Design and implementation of a mixture fraction diagnostic for aerosol-laden turbulent jets

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ABSTRACT

A facility has been built to study the aerosol formation processes in supersaturated jets using Phase-Doppler Interferometry (PDI). Experimental studies with this facility will investigate theories which propose a critical relationship between the local temperature and vapor concentration, properties which can be related to the local mixture fraction, and the aerosol density and growth rate. In order to be able to condition the PDI measurements on these local properties, a spatially and temporally resolved tracer-based laser-induced fluorescence diagnostic has been designed to measure the mixture fraction of fluid originating from the vapor laden jet. The diagnostic uses toluene vapor excitation at 266 nm to produce a fluorescence signal which is measured and related to a corresponding mixture fraction. This work presents a summary of the performance models and subsequent design analysis that has been used to finalize the design of the mixture fraction diagnostic. Preliminary measurements will be carried out in the summer of 2016.

1. Introduction

Particle nucleation and growth in supersaturated vapors is a primary concern in the design of industrial processes (Kodas, 1999) and the study of natural phenomena such as cloud formation (Pruppacher and Klett, 2010). Many of the processes of interest in aerosol-laden flows occur in regions of turbulent mixing where the vapor concentration and temperature distributions are non-uniform in both space and time. A typical industrial process might involve a warm flow that is saturated with an aerosol precursor vapor mixing with a cold inert gas, as is the case with the production of some commercial powders (Ingebrethsen, 1983). Recent developments in aerosol modeling for use in high-fidelity simulation of the turbulent mixing fields that create supersaturated conditions have been made that predict nucleation rates and particle size distributions in turbulent flowfields (Attili et al., 2014; Attili et al., 2015; Bisetti et al., 2012; Zhou et al., 2014). This motivates the development of a database of experimental studies of the aerosol
formation processes in turbulent mixing fields so that the simulations can be benchmarked and validated.

A turbulent aerosol jet facility has been developed to study the formation and growth of aerosols in well-controlled turbulent mixing fields. The facility generates a hot, turbulent jet that is saturated with dibutyl phthalate (DBP) vapor. The jet mixes with a low-speed, cold co-flow and the resulting supersaturated DBP vapor condenses into aerosol particles. Spatially and temporally resolved measurements of particle size, number density and two-component velocity are made using a Phase-Doppler Interferometry (PDI) system. For the particle properties and growth data to be interpreted in the context of current theory and simulations based on the theory’s application, it is critical that the local conditions of the fluid be known for those measurements. The local conditions of interest include saturation ratio and temperature which can be related to the local degree of mixedness, i.e. the local mixture fraction. It is therefore necessary to design a non-intrusive diagnostic capable of making spatially and temporally resolved measurements of mixture fraction in a flow field with inhomogeneous distributions of vapor concentration and temperature. These complementary mixture fraction measurements will be conducted simultaneously with the PDI measurements.

Some of the challenges of this measurement are similar to those any other non-intrusive spatially and temporally resolved diagnostics. However, an important additional complexity that is unique to this project is the presence of the DBP vapor and its effect on mixture fraction measurements. The broadband UV absorption spectrum of DBP has been computed and measured by Du et al (Du, 2014). It is known to absorb at wavelengths associated with many common UV laser induced fluorescence (LIF) target species, however, to the authors’ knowledge, the fluorescence properties of its vapor state have yet to be characterized.

This work presents a comprehensive design analysis related to the development of a laser-based mixture fraction diagnostic for use in supersaturated, aerosol laden turbulent jets. The analysis includes modeling of the fluorescence and detection processes and potential noise sources in the diagnostic. An analysis the feasibility of an indirect measurement of mixture fraction using a two-color toluene LIF thermometry is described. The methodology for choosing an appropriate tracer and excitation strategy with consideration of potential DBP interference is discussed. A brief summary of the design of the LIF diagnostic and its subsystems is described as well as a strategy
for integration with the facility and PDI system. This design has yet to be tested and preliminary measurements using the integrated system are scheduled to be made in June of 2016.

2. Model of organic tracer fluorescence

In designing a LIF diagnostic like the one described in this work, it is critical to estimate the effective fluorescence yield in order to predict signal and noise levels. The first step in this design analysis was the development of a mathematical model of the absorption, quenching, and fluorescence processes experienced by organic tracer species, specifically toluene and acetone. In general, a model for LIF excitation may be expressed with the spectroscopic properties of the target species condensed into the product of an absorption cross-section, $\sigma$, and a fluorescence quantum yield (FQY), $\phi$, if the function dependencies of those quantities are well characterized. In the case of toluene and acetone, the absorption cross-section and FQY are both functions of excitation wavelength, $\lambda_{ex}$, and local gas temperature, $T$. Additionally, the FQY is also dependent on the fluorescence wavelength that is observed, $\lambda_f$, and potentially on the number density of particular quenching species, $n_i$. Equation (1) gives an expression for the emitted LIF spectral density, $dS_f/d\lambda_f$, in photons per unit wavelength where $E$ is the energy per laser pulse, $h$ is Planck’s constant, $c$ is the speed of light, $L$ is the length of the probe volume, $X_t$ is the local mole fraction of the tracer, $P$ is the local pressure, and $k$ is the Boltzmann constant.

