COMPUTATIONAL AND EXPERIMENTAL STUDY OF LAMINAR FLAMES

Grant Number: DE-FG02-88ER13966

Institution: Yale University

Street Address/City/State/Zip:
P.O. Box 208284
New Haven, CT 06520

Principal Investigator: Mitchell D. Smooke

Address:
P.O. Box 208284
New Haven, CT 06520

Telephone Number: (203) 432-4344

Email: mitchell.smooke@yale.edu

DOE/Office of Science Program Office:
Basic Energy Science/Chemical Sciences

DOE/Office of Science Program Technical Program Manager Contact:
Dr. Mark Pederson

PROGRAM OBJECTIVES:

During the past three years, our research has centered on an investigation of the effects of complex chemistry and detailed transport on the structure and extinction of hydrocarbon flames in coflowing axisymmetric configurations. We have pursued both computational and experimental aspects of the research in parallel on both steady-state and time-dependent systems. The computational work has focused on the application of accurate and efficient numerical methods for the solution of the steady-state and time-dependent boundary value problems describing the various reacting systems. Detailed experimental measurements were performed on axisymmetric coflow flames using two-dimensional imaging techniques. Previously, spontaneous Raman scattering, chemiluminescence, and laser-induced fluorescence were used to measure the temperature, major and minor species profiles. Particle image velocimetry (PIV) has been used to investigate velocity distributions and for calibration of time-varying flames. Laser-induced incandescence (LII) with an extinction calibration was used to determine soot volume fractions, while soot surface temperatures were measured with three-color optical pyrometry using a color digital camera. A blackbody calibration of the camera allows for determination of soot volume fraction as well, which can be compared with the LII measurements. More recently, we have concentrated on a detailed characterization of soot using a variety of techniques including time-resolved LII (TiRe-LII) for soot primary particles sizes, multi-angle light scattering (MALS) for soot radius of gyration, and spectrally-resolved line of sight attenuation (spec-LOSA). Combining the information from all of these soot measurements can be used to determine the soot optical properties, which are observed to vary significantly depending on spatial location and fuel dilution. Our goal has been to obtain a more fundamental understanding of the important fluid dynamic and chemical interactions in these flames so that this information can be used effectively in combustion modeling.

SUMMARY OF COMPLETED RESEARCH

Computational and Experimental Flame Studies

Bridging the significant gap between steady flames in simple burner configurations and actual time-dependent engineered combustion systems will continue to require a long and concerted effort encompassing the study of both laminar and turbulent reacting flows and drawing on a large variety of algorithms and approaches. In this endeavor, a lot can be gained from the detailed study of unsteady laminar flames. Even at relatively small Strouhal numbers, such flames often display a much wider range of interactions between fluid dynamics and thermochemistry than are seen at steady-state conditions, generating remarkable spatial structure and sampling regimes of temperature, mixture fraction, residence time, strain, and scalar dissipation that are still in need of thorough scientific analysis. A very useful type of unsteady laminar flame may be created in the laboratory by simply imposing a periodic forcing on the fuel flow rate in a steady coflow laminar diffusion flame.

While significant preliminary results were generated in the comparison of computations with experiments in this system, several important details emerged. First, although the computed flame structure agreed well with the experiments lower in the flame, differences developed as one moved upwards in the flame. These differences became more pronounced as the forcing

amplitude increased. Second, in the case of the sooting flames, to be able to predict the region of soot pinch-off that was observed in the experiments, we had to increase the forcing modulation above that used in the experiments. To be able to reduce the errors in the computational and experimental comparisons, modifications of the computational algorithm we used previously in our time-dependent flame studies were required.

