation.

Excitation in the probe volume just above the nozzle was provided by focusing the fourth harmonic of an Nd:YAG laser at 266 nm with a  $f = 500 \text{ mm}$  spherical lens. The resulting tracer fluorescence was detected by an imaging system with specifications as given in Fig. 1. Between 25 and 200 shots were averaged, and spectra were extracted by spatially integrating over 2 mm in the central portion of the jet with the beam waist located 1.5 mm above the nozzle exit. By PLIF visualization, it was verified that throughout the probe volume the jet is homogeneous in composition. The temperature profile above the nozzle was measured with a small-diameter thermocouple for various nominal temperatures. The temperature deviation throughout the probe volume was found to be less than 4 K. Pulse energies for these spectral measurements were less than  $25 \mu\text{J}/\text{shot}$ , which is somewhat more than the saturation limit, that is, the fluorescence is not necessarily linear in

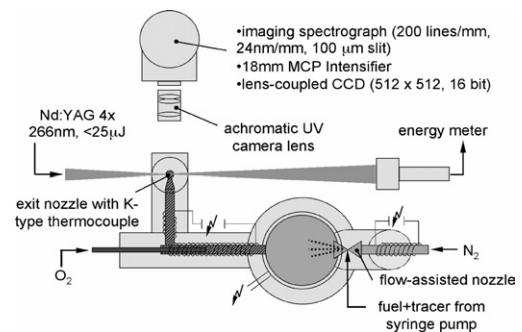


Fig. 1. Experimental arrangement for spectrally resolved PLIF in a heated fuel– $\text{N}_2$ – $\text{O}_2$  jet.

laser energy. However, variation of laser energy did not lead to a change in the shape of the spectrum. In a time-resolved study, the authors of [5] found the fluorescence decay times of naphthalene to be insensitive to laser pulse energy.

A typical flow rate was 5.0 L/min of nitrogen (at standard temperature and pressure) with various flow rates of oxygen added. Mixtures of 79%  $N_2$  and 21%  $O_2$  will be referred to as “air.”

Measurements were performed with both fuel and tracer injected into the gas flow, as well as with the tracer only. Fuel flow rates are given in terms of the equivalence ratio with respect to a final gas composition containing 21%  $O_2$ , even when this is not the actual composition. Where the tracer was used without fuel, the equivalence ratio is given as if the fuel were added, too. In this case, the fuel (dodecane) was replaced by an equal gaseous flow rate of  $N_2$  since, as will be shown below, both dodecane and  $N_2$  are essentially inert with respect to the fluorescence process. For 6.3 L/min air and  $\Phi = 1$  for example, the liquid fuel flow rate is 40.02 ml/h, equivalent to a 1-MN flow rate of 1.52 ml/h at 5% (by weight) tracer concentration. The boiling point of dodecane is 498 K, those of 1-MN and naphthalene are 515 and 490 K, respectively. 1-MN is liquid, naphthalene is solid at room temperature.

## 2.2. Spectrally resolved results

Figure 2 shows the fluorescence spectra of 5% naphthalene in dodecane for various partial pressures of oxygen at 500 K. As has been found by Ossler et al. [5] in their extensive study of the fluorescence of polycyclic aromatic hydrocarbons (PAHs), an addition of oxygen leads to a broadening of the spectrum and a red-shift of the falling edge of about 4 nm. This effect saturates at about 10% oxygen, which is the maximum concentration investigated by the above authors. Further addition of oxygen does not yield any changes in spectral shape. This spectral change should therefore not be of importance in most combustion-relevant