$$\frac{dS_f}{d\lambda_f} = \frac{E \lambda_{ex}}{hc} L \frac{X_t P}{kT} \sigma(\lambda_{ex}, T) \phi(\lambda_{ex}, \lambda_f, T, n_i)$$  \hspace{1cm} (1)

To put the model in the context of measured fluorescence signal, a spatial collection factor must be included and the effect of any optical filters deployed in the detection system must be considered. Equation (2) gives a derivation of the fluorescence photons that are collected onto the detector, $S_m$, where $\Omega$ is the solid angle subtended by the collection optics and $F(\lambda_f)$ is the spectral transmittance of all of the optics, including filters and lenses, that are used in the detection system.

$$S_m = \frac{\Omega}{4\pi} \int \frac{dS_f}{d\lambda_f} F(\lambda_f) d\lambda_f = \frac{E \lambda_{ex}}{hc} L \frac{X_t P}{kT} \sigma(\lambda_{ex}, T) \frac{\Omega}{4\pi} \int \phi(\lambda_{ex}, \lambda_f, T, n_i) F(\lambda_f) d\lambda_f$$  \hspace{1cm} (2)
Equation (2) is a general formulation for the collected fluorescence for both toluene and acetone. In order to fully express the absorption cross-sections and FQYs used in the model, toluene and acetone must each be considered individually.

### Toluene

For toluene, the spectroscopic properties related to 266 nm excitation are well characterized by previous works by Koban et al. (Koban, 2004; Koban, 2005a; Koban, 2005b; Koban, 2005c). Additionally, high-repetition rate, high-power solid state lasers are available that emit 266 nm radiation which provide adequate signal and sub-millisecond time resolution. For these reasons, the toluene fluorescence model presented in this work considers only 266 nm excitation and so the excitation wavelength variable, $\lambda_{ex}$, will be removed from all of the subsequent equations related to toluene fluorescence.

The only non-negligible source of non-radiative quenching for excited toluene in this experiment is molecular oxygen. Koban et al. model the effect of oxygen quenching as a function of temperature and oxygen number density, $n_{o_2}$ (Koban et al., 2005b). This work incorporates that model of oxygen quenching rate as a corrective factor, $C_{Q}^{O_2}$, which diminishes the FQY that would be expected in an anaerobic nitrogen bath, $\phi^{N_2}$, according to:

$$\phi(\lambda_f, T, n_{o_2}) = C_{Q}^{O_2}(T, n_{o_2})\phi^{N_2}(\lambda_f, T)$$

(3)

It should be noted that the quenching factor is cast as independent of the emission spectrum. This is justified by observations by Koban et al. which showed no significant effect of oxygen quenching on the shape of the fluorescence spectrum for 266 nm excitation. On this basis, this work makes the approximation that the entire emission spectrum is uniformly quenched and therefore $C_{Q}^{O_2}$ is independent of $\lambda_f$.

To simplify both notation and the complexity of the computations required by the model, it is desirable to define the effective FQY based on the collected portion of the fluorescence spectrum as a new variable, $\phi_{m}^{N_2}$, which can be pre-computed using measured toluene fluorescence spectra and transmittance curves for optical filters of interest as reported by the manufacturer (Semrock).

$$\phi_{m}^{N_2}(T) = \int \phi^{N_2}(\lambda_f, T)F(\lambda_f)d\lambda_f$$

(4)
By making the assumption of the spectral independence of oxygen quenching as previously noted, the following expression analogous to the definition shown in equation (3) can be made for the integrated FQY:

\[
\phi_m(T, n_{o_2}) = C^{O_2}_{Q}(T, n_{o_2}) \phi^{N_2}_m(T)
\] (5)

The integrated, oxygen quenched FQY may now be substituted into equation (2) to simplify it to:

\[
S_m = \frac{E \lambda_{ex} L}{h c} X_{tot} P \sigma(T) \Omega \frac{4 \pi}{3} \phi_m(T, n_{o_2})
\] (6)