Time-Dependent Modeling of Forced Flames

We applied an implicit Newton-Krylov (NK) algorithm employing high-order compact spatial discretization to a time-dependent jet diffusion flame [1] for a range of axial velocity perturbation amplitudes α: 30%, 50%, 70%, and 90%. The frequency of oscillation γ was fixed at 20 Hz in order to facilitate interaction with the experiments [2, 3]. (In the laboratory, the forcing frequency was chosen to be a multiple of the 10 Hz frequency of the laser). The average axial velocity in the fuel tube and the constant axial velocity in the oxidizer tube were both set to 35 cm/s. For this flow configuration, the Reynolds number was O(100). Emerging from the fuel tube was 65% CH_4 (by mole) diluted with N_2 into air. The inner radius of the fuel tube was 0.2 cm, the inner radius of the oxidizer tube was 2.5 cm, and the width of the fuel tube was 0.038 cm, all corresponding to the specifications of the experimental burner. Steady solutions using both compact spatial discretizations and a conventional low order method were generated on a highly nonuniform grid. These steady solutions were taken as the initial conditions of the time-dependent flame calculations. In the time-dependent systems below z = 1 cm, the experimental and numerical flames (for both methods) are structurally very similar; significant structural disparities only arise well above this region, where a complicated interplay between buoyancy, vortical motion of the fluid, heat and mass transfer, and chemical reaction lead to remarkable structures that have been captured in the laboratory. The "signature" of this oscillating flame system is the pinching off of a parcel of hot fluid which continues to burn as it accelerates upward until heat is diffused away and the reaction is quenched; but structures even more striking than this are found in the radical species concentrations and, for heavier fuels, the soot distributions in the vicinity of the pinch-off [4]. To ascertain the importance of the high order implicit-compact method and to evaluate its promise in future computations, it was important to be able to perform a like-for-like comparison with a conventional low order method, paying special attention to the region well downstream of the base of the flame. One way of doing this was to use a single grid for all computations, with the sole requirement that this grid be sufficiently refined to resolve the triple flame for both the low order and the high order computations.

In Fig. 1, we include plots of the temperature field of the compact method (CS) versus the low order (LO) unsteady flame for the 90% amplitude perturbation. The CS method does a credible job of simulating the pinch-off, capturing the distinctive arrowhead shapes of these hot spots as they convect downstream, and resolving the fine structure of the vorticity field, whereas the conventional LO method damps and smears the solution in the post-flame region. If the problem sizes (the number of unknowns times the number of grid points) of reacting flow computations were only moderately larger than that of similar nonreacting flow simulations, the best solution might be to persist with current validated low order flame solvers by simply employing more grid points. But this is not the case. Clearly, calculations of unsteady flames will benefit from the use of high order methods in a fully implicit implementation. Moreover, with the successful

implementation of the transient compact method, we will be able to compare high-fidelity computational results with measurements of major and minor species, temperature, and velocities in the forced flow for different velocity modulations at significantly longer physical times.