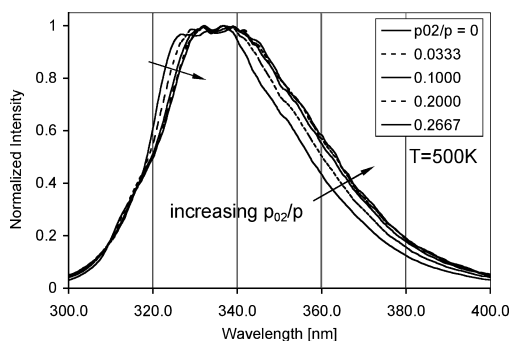


Fig. 2. Emission spectra of 5% naphthalene/95% dodecane for different partial pressures of oxygen.

situations, except for very high rates of exhaust-gas recirculation. The spectral results for 1-MN in dodecane and 1-MN by itself (not shown) are identical. The spectral shape of 1-MN fluorescence is nearly identical to that of naphthalene, though for each oxygen concentration there is an additional red-shift of about 4 nm as compared to naphthalene at the same oxygen concentration.

This similarity and results obtained from the imaging configuration described below indicate that LIF from 1-MN is equivalent to that of naphthalene and independent of the presence of the fuel it is meant to trace. All experiments involving temperatures higher than 500 K were conducted using 1-MN only. Dodecane was found to auto-ignite at a wide range of equivalence ratios at temperatures above 600 K, resulting first in temperature fluctuations as indicated by the exit thermocouple, and then in a weak bluish flame. These phenomena were not observed with 1-MN only, neither at the low concentrations typical for use as a tracer, nor in higher concentrations. A typical residence time from the point of oxygen addition to the nozzle exit was 80 ms. An initial attempt to characterize the chemical stability of 1-MN as a function of temperature and residence time was inconclusive, and the possibility that there is pyrolysis of the tracer should be considered for each application.

Figure 3 shows the effect of temperature variation on the 1-MN fluorescence spectrum. From Fig. 3A, it can be seen that the total fluorescence emission in air stays roughly constant up to 600 K and then decreases monotonically. The normalized emission spectra, shown in Fig. 3B, show a broadening and a red-shift of the falling edge. This spectral change was also found to be the case for 1-MN in nitrogen, (not shown), again confirming the findings of Ossler et al. [5], who observed similar behavior for naphthalene in nitrogen. In air, the red-shift at half maximum is about 16.4 nm, while in nitrogen it is 21.1 nm. However, in nitrogen the total signal monotonically decreases for all temperatures, presumably due to the lack of oxygen quenching, whose influence is also temperature dependent, as will be shown below.

## 3. Imaging measurements

For ketonic tracers, a temperature-induced shift of the absorption spectrum has been used to obtain simultaneous two-dimensional measurements of temperature and fuel concentration [10–14]. In support of quantitative PLIF with these tracers, spectral temperature dependencies, particularly of the laser absorption, are now well documented. Temperature has been measured as the ratio of the fluorescence for two different excitation wavelengths. Conversely, for naphthalene, for which the extinction coefficient at 266 nm

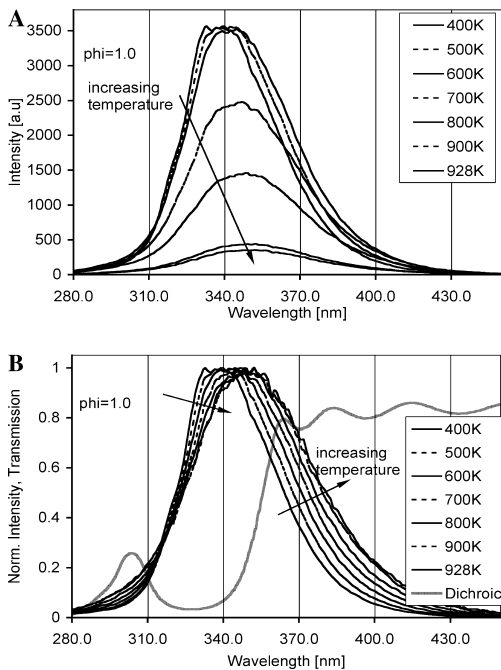


Fig. 3. Emission spectra of 1-MN in air for different temperatures: (A) absolute values and (B) normalized with respect to the maximum. In (B), the transmission curve of the 350-DCLP dichroic combined with a WG-295 is shown in grey.