At the heart of the oxygen quenching model described by Koban et al. is a decomposition of the spectrally integrated FQY, \( \phi_i^{N_2} \), into a short-lived component, \( A_1 \), and a long-lived component, \( A_2 \), the sum of which describe toluene fluorescence from 266 nm excitation (Koban et al., 2005b). Each of these has been observed to experience its own quenching dynamics described by unique Stern-Volmer coefficients, \( k_{SV,1} \) and \( k_{SV,2} \) respectively. The oxygen quenched, integrated FQY is then expressed by the following model:

\[
\phi_i^{N_2}(T) = A_1(T) + A_2(T)
\] (7)

\[
\phi_i(T, n_{o_2}) = \frac{A_1(T)}{1 + n_{o_2} k_{SV,1}(T)} + \frac{A_2(T)}{1 + n_{o_2} k_{SV,2}(T)}
\] (8)

This model can be related to the oxygen quenching correction factor defined in equation (3) by again invoking the spectral independence approximation for the effect of oxygen quenching. This allows a comparison of the quenching ratios of the experimentally observed, spectrally filtered FQY, \( \phi_m \), with that of the integrated emission FQY, \( \phi_i \). From this follows a derivation for the expression for \( C^{O_2}_{Q} \):

\[
C^{O_2}_{Q}(T, n_{o_2}) = \frac{\phi_m(T, n_{o_2})}{\phi^{N_2}_m(T)} = \frac{\phi_i(T, n_{o_2})}{\phi_i^{N_2}(T)} = \frac{A_1(T)/\phi_i^{N_2}(T)}{1 + n_{o_2} k_{SV,1}(T)} + \frac{A_2(T)/\phi_i^{N_2}(T)}{1 + n_{o_2} k_{SV,2}(T)}
\] (9)

The model for toluene fluorescence is now fully described with the expression for \( \phi_i^{N_2}(T) \) taken from Koban et al. (2004) and those expressions for \( A_1(T), A_2(T), k_{SV,1}(T) \), and \( k_{SV,2}(T) \) taken from Koban et al. (2005b).
Acetone

Acetone is another candidate as an organic tracer and its use as a LIF target has been well studied (Lozano et al., 1992; Thurber et al., 1997; Hu et al., 2002). The model for acetone fluorescence used in this work is based on measurements of LIF emission per unit mole fraction per unit laser energy, $S_f^+$, as reported by Thurber et al. (1997) and defined as:

$$S_f^+ \propto \frac{P}{T} \sigma(\lambda_{ex}, T) \phi_l(\lambda_{ex}, T)$$  \hspace{1cm} (10)

Equation (2) must now be recast in terms of the ratio provided in Thurber et al. as shown in equation (10). To cast the fluorescence signal into an absolute fluorescence estimate instead of a relative measure, this model takes the relationship in Equation (10) to be an equality and uses values for $\sigma$ and $\phi_l$ found in Lozano et al. (1992) to compute an estimate for $S_f^+(\lambda_{ex}, 295K)$.

$$S_m(\lambda_{ex}, T) = \frac{E \lambda_{ex}}{hc} \frac{X_{ace}}{k} \sigma(\lambda_{ex}, T) \Omega \frac{4\pi S_f^+(\lambda_{ex}, T)}{S_f^+(\lambda_{ex}, 295K)} S_f^+(\lambda_{ex}, 295K)$$ \hspace{1cm} (11)

Data shown in figure 1 of Thurber et al. was digitized and best fit curves were used to find the variation with temperature in the ratio of $S_f^+$ to its reference value. This was done for 266 nm and 308 nm excitation because these were the operating wavelengths of the two lasers that were being considered for this experiment. Equations (12) and (13) show the temperature correlations that were used to predict the fluorescence yield for acetone in the model.

$$\frac{S_f^+(266nm, T)}{S_f^+(266nm, 295K)} = \frac{-0.5501T + 628.6K}{T + 173.6K}$$ \hspace{1cm} (12)

$$\frac{S_f^+(308nm, T)}{S_f^+(308nm, 295K)} = \frac{-0.03642Y^3 + 0.01216Y^2 - 0.1151Y + 0.6712 \text{ where } Y}{221.6K}$$ \hspace{1cm} (13)

It has been noted that acetone exhibits significant long-lived phosphorescence emission in addition to fluorescence when excited with UV radiation. While this could cause interference issues in an anaerobic environment, the phosphorescence signal is strongly quenched by the oxygen present...
in the air co-flow stream used in this experiment. Hu and Koochesfahani (2002) demonstrated a reduction in acetone phosphorescence emission of greater than four orders of magnitude at mixture fractions of less than 0.5 for 308 nm excitation. All of the measurements considered in the planning of this experiments were predicted to exhibit mixture fractions of no more than 0.6 and so phosphorescence interference is not a concern in this analysis and design.