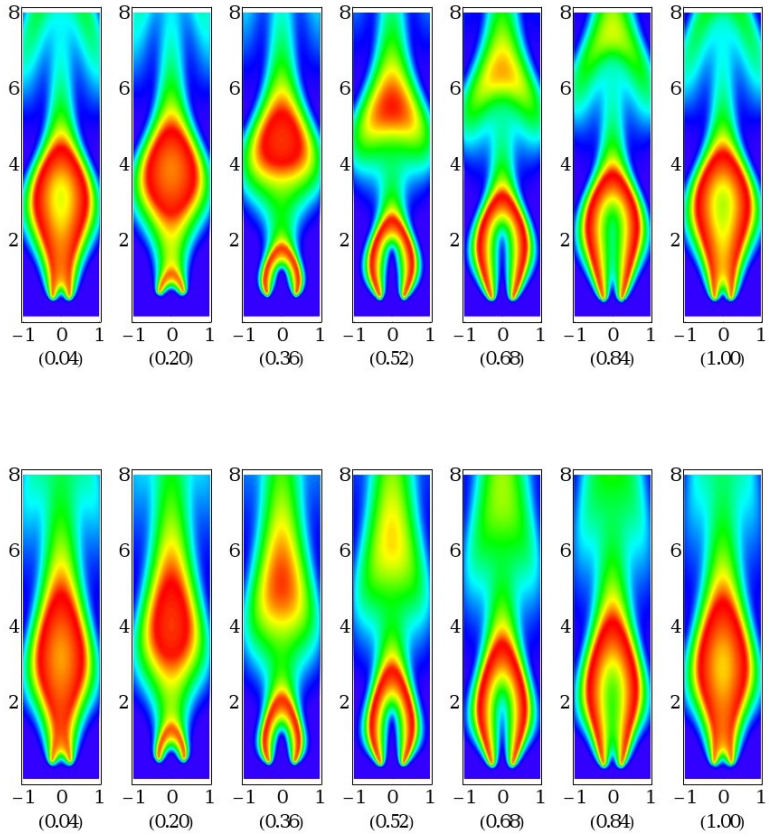

Fig. 1. Temperature field in an oscillating flame with 90% amplitude perturbation, as computed by the implicit solver with compact scheme (top) versus low order (bottom) spatial discretization. The frames are labeled by nondimensional time, i.e., the fraction of the period of oscillation.

NO LIF Measurements in Time-Varying Flames

To expand the characterization of minor species in our nonsooting time-varying flames, experimental measurements of the nitic oxide distribution and absolute concentration have been performed in the 65%/35% methane/nitrogen coflow laminar diffusion flames using laser-induced fluorescence (LIF). Previous measurements of *NO* concentration have been performed by spectrally resolving the *NO* fluorescence emission. The current approach relied on the imaging of the fluorescence, with an improvement in the signal collection efficiency, as well as a reduction of the acquisition time.

The output of an Nd:YAG pumped dye laser was focused into a nitrogen-diluted methane flame (35% and 65% by volume, respectively) and used to excite the NO $A^2\Sigma^+ \leftarrow X^2\Pi$ transition near 226 nm. A cooled CCD camera optically-coupled to an image intensifier collected and imaged the $A^2\Sigma^+ \rightarrow X^2\Pi$ fluorescence emission - (0,0) to (0,5) bands - through a UV bandpass filter. A series of 0.25 mm-spaced 1D measurements allowed for a two dimensional reconstruction of the fluorescence spatial distribution. The $Q_1(18)$ excitation transition was chosen due to its strength; it is well populated at room temperature, it does not overlap with other allowed transitions, and is free from vibrationally hot oxygen interference. The subtraction of an off-resonant signal accounted for molecular broadband fluorescence, incandescence interference, and flame luminosity. A *NO* gas mixture of known composition was used for calibration purposes. As done in previous work [5], and to reduce experimental uncertainties, the comparison has been done between measured and computed fluorescence signal rather than species mole fraction. The implemented fluorescence model relied on a two-level system, under the assumption of linear fluorescence and quenching. Computed temperature field and species mole fractions derived from the numerical simulation accounted for Boltzmann population distribution and quenching correction. To ensure consistency with the model, measurements were performed in the linear regime, below the *NO* saturation threshold. Figure 2 shows measured and computed fluorescence (left and center) in the steady flame as well as a centerline plot (right), which includes results of previous spectroscopic measurements. The agreement in the NO distribution for the steady flame showed excellent consistency between the two experimental methods, and the numerical results deviate from the experimental ones only in the downstream region. Figure 3 shows five phases of two different modulations, 30% and 50%.