was previously found to be insensitive to temperature [5], it should be possible to obtain the local temperature from a suitable ratio of different parts of the fluorescence spectrum, with one single excitation wavelength. For imaging, the chosen strategy was to use a dichroic beamsplitter to separate the spectrum into “blue” (short wavelength) and “red” (long wavelength) portions. This results in a “blue” and “red” image, whose pixel-by-pixel ratio will reflect the spectral red-shift and can be related to temperature through suitable calibration. In Fig. 3, the transmission curve of the chosen dichroic (350-DCLP, Chroma Technology), combined with a standard UV-longpass filter (WG-295) to reject elastic scattering, is shown superimposed on the normalized 1-MN emission spectra. Wavelengths shorter than the cut-on at 360 nm are reflected. Wavelengths below roughly 300 nm are rejected from both cameras through the absorption in the longpass filter.

### 3.1. Imaging configuration

The complete optical arrangement for imaging experiments in the burner is shown in Fig. 4. Excitation was provided by the same laser as above (Nd:YAG, 4th harmonic at 266 nm). The laser sheet was formed to cover almost the entire height of the burner and was carefully clipped and

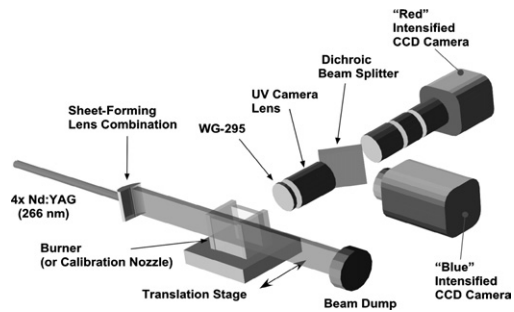


Fig. 4. Experimental arrangement for two-color PLIF.

aligned with the catalyst grid on top of the burner (not shown in image) to be able to extend the useful imaging region to very close proximity of the catalyst. The sheet thickness was estimated at 0.5 mm. An achromatic UV camera lens (105 mm, f/4.5) with filter and dichroic as described above imaged the fluorescence onto two intensified CCD cameras. The “red” (transmission) camera was lens-coupled to an 18 mm intensifier. Its full resolution of  $2016 \times 2016$  pixels was binned to either  $504 \times 504$  for multi-shot images or to  $168 \times 168$  for single-shot measurements. The “blue” (reflection) camera was fiber-coupled to an 18 mm intensifier and was used at its full resolution with  $575 \times 380$  pixels readout for multi-shot images and binned to  $115 \times 68$  for single shots. The resulting spatial resolution of both cameras was matched for single-shot images with a nominal pixel volume of  $320 \times 320 \times 500 \mu\text{m}$ . For multi-shot exposures, the resolution was better according to the reduced binning of each camera. The burner could be translated through the sheet to image in different locations. At each location, 20 single shots and 1 exposure of 50 shots were taken. Catalyst luminosity was suppressed through the short gating time of  $1 \mu\text{s}$ . The total dimensions of the mixing chamber were  $30 \times 30 \times 25 \text{ mm}$  (width  $\times$  depth  $\times$  height). In each image, a region extending to within about 0.5 mm of both side windows, to 0.5 mm below the catalyst grid, and 2 mm above the fuel nozzles was evaluated. Therefore, the images covered about 29 mm in width and 22.5 mm in height.

For signal normalization and flat-field correction, the intake of the burner could be supplied with a uniform air/tracer mixture from the same delivery system as used for the nozzle experiments. These calibration images were taken at the lowest temperature and highest seeding rate for which condensation of 1-MN in the delivery system and the nozzle or burner could be avoided. This was at 348 K and a 1-MN flow rate of 0.2 ml/h injected into 6.33 L/min of air, yielding a tracer concentration corresponding to an equivalence ratio of 0.333 for 1% 1-MN doped into dodecane. This condition then also provided a normalization for calibration purposes, as it could be reproduced

in both the insulated and heated nozzle, and the less well-controllable burner.