**Measurement SNR**

The relatively small collected light intensities of the LIF measurements require the use of high-sensitivity, low-noise detectors. Photon fluxes for measurements using either acetone or toluene are predicted to fall in the range of $10^7$ to $10^{10}$ photons per second, a regime where photomultiplier tubes (PMTs) are typically considered the best detection option. This design will employ PMTs that are integrated in time using Stanford Research Systems (SRS) SR250 boxcar averagers to improve detection Signal-to-Noise Ratios (SNRs) and reduce the data sampling rate required to resolve the fluorescence signal.

A model for the noise characteristics of the integrated detection system is needed in order to compare the performance of different diagnostic designs. An application note published by SRS provides the following noise model for PMTs integrated with a boxcar averaging system (SRS, 2012):

\[
\text{SNR} = \frac{0.8\sqrt{N_S}}{\sqrt{1 + \frac{N_D + N_{DAQ}^2}{N_S} + \frac{V_n \Delta t}{1.5N_S G^2 R^2 e^2}}}
\]

(14)

with the additional term $N_{DAQ}$ defined as

\[
N_{DAQ} = \frac{V_{DAQ} \Delta t}{MeGR}
\]

(15)

representing the noise introduced by the data acquisition system. Because of the dynamic range required and low light levels associated with most of the measurement points, measurements may be sensitive to even very low magnitude voltage fluctuations occurring during sampling. Therefore, $N_{DAQ}$ is an equivalent photon count that accounts for noise from the high-speed data acquisition system.
The 0.8 factor in equation (14) is a noise factor that accounts for the fluctuations in gain amplitude due to stochastic processes in the dynode amplification stages. The number of fluorescence signal photons collected during an integration gate duration, \(\Delta t\), is represented by \(N_S\); \(N_D\) is the number of dark photons during the gate; \(V_n\) is the input noise density of the boxcar average; \(G\) is the PMT gain; \(R\) is the load resistance; and \(e\) is the magnitude of the charge of an electron. In equation (14), \(M\) represents the gain factor due to the adjustable sensitivity setting of the boxcar integrator which must be considered because noise introduced by data acquisition will not have experience this amplification.

3. Study of thermometry feasibility

An initial strategy for making local mixture ratio was an indirect one using two-color toluene thermometry to measure temperature which could be related to mixture fraction. The technique was described in detail by Miller et al. (2013) and has been applied by several other researchers to make 2-D imaging measurements of temperature in gas flows (Luong et al., 2006; Kaiser et al., 2013; Miller et al., 2014; Estruch-Samper et al., 2015; Gamba et al., 2015). The technique leverages the red-shift in toluene fluorescence for 266nm excitation that has been observed as local temperature rises. By using two detectors with mutually exclusive bandpass spectral filters, one that transmits near 280nm and another that transmits above 300nm, the ratio between the two filtered fluorescence signals can be shown to be a monotonic function of temperature with acceptable sensitivity, SNR and accuracy. While the exact shifting mechanism is unknown, the behavior of the spectrum has been well-studied and toluene thermometry techniques have been used to make high-accuracy single-shot PLIF measurements in previous experiments.

Analysis of the two-color toluene thermometry technique in the context of the jet-air mixing of the turbulent jet shows temperature measurement SNRs in the range of 10 to 25 for mixture fractions as low as 0.02. However, the goal of this diagnostic is to measure mixture fraction rather than temperature. If it is assumed that flow times are much significantly shorter than those of thermal diffusion so that the temperature field effectively behaves as a passive scalar, then one may relate the mixture fraction and temperature using conservation of energy through the following linear relation:

\[
Z = \frac{T - 295K}{138K}
\]  

(16)
which can be used to derive the SNR of a mixture fraction measurement based on the SNR of the corresponding thermometry measurement as:

\[
\text{SNR}_Z = \frac{Z}{\sigma_Z} = \frac{T - 295K}{138K} = \left(1 - \frac{295K}{T}\right)\text{SNR}_T
\]  

(17)

Most importantly, equation (17) demonstrates that the SNR of the mixture fraction measurement is always less than that of temperature and unless \(T >> 295 \text{K}\), the reduction in mixture fraction SNR relative to temperature SNR will be significant. Figure 1 shows the deterioration in mixture fraction SNR compared to that of the corresponding temperature measurement for the entire range of mixture fractions. The decrease in SNR for mixture fractions below 0.2 is greater than 90% when compared to that of temperature and will cause the SNR to trend toward zero much faster for mixture fraction measurement. This trend is intuitive when one considers that even a small amount of temperature noise at 296 K, which would give a very high temperature SNR, would result in a nearly zero SNR for an inferred mixture fraction measurement.