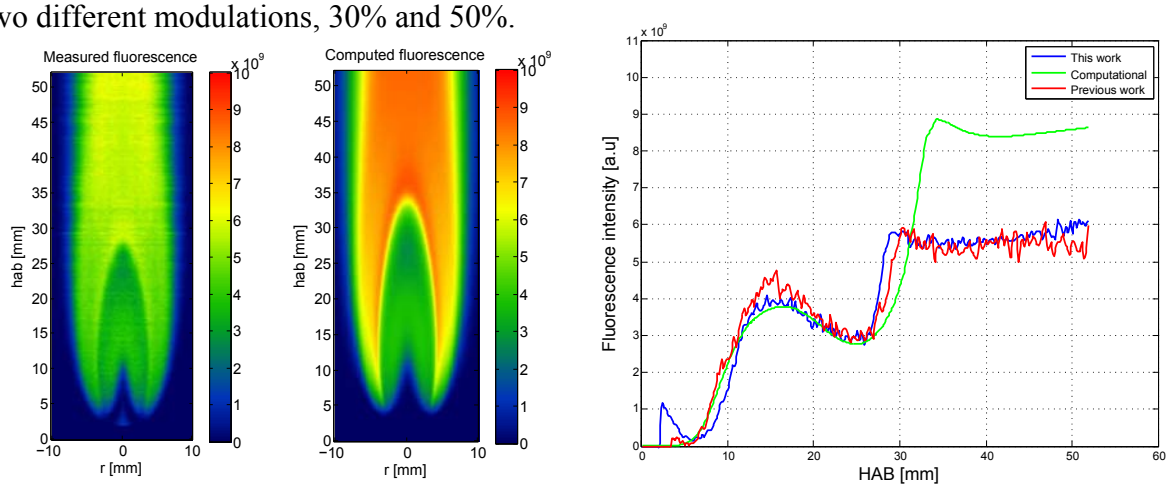

Fig. 2. Measured fluorescence (left), simulated fluorescence (center) and centerline fluorescence profile for the steady flame.

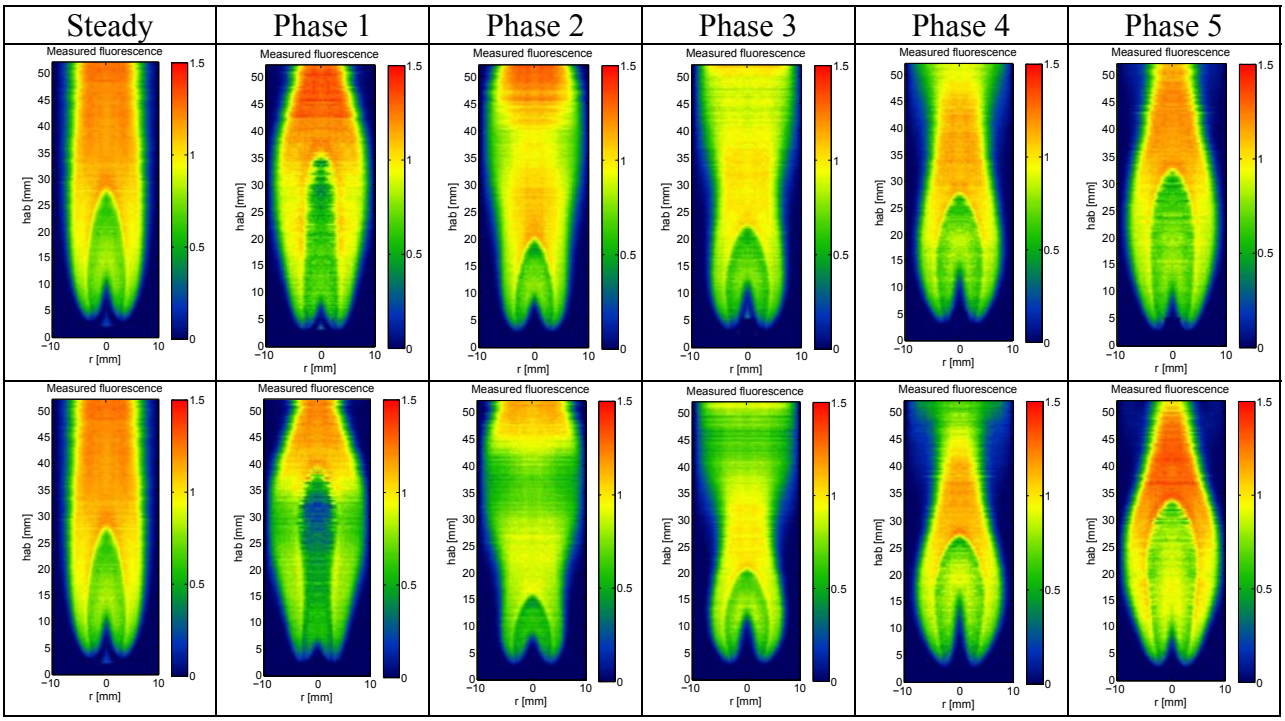

Fig. 3. Measured fluorescence in the time-varying flame: 30% modulation (top), 50% modulation (bottom).