For imaging in the burner, dodecane with 1.5% 1-MN was used, which should be low enough to yield negligible absorption throughout the beam path, despite the high absorption coefficient of 1-MN. Laser energy was about 0.4 mJ/shot, which was verified to be in the linear regime.

### 3.2. Imaging results

In a first step, the imaging experiment was used with the jet nozzle to quantify some observations that had been made earlier in the spectral measurements but could not be quantified correctly because these measurements had been performed outside of the linear regime. For this purpose and for calibration measurements described below, the fluorescence signal was spatially integrated in a region of uniform temperature and gas composition in the potential core of the jet. Measurements represented in Fig. 5 were made with the dichroic removed such that the total spectral emission was imaged onto the “red” (transmission) camera.

Figure 5 shows the oxygen quenching of the tracers naphthalene and 1-MN, each at 1% in dodecane (DD), as well as 1-MN without dodecane at 500 K. Flow rates for all of the above correspond to an overall dodecane–air equivalence ratio  $\Phi = 0.75$ , with the flow rate of pure 1-MN adjusted to yield the mass flow of 1% 1-MN in dodecane at that equivalence ratio and the “missing” dodecane replaced by nitrogen. Also shown are results for 1-MN at  $T = 800$  K. Results are shown as a Stern–Volmer plot in which the fluorescence intensity for a given partial pressure of oxygen is normalized by the fluorescence without oxygen (only nitrogen as a carrier gas). A straight line therefore indicates that indeed oxygen is the major quencher, and the slope of that line gives the Stern–Volmer coefficient  $k_{SV}$ . It can be seen that the three tracer/fuel combi-

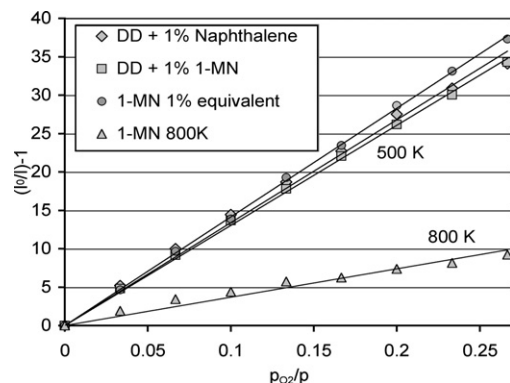


Fig. 5. Stern–Volmer plot for different naphthalenes with and without fuel at 500 K, and for 1-MN at 800 K. (See text for a discussion of normalization of the axes.)

nations yield straight lines with similar slopes of 1340, 1300, and 1410  $\text{MPa}^{-1}$  for naphthalene in DD, 1-MN in DD, and 1-MN only, respectively. This variation is within the error of measurement, for which the slope of the line is estimated to be about 10%. Therefore at least at this temperature (500 K), both naphthalene and 1-MN behave very similarly with respect to oxygen quenching, are not quenched by the fuel, and it is possible to perform experiments without the fuel surrogate dodecane. The Stern–Volmer coefficient significantly decreases at high temperatures and is only 369  $\text{MPa}^{-1}$  at 800 K. This has to be kept in mind since the proportionality of fluorescence and equivalence ratio depends on effective oxygen quenching.

With the heated jet still in place but imaging geometry identical that to be used in the burner (with the beam splitter separating the two wavelength regions), calibration measurements for temperature and equivalence ratio were obtained by recording the “red” and “blue” fluorescence throughout a practical range of these parameters. Fig. 6 shows the ratio “red/blue” as a function of temperature for different equivalence ratios. Through the flat-field correction that was applied to each “red” and “blue” image, this ratio is unity for the temperature at which the flat-field images were taken (348 K). To be able to apply the calibration in pixel-by-pixel calculations, a second order polynomial was fit to the averages over the different equivalence ratios. The variation of the “red/blue” ratio at each temperature gives an estimate of the uncertainty of the measurement ( $\leq 10\%$ ), since this ratio should be identical for all equivalence ratios and depend only on temperature.