Because of the large SNR penalty of using an indirect thermometry measurement of mixture fraction, it is clear that the two-color toluene thermometry technique would be infeasible in this application. We move instead to direct measurement of the mixture fraction based on the fluorescence intensity of the tracer species. It should also be noted that although the assumption
of unity Prandtl number is not as central to the direct mixture fraction measurement as it is to the thermometry technique, it is still implicit in the temperature dependence of certain spectroscopic properties which required the same linear relationship between temperature and mixture fraction to be made in the fluorescence model.

4. Choice of tracer

Selection of the tracer species to be used was determined by an analysis of the predicted signal to noise performance of acetone and toluene as well as consideration of potential interference with DBP vapor, which will be seeded into the jet mass flow downstream of the tracer seeding. Work performed by Du et al. (2014) determined that DBP exhibits a relatively large absorption cross-section in the UV spectrum below 300 nm. Although the absorbance of the DBP vapor will not be strong enough to cause significant attenuation of the laser beam, it is likely to be large enough at 266 nm that electronic excitation leads to measurable DBP fluorescence. Because the fluorescence properties of DBP in this type of environment have not been quantified, it is important to either avoid exciting DBP by using a laser wavelength outside of its absorption bandwidth, or by blocking DBP fluorescence with optical filters.
The use of acetone as a tracer with 308 nm laser excitation will avoid significant excitation of the DBP vapor, however excitation strategies for acetone or toluene excitation at 266 nm will cause significant DBP excitation. It is important then that there is as little overlap as possible between the fluorescence spectra of the potential tracers and DBP in order for the interfering DBP fluorescence to be blocked without a significant reduction in signal. If the overlap between the fluorescence spectrum of either tracer and the DBP vapor is too great, then it will not be possible to make a quantitative interpretation of LIF measurements and that strategy will not be viable.

The relative fluorescence spectral intensities of toluene, acetone, and DBP were measured using a fluorimeter with the substances in liquid phase. Figure 2 shows a comparison between the fluorescence spectra of acetone, toluene and DBP at a temperature of 340 K and an excitation wavelength of 266 nm. Nearly all of the toluene fluorescence occurs below 320 nm, while the DBP fluorescence extends from approximately 310 nm to 400 nm. This shows that a 310 nm longwave bandpass filter can be used to eliminate the DBP fluorescence without a significant impact on the magnitude of the toluene LIF signal. In contrast, the fluorescence spectrum of acetone at this excitation wavelength is quite similar to that of DBP. It is clear that the DBP fluorescence could not be blocked without a major attenuation of the acetone LIF signal. For this reason, it was determined that acetone LIF was only viable with 308 nm laser excitation.

The choice of tracer between toluene excited at 266 nm and acetone excited at 308 nm was based on the predicted SNR values for each approach in the context of the turbulent mixing jet. Mixture fractions in the region of interest in the flow field range from 0 to 0.6 with a majority of

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**Fig. 2** A comparison between the fluorescence spectra of the tracer candidates and DBP.

**Fig. 3** Comparison between the mixture fraction measurement SNRs of toluene and acetone LIF.
measurements of interest to be made at a mixture fraction of 0.2 or below. The 308 nm excitation of the acetone was modeled using parameters corresponding to a Spectra-Physics Tsunami Ti:Sapphire laser operating with a nominal power of 140 mW and the 266 nm toluene excitation was modeled using parameters from a Frankfurt Laser Company DPSS Nd:YAG laser with a nominal power of 65 mW. The fluorescence and measurement noise models described in section 2 were applied to both tracer strategies for the range of mixture fractions of interest. The SNR was determined for the mixture fraction measurement itself based on random variations in the measured fluorescence signal. Figure 3 shows a comparison of the predicted SNR values for each of the two tracer strategies as a function of local mixture fraction in the probe volume.

It should be noted that by the definition of mixture fraction SNR as the ratio of the mixture fraction to the random variation in mixture fraction, the SNR necessarily decreases to zero at a mixture fraction of zero. The SNR of toluene decreases very rapidly with decreasing mixture fraction because of the dual effects of lower toluene number densities and increased oxygen number densities which exacerbate non-radiative quenching. Acetone is not strongly quenched by oxygen which explains its higher SNR values compared to toluene for low mixture fractions. Despite the lack of quenching and a higher number density than toluene (due to a higher vapor pressure) associated with acetone, the toluene SNR is greater than that of acetone for all mixture fractions of 0.05 or greater. Not only does toluene provide a higher quality signal for a larger range of anticipated mixture fractions, it provides a dramatically higher SNR for mixture fractions of 0.2 and above where vapor concentrations are high and aerosol particles are predicted to be the densest. In consideration of the different predicted performances, toluene was chosen as the tracer species for this diagnostic.