Full Field Temperature Measurements in Sooting Coflow Diffusion Flames

The availability of temperature maps over the entire flow field of sooting coflow diffusion flames would allow significant simplifications to the radiation corrections in our computational work. Mapping temperature over the entire flow field involves measurement over a wide range of temperatures, flow velocities, and scattering characteristics. For this reason, three different techniques were selected and spatially overlaid in order to map the entire flow region. A two-color ratio soot pyrometry technique was used in the sooting region [6], thin-filament pyrometry (TFP) captured the gas temperature adjacent to the sooting region of the flame [7], and a new Rayleigh scattering technique based on Structured Laser Illumination Planar Imaging (SLIPI) was used in the regions below 1150 K [8]. The advantage of the SLIPI approach is that for experiments that involve high background levels (as in the sooting flames), an intensity modulated light sheet serves as a signature that can be used to separate light scattered from within the sheet from background scattering. Our approach involves splitting the output of an Nd:YAG and propagating two beams over 6.9 meters before interfering them above the burner. Relay imaging the signal to the camera and implementation of SLIPI was necessary due to orders of magnitude difference in signal strength between scattering from soot and Rayleigh scattering. The full field temperature maps for the 60% and 80% ethylene flames are shown in Fig. 4, where the data is spatially overlaid from Rayleigh SLIPI, TFP, and soot pyrometry. The figure shows a region of no data from the fuel tube to approximately 3 cm above the burner in both flames. This is a location where measurement with the three diagnostic techniques was not possible. The soot volume fraction is too low for two-color ratio soot pyrometry, temperature is too low for TFP, and there is too much soot for Rayleigh SLIPI.

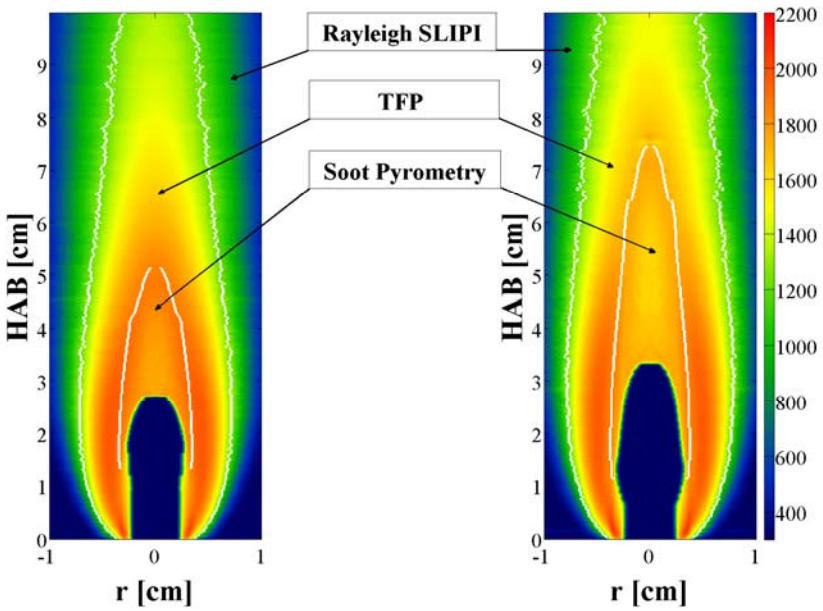

Fig. 4. Flame temperature profiles for 60% and 80% ethylene flames where white lines indicate the interface of two adjoining thermometry techniques. The temperature color bar is shown in Kelvin.

DOE SUPPORTED PUBLICATIONS DURING THE FUNDING PERIOD

M. Sanchez-Sanz, B. A. V. Bennett, M. D. Smooke and A. Linan, "Influence of Strouhal Number on Pulsating Methane/Air Coflow Jet Diffusion Flames," *Comb. Theory and Modelling*, **14**, (2010).

M. B. Long, "Imaging Flames: From advanced laser diagnostics to snapshots," in *Optical Processes in Microparticles and Nanostructures*, A. Serpengüzel and A.W. Poon, Editors. 2010, World Scientific.

R. B. Dobbins and M. D. Smooke, "A Fully Implicit Compact Finite Difference Method for the Numerical Solution of Unsteady Laminar Flames," *Flow, Turb. and Comb.,* **85,** (2010).