Figure 7A shows the “red” fluorescence as a function of equivalence ratio for different temperatures. Through the flat-field correction the measurements are normalized to yield unity for  $\Phi = 1$  and  $T = 348$  K. Greater precision in this normalization could be obtained if it were possible to extend flat-field (i.e., base-line) measurements to

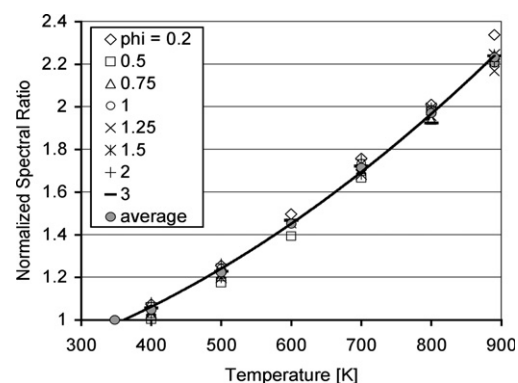


Fig. 6. Temperature calibration curve (quadratic fit to the series “average”) as obtained from two-color imaging of the heated jet.

higher equivalence ratios (as for the higher temperatures), but this is difficult if condensation is to be avoided. All data points are scattered reasonably tightly around their respective linear fit, indicating that at least for the moderate equivalence ratios considered here fluorescence is indeed linear in equivalence ratio for all temperatures. Separately, linearity up to an equivalence ratio of 10 was confirmed for 500 K, both for dodecane with 1-MN and for 1-MN only. A “correction factor” relating the normalized “red” fluorescence to the true equivalence ratio is then given by the inverse of the slope for each temperature. This factor is plotted as a function of temperature in Fig. 7B. As could be expected from the spectral measurements, with increasing temperature the “red” signal first increases (the correction factor decreases) because the overall fluorescence is nearly constant for moderate temperatures and the spectrum shifts to the red, but then decreases (the correction factor increases) because the sharp decrease in total signal is not compensated by the continuing red-shift. A cubic polynomial was fit to the data to facilitate interpolation. The point representing reference conditions is at 348 K and 1.00, which is not quite

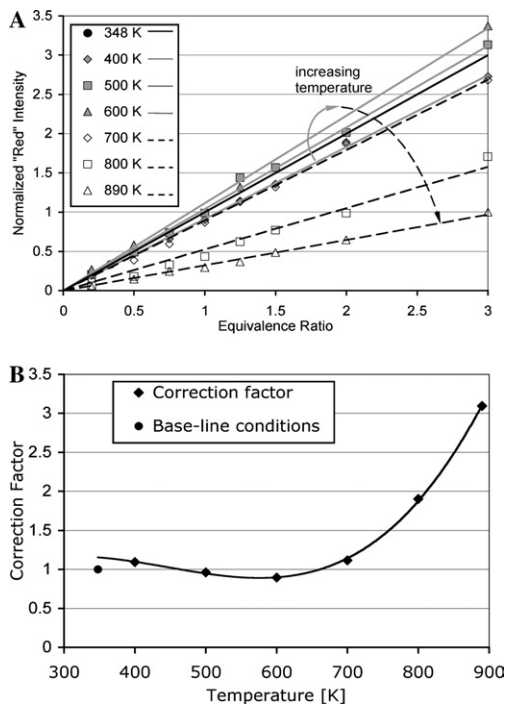


Fig. 7. (A) Equivalence ratio calibration curve. Values are normalized such that  $\Phi = 1.0$  at 348 K yields unity slope (shown in solid black). Slopes successively increase for 400–600 K (grey lines) and then decrease from 700 to 890 K (dashed lines). (B) Resulting correction factor for the “red” image. The base condition for obtaining flat-field correction images at  $\Phi = 0.333$  and  $T = 348$  K is shown as a full circle, but not included in the cubic fit.