5. Design of toluene LIF diagnostic

Collection optics

The design of the collection optics for the LIF system was created with the aid of Zemax optics simulation software. Primary concerns were the efficient collection of fluorescence emission, spatial resolution, and reduction of chromatic distortions. The focal length of the collimating lens must be at least 350 mm in order to prevent the collection optics from disturbing the co-flow and to avoid coating the optics with toluene and DBP. Zemax simulations were used to perform ray tracing from different spatial points near the probe volume onto the focal plane of the collection optics in order to select the appropriately sized pinhole and to evaluate the overall spatial
resolution of the diagnostics. Several different designs of different numbers and combinations of optics were evaluated in order to optimize the collection performance. Eventually, trial and error indicated that a two-element design offered the best performance with a reasonable tolerance to misalignment of the optics. Figure 4 shows the simulated ray tracing of the final design which uses 500 mm and 150 mm plano-convex lenses in series. Two sets of collection optics and detectors will be deployed in the experiment in order to verify measurements and increase the SNR through averaging.

![Fig. 4 Simulated ray tracing of the probe volume center point (left-hand side) focused onto the pinhole (right-hand size) by the selected LIF collection optics configuration.](image)

In addition to the collection optics, the toluene diagnostic system includes two subsystems, an absorption cell to monitor the level of toluene seeding in the nitrogen stream and a toluene vapor seeder to produce the desired toluene concentration. An analysis to model the performance of these subsystems and optimize their performance was conducted as part of this work.

**Absorption Cell**

One of the attractive aspects of the indirect thermometry diagnostic was that its signal would be independent of the local toluene concentration as well as of the concentration of toluene in the central jet flow. With the direct toluene measurement, it is critical to know the concentration of toluene in the central jet because this determines the relationship between the measured LIF signal and the mixture fraction in the probe volume. This motivates the need for an in-line absorption

![Fig. 5 Notional schematic of absorption cell system. A: UV beam sampler; B: 50-50 beamsplitter; C: Focusing lenses; D: Reference detector; E: Absorption cell with toluene vapor; F: Absorption cell detector; G: Breadboard.](image)
measurement in between the toluene seeder and the DBP evaporator. The absorption measurement monitors any temporal variations in the toluene concentration of the central jet. Fig. 5 provides a notional schematic of the design of the absorption cell system.

A model of the noise characteristics of the toluene absorption cell was created to size the length of the absorption cell. In order to accurately determine the absorption through the optical cell, the ratio of the signal through an empty cell, $S_{cell,0}$, to the signal directed into the reference detector, $S_{ref}$, can be defined as the split ratio factor, $C_{sp}$:

$$C_{sp} = \frac{S_{ref}}{S_{cell,0}}$$  \hspace{1cm} (18)

It is straightforward to measure the value of this split ratio which can then be used to correct an expression of Beer’s law. Solving the Beer’s law expression in equation (19) for the mole fraction of toluene yields the conversion from signal strength to absorber mole fraction shown in equation (20).

$$C_{sp} \frac{S_{cell}}{S_{ref}} = e^{-\sigma \ln x_{tot}}$$  \hspace{1cm} (19)

$$X_{tot} = \frac{\ln(S_{ref}) - \ln(S_{cell}) - \ln(C_{sp})}{\sigma n L}$$  \hspace{1cm} (20)

In equation (20) we can assume that the absorption cross-section, $\sigma$, and number density, $n$, remain constant so that the only two sources of noise in the measurement are the noise associated with each detector. Under this assumption, it can be shown that the SNR for measurement of the toluene mole fraction in the absorption cell flow is given by:

$$\text{SNR}_{X_{tot}} = \frac{-\ln \left( C_{sp} \frac{S_{cell}}{S_{ref}} \right)}{\sqrt{\frac{1}{\text{SNR}_{cell}} + \frac{1}{\text{SNR}_{ref}}}}$$  \hspace{1cm} (21)

In equation (21), $\text{SNR}_{cell}$ and $\text{SNR}_{ref}$ represent the SNRs of the photodetectors for the absorption cell and reference beam paths respectively. The SNR for a general photodiode detector is modeled as
where $\mathcal{R}$ is the responsivity of the detector in terms of signal current per unit optical power; $W$ is the incident optical power; $B$ is the bandwidth of the detector circuit; NEP is the noise equivalent optical power of the detector; and $I_{DAQ}$ is the equivalent noise current produced by the data acquisition system.

Using this model, the signal to noise ratio of the absorption cell toluene mole fraction measurement can be plotted as a function of toluene mole fraction for any particular absorption cell length. The detector properties were taken from the manufacturer’s specifications for the Thorlabs DET25K biased photodiode which is the detector that will be used in the planned experimental measurements. Figure 6 shows the SNR for a range of toluene saturations for the three absorption cell lengths that were available from suppliers: 5 cm, 10 cm, and 20 cm.