P. B. Kuhn, B. Ma, B. C. Connelly, M. D. Smooke, and M. B. Long, "Soot and Thin-filament Pyrometry Using a Color Digital Camera," *Proceedings of the Combustion Institute*, **33**, 2011.

B. Coriton, J. H. Frank, A. G. Hsu, M. D. Smooke and A. Gomez, "Effect of Quenching of the Oxidation Layer in Highly Turbulent Counterflow Premixed Flames," *Proceedings of the Combustion Institute*, **33**, 2011.

L. Tosatto, B. A. V. Bennett and M. D. Smooke, "Parallelization Strategies for an Implicit Newton-based Reactive Flow Solver," *Comb. Theory and Modelling*, **15**, (2011).

L. Tosatto, T. Lu, B. A. V. Bennett and M. D. Smooke, "A Flux-Based Directed Relation Graph Method for the Adaptive On-the-Fly Reduction of Chemical Mechanisms," *Comb. and Flame*, **158**, (2011).

J. D. Herdman, B. C. Connelly, M. D. Smooke, M. B. Long, and J. H. Miller, "A Comparison of Raman Signatures and Laser-Induced Incandescence with Direct Numerical Simulation of Soot Growth in Non-Premixed Ethylene/Air Flames," *Carbon*, **49**, (2011).

M. B. Colket, S. P. Zeppieri, M. D. Smooke and W. W. Kim, "Laminar Flame Speeds, Flammability Limits, and Flame/Reaction Zone Thicknesses for a Surrogate Kerosene Fuel at Engine Operating Conditions," AIAA Paper 2012.

L. Tosatto, F. Mella, M. B. Long, and M. D. Smooke, "A Study of JP-8 Scoflow Flame Structure by Combined Use of Laser Diagnostics and Numerical Simulation," *Comb. and Flame*, **159**, (2012).

J. Cancian, B. A. V. Bennett, M. B. Colket, and M. D. Smooke, "Prediction of Electron and Ion Concentrations in Low-Pressure Premixed Acetylene and Ethylene Flames," accepted for Publication, *Comb. Theory and Modelling*, **17**, (2013).

L. Tosatto, B. A. V. Bennett, and M. D. Smooke, "Comparison of Different DRG-Based Methods for the Skeletal Reduction of JP-8 Surrogate Mechanisms," *Comb. and Flame*, **160**, (2013).

M. D. Smooke, "The Computation of Laminar Flames," *Proceedings of the Combustion Institute*, **34**, 2013.

B. Ma and M.B. Long, "Absolute light calibration using S-type thermocouples," Proc. Combust. Inst. **34**, 3531–3539 (2013).

B. Ma, G. Wang, G. Magnotti, R. S. Barlow and M. B. Long, "Intensity-ratio and color-ratio thin-filament pyrometry: Uncertainties and accuracy," Combust. Flame, **161(4)**, 908–916 (2014).

B. Ma and M.B. Long, "Combined soot optical characterization using 2-D multi-angle light scattering and spectrally resolved line-of-sight attenuation and its implication on soot color-ratio pyrometry," Applied Physics B, 1-17 (2014).

S. Cao, B. Ma, B. A. V. Bennett, D. Giassi, D. P. Stocker, F. Takahashi, M. B.Long, and M. D. Smooke, "A Computational and Experimental Study of Coflow Laminar Methane/Air Diffusion Flames: Effects of Fuel Dilution, Inlet Velocity and Gravity, to be published *Proceedings of the Combustion Institute*, **35**, (2014).

B. Ma, S. Cao, D. Giassi, D. P. Stocker, F. Takahashi, B. A. V. Bennett, M. D. Smooke, and M. B. Long, "An Experimental and Computational Study on Soot Formation in a Coflow Jet Flame under Microgravity and Normal Gravity," *Proceedings of the Combustion Institute*, **35**, (2014).

References

1. E. Fernández-Tarrazo, A.L. Sánchez, A. Liñán and F.A. Williams, "A simple one-step chemistry model for partially premixed hydrocarbon