in continuation of the data obtained from the line slopes towards lower temperatures. This discrepancy reflects the inaccuracy associated with referencing to only one equivalence ratio. The underlying measurement error is probably associated with difficulties in accounting for the luminous background in the fluorescence images, a problem that is expected to increase in the actual burner.

Figure 8 shows processed images obtained by operating and scanning the burner, as described in *Experimental details*. The flow rates were 13 ml/h of dodecane and 3.6 L/min of air, yielding an overall equivalence ratio of 0.565. The electrosprays were operated at 9 kV with respect to the grounded catalyst grid. This leads to unsteady structures in the flow, which improve mixing, as shown in [2]. Images shown are from the center of the burner (15 mm from each side). Because of the non-linear processing required, in particular the division of the “red” and “blue” image to yield temperature, single-shot measurements are required. Averaging should be performed on the final scalar fields for equivalence ratio and temperature. However, this is time consuming and potentially suffers from noise problems. Therefore, multi-shot images were also taken at each position in the scan and processed along with the single shots for comparison. Row (a) shows the images obtained via such “incorrect” averaging, while the images in row (b) were obtained through averaging the 20 available single shots after all corrections. The degradation of spatial resolution due to binning is evident, but in return, the  $S/N$  of the images for both cases are comparable, being between 15 and 25 for the “red” images, and about 11 for the temperature images (as measured in relatively homogeneous zones with average equivalence ratios and temperatures).

Single shot images (row c) of the “red” and “blue” signals show the expected shot noise but are still acceptable. As is evident in the temperature image, the division of these two noisy images inevitably creates more noise. Typical  $S/N$  for “red” single-shot images is between 6 and 10, with  $S/N$  for one single-shot temperature field being about 5. The “red” and “blue” single shot images look similar in structure to the average. Other images in the scan showed greater signs of the unsteady operating conditions of the burner.

As outlined above, the temperature images were used to calculate a correction coefficient, to be multiplied with the “red” image to yield the true equivalence ratio. It was found, however, that for the single-shot images this process added an unacceptable amount of noise to the otherwise relatively good “red” images. Considering that most of the images showed that the prevailing temperatures, even close to the catalyst, were in the range where the correction factor is only weakly dependent on temperature, this correction was omitted. For temperatures between 348 and 700 K, this potentially