The results of the simulation show that the SNRs of the absorption systems of various lengths will rise to the same peak value at a particular toluene concentration depending on the length of the absorption cell. Above this value, the SNR quickly falls off due to low light levels passing through to the cell transmission detector because of the strong absorption, which causes low values for $\text{SNR}_{cell}$ (note that $\text{SNR}_{ref}$ will be constant for a given $C_{sp}$ which was taken to be 0.5 for this
analysis). The vertical dashed line on Fig. 6 shows the saturation concentration of toluene at 22°C, the highest mole fraction that would be expect during the experiment. This peak concentration value falls nearest to the concentration of peak SNR for the 10 cm long absorption cell and is on the low concentration side of the peak, away from the steep SNR drop-off. Using this analysis, 10 cm was chosen as the length of the toluene absorption cell.

**Toluene seeder**

The toluene seeder, a reservoir of liquid toluene through which the nitrogen gas flow is bubbled prior to the center jet nozzle, is a critical component of the LIF system as it must produce sufficient toluene concentrations in the seeded stream with as little temporal variation as possible. The performance of a vapor seeder is determined mostly by: (1) the sizing of the vessel; (2) the way that the gas flow is injected into the liquid; and (3) the thermal management of the heat losses due to evaporation of the liquid tracer. A model of the toluene vaporization process in the seeder has been developed to guide the decisions about sizing for the flow rates that will be used for the planned measurements.

For the purpose of modeling the toluene concentration in the seeder output gas, it is assumed that the toluene vapor is saturated at its vapor pressure for a given temperature. Using Antoine equation coefficients computed from measurements made by Pitzer and Scott (1943), the previous assumption can be leveraged to express the mole fraction of toluene in the outlet gas as a single variable function of the liquid toluene temperature, $T$:

\[
X_{tol}(T) = \frac{1}{1.013} \times 10^{4.14157 \frac{1377.6 K}{T+50.5 K}}
\]  
(23)

As the flow rate through the seeder increases, the heat loss due to latent heat of vaporization increases. Equation (23) shows that if this heat loss is sufficiently high and is not compensated for with a heating strategy, then the concentration of toluene within the seeded flow will decrease. Fluctuations in toluene concentration that are either too rapid or excessive decreases in toluene concentration could compromise the performance of the diagnostic and must be addressed with the design of the seeder. This evaporative heat loss will be equal to the product of the rate of toluene mass flow out of the reservoir, $\dot{m}_{tol}$, and the mass specific latent heat of vaporization for toluene, $\Delta_{vap} h_{tol}$:
\[ \dot{Q}_{\text{vap}} = \dot{m}_{\text{tol}} \Delta_{\text{vap}} h_{\text{tol}} \quad (24) \]

The latent heat of vaporization of liquid toluene is also a function of \( T \) and this work accounts for that variation with a model for the molar latent heat of vaporization that was found by Mayer and Svoboda (1985).

\[ \Delta_{\text{vap}} H_{\text{tol}}^0(T) = 53.09 e^{-0.2774 T_r (1 - T_r)^{0.2774}} \text{kJ/mol} \quad \text{where} \quad T_r = T/591.7K \quad (25) \]

In addition to the heat lost to evaporation, the only other significant source of heat transfer is from the external heating system, if present. For this work, water heating circuits made of coils of copper tubing were considered as the potential heating system to stabilize the temperature of the liquid toluene. The heat transfer from the water circuit coils into the liquid toluene, \( \dot{Q}_{\text{coil}} \), is a function of the temperature of the liquid toluene, \( T_{\text{tol}} \), the thermal resistance of coil walls and thermal boundary layers between the core water flow and the quiescent toluene, \( R_{T,\text{coil}} \), and the inlet and outlet temperatures of the water circuit, \( T_{H_2O,i} \) and \( T_{H_2O,e} \) respectively. Because the temperature of water changes relatively little from the inlet to the outlet of the coils, it is reasonable to assume a constant heat flux boundary condition for the length of the coil. This yields the following relation for the variation of the exit temperature of the water circuit,

\[ T_{H_2O,e}(t) = T_{\text{tol}}(t) - \left( T_{\text{tol}}(t) - T_{H_2O,i} \right) e^{-1/(\dot{m}_c R_{T,\text{coil}})} \quad (26) \]

which can then be used in the following heat exchanger equation (Incropera et al., 2007) to describe the heat transfer from the water circuit to the toluene reservoir:

\[ \dot{Q}_{\text{coil}} = \frac{T_{H_2O,i} - T_{H_2O,e}}{R_{T,\text{coil}} \ln \left( \frac{T_{H_2O,i} - T_{\text{tol}}}{T_{H_2O,e} - T_{\text{tol}}} \right)} \quad (27) \]