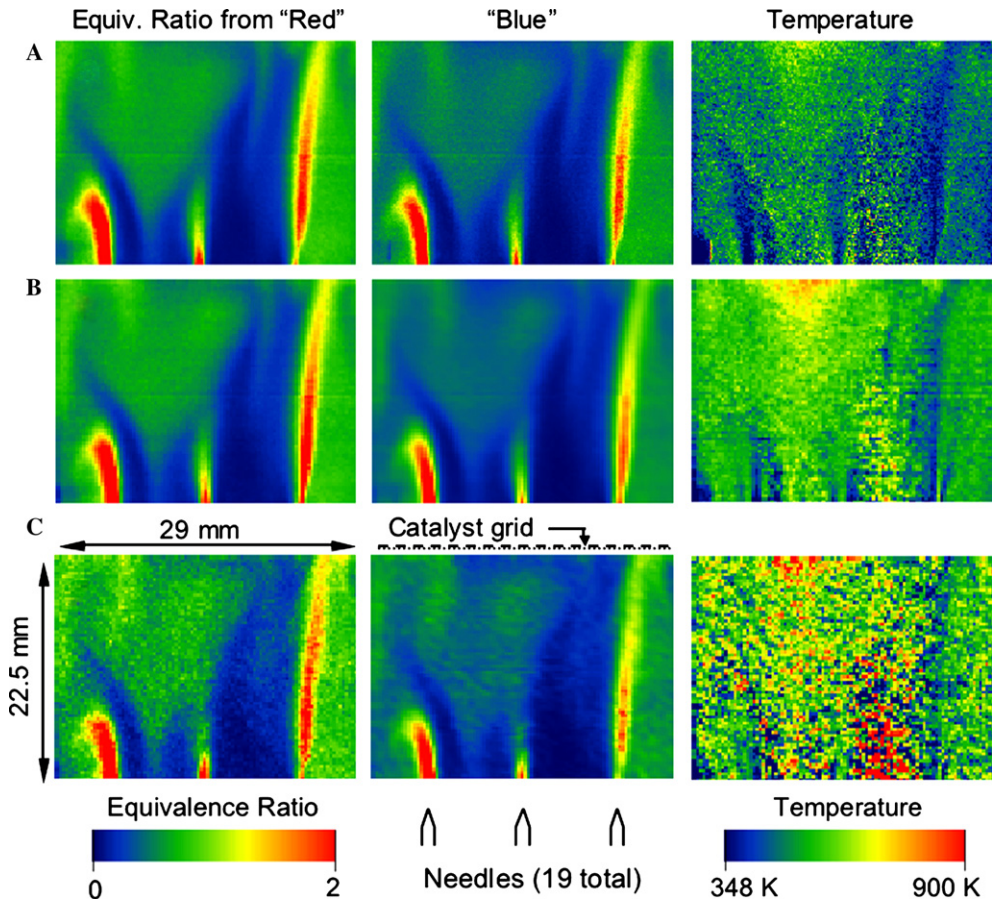


Fig. 8. Row (A): images obtained from a multi-shot exposure (averaged on-chip before processing). The “red” image (left) is converted to local equivalence ratio (uncorrected for temperature). Row (B): images obtained from single shots and then averaged after (non-linear) processing. Row (C): single-shot images. The equivalence ratio color scale is valid for the “red” image only, since the “blue” image is not converted to an equivalence ratio. Electro spray needles and the catalyst grid are schematically shown in their positions relative to the imaged area.  $\Phi_{\text{global}} = 0.57$  and  $T_{\text{inlet}} = 348$  K.

results in a 15% error; for high temperatures however, the possibility of large errors exists.

The temperature images indicate a very inhomogeneous region in the lower part of the burner. Cool regions surrounding the jets of fuel-rich mixture issuing from the vicinity of the nozzles can be seen. While it is expected that evaporation of the fuel leads to local cooling, the temperature in some of those regions is measured to be less than 350 K, which is unlikely. Also, the averaged single-shots show a hot region between the jets in the middle and on the right, while the multi-shot image indicates that this region is at about average temperature. Since almost no unsteadiness is observed here, both images should agree. These errors and discrepancies are most likely due to the exaggeration of somewhat incorrect background subtraction through very low overall signal, as those are regions that contain almost no fuel ( $\phi < 0.05$ ).

The temperature images corresponding to the central part of the burner (such as those shown

here) show a hot region in the top center, while others away from the center show large regions close to the side walls with high equivalence ratio. This could be interpreted as recirculation up along the sides and corners of the burner into the center, with the mixture heating up as it flows along the catalyst grid. The flow would then turn downward and mix with the incoming, inhomogeneous fuel/air jets that can be clearly seen on the images. However, for a precise evaluation velocity data would be necessary.

The overall temperature, as calculated by spatially integrating the top region (away from the artifacts in the lower parts of the image and keeping some distance to both the presumably cold walls and hot catalyst), varies between 470 and 530 K for the multi-shot averages, and between 470 and 550 K for the single-shot averages. This is somewhat elevated from the air inlet temperature of 475 K, as would be expected.

#### 4. Conclusions

A two-color PLIF technique was developed to simultaneously measure temperature and equivalence ratio with naphthalenes as tracer for fuel surrogates for diesel-like fuels. Some temperature and quenching effects were quantified. Two-color PLIF was applied to a mesoscale burner prototype. The accuracy of the temperature measurements was limited by the flat-field and background corrections applied, which are somewhat difficult due to the high boiling point of the tracer and scattering of fluorescence on burner surfaces. Weak single-shot signal required binning, but the spatial resolution at reasonable noise levels was still sufficient to observe the important features of the mixing process in the burner. Signal/noise will have to be improved through faster optics and more extensive averaging to effectively perform temperature corrections on the equivalence ratio images. The temperature field close to the catalyst grid was found to be surprisingly uniform, except for a hot center portion that might be attributed to recirculation in the chamber.

#### Acknowledgment

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#### Comments

*Wieland Koban, Universität Heidelberg, Germany.* I have a comment concerning the figure showing that the LIF signal is proportional to the fuel/air ratio. It should be explicitly mentioned that equivalence ratio is varied by varying tracer concentration. The axis could also be labeled as tracer number density. It is misleading to conclude from this plot that the signal is proportional to the fuel/air ratio in general, as the situation changes when keeping number density constant by varying the oxygen concentrations. In presentation at this symposium [1] we show that in general the LIF-signal of aromatics is *not* proportional to the fuel/air ratio.

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*Reply.* It is correct that to obtain the data underlying Fig. 8A, the equivalence ratio  $\Phi$  was varied by varying the tracer concentration. This was selected to correspond to the case in our burner, where we have fuel mixing with air only at constant pressure. The

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statement that fluorescence is proportional to equivalence ratio should therefore be qualified with respect to the given operating conditions. Your group ([1] in paper and references therein) has indeed given a much more thorough analysis of this aspect of aromatic-tracer LIF for the case of toluene. The data used to obtain the Stern–Volmer plots of Fig. 6 can be re-plotted in terms of equivalence ratio and then show that at 500 K 1-MN fluorescence is linear in  $\Phi$  down to an oxygen concentration of about 7%, but at 800 K only to 15% or 20% oxygen, i.e., very little less than in air. This is the effect of decreasing quenching efficiency, as is pointed out in the paper. In a very limited comparison with toluene, we found that the Stern–Volmer coefficient for oxygen quenching of 1-MN is greater than that of toluene at all temperatures.

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*Jerry Seitzman, Georgia Institute of Technology, USA.* Could you describe the dynamic range requirements for a camera in this technique? It seems that the large drop in signal with temperature will make temperature measurements across a large range very difficult. This would be



even worse in a system where hot air mixes with fuel (so high temperatures correlate to low tracer mole fraction).

*Reply.* In our imaging experiments in the burner, the single-shot S/N was clearly shot-noise limited. It is correct that the temperature sensitivity exacerbates the problems that stem from seeding one stream only in a two-stream problem. As can be seen from Fig. 8B, the loss in signal for the “red” part of the spectrum from 700 K to 900 K is a factor of 3 (and about a factor of 5 for the total signal, not shown), multiplying whatever dynamic requirements would be made by isothermal mixing in the case of a cold, rich jet with a hot, lean environment.



*Robert P. Lucht, Purdue University, USA.* What was the reason that you restricted your laser energy to 20  $\mu\text{J}$ /pulse? Higher pulse energies would give you higher S/N ratios.

*Reply.* Pulse energies of  $\sim 20 \mu\text{J}$  were used for the spectrally resolved experiments in which the laser was formed into a line. For the imaging experiments, pulse energies were restricted to 0.4 mJ to avoid saturation. The beam profile was not very uniform, and with a cleaner profile it might be possible to stay in the linear fluorescence regime for somewhat higher energies. We found 1-MN and naphthalene fluorescence to saturate at much lower energies than toluene, for example. This is probably due to the stronger absorption.