Because the heat transfer through the external walls of the vessel from the room air to the toluene reservoir was computed to be negligibly small compared to the heating circuit, the time evolution of the liquid mass and temperature of the toluene reservoir can be described by the following system of differential equations:
These equations were solved using the ODE45 function in MATLAB. By changing the mass flow of water through the circuit and the geometry of the circuit, which determines the value of $R_{T, coil}$, the seeding performance of different designs can be tested. The most immediate question that can be addressed with this model is whether or not a water heating circuit is necessary for this application, or if the evaporative cooling is small enough to not disrupt operation of the LIF system.
Analysis of seeder performance considered a nitrogen gas mass flow rate of 5 g/s which was the highest mass flow rate of the cases for the planned experimental campaign. This case was analyzed both with and without a water heating coil to determine the effect and necessity of water heating. The heating coil considered was 76 cm in height and had a linear pipe length of approximately 7.6 m. Copper piping with a 5 mm inner diameter was chosen because it was the largest diameter pipe size that could be bent to the necessary radius for fabrication of the heating coil. Figures 7a and 7b show a comparison of the predicted seeder performance with water heating at a flow rate of 7.5 lpm with a water inlet temperature of 20°C and without water heating. An inlet water temperature of 20°C is appropriate for this application because although a higher water temperature would lead to a higher saturation concentration, the liquid toluene should not be heated above room temperature or condensation may occur in the lines downstream as due to cooling of the seeded stream. Anticipated run times to achieve converged statistics with PDI measurements is on the order of 30 minutes.

The simulation results demonstrate the need for a water heating coil as the unheated seeder produces continuously decreasing toluene concentrations due to a drop in liquid toluene temperature. The use of the water heating coil causes the liquid toluene temperature to stabilize at approximately 16°C after about an hour of continuous run-time. This model was also used to determine the minimum required water flow rate to maintain thermal stability of the toluene reservoir.
Fig. 8 Effect of water flow rate on stabilized toluene temperature for a water heating circuit with an inlet water temperature of 20°C.

Figure 8 shows the results of steady state simulations using a variety of heating water flow rates in the same configuration described above. A distinct knee can be observed in the plot near a flow rate of approximately 0.75 lpm, above which little increase in stabilized temperature is predicted. This result indicates that if the facility can supply at least 1 lpm of 20°C water flow, it will be sufficient to maintain the temperature and performance of the seeder system.

6. Implementation of diagnostic

The toluene LIF diagnostic must be effectively integrated with the turbulent jet facility and the PDI system in order to make meaningful measurements. Figure 9 shows a schematic of the instrumentation used to make measurements in the seeded jet. Several aspects of the LIF and PDI systems must be carefully coordinated to ensure that the measurements can be interpreted properly. A chief concern is the alignment of the focused light sources and detection optics to ensure that the same probe volume is being interrogated by both measurements. While alignment represents coordination of the spatial resolution of the two system, it is also critical that the two systems be precisely timed together so that the temporal resolution of the measurements is meaningful.
A LabView controller program will be used to coordinate the timing of the systems as well as the data collection, with the exception of the PDI measurements which will be collected by software developed by the manufacturer, Artium Technologies. The PDI system has a resettable clock feature which will be triggered at a known reference time, $t_0$, using a digital output from the LabView DAQ card. The laser’s Q-switch signal is used to trigger the integration gate of the boxcar integrator and all output signals from the signal integrator and the photodiodes will be sent to the DAQ card for sampling. A schematic representation of the signal paths to be used in the experimental setup are shown in Fig. 10.
Preliminary measurements using the integrated system are scheduled to be made in June, 2016. Central jet nozzles and injection velocities will be varied to operate the facility at different Reynolds numbers. A three-axis mechanical translation stage will move the jet and co-flow exit relative to the probe volume allowing a spatial grid to be sampled in the far-field of the turbulent jet. Simultaneous measurements of mixture fraction and aerosol particle properties will be used to construct conditional statistics of particle formation and growth processes.

7. Conclusion

A mixture fraction diagnostic has been designed to produce precise spatially and temporally resolved measurements in the presence of DBP vapor and aerosol particles. The design considered several alternative tracer and excitation schemes for both direct and indirect measurement of mixture fraction and the analysis demonstrated that a direct measure of mixture fraction using toluene fluorescence excited at 266 nm offered the best performance. This included both predicted signal strength predictions and avoidance of interference by DBP vapor fluorescence. The design of the toluene LIF system’s collection optics and subsystem were optimized through design modeling and analysis of their predicted performance. A strategy for implementation and integration of the diagnostic in the turbulent jet facility has been presented. Preliminary measurements have been scheduled for June of 2016, the results of which will be presented at the 18th Lisbon Symposium in July of 2016.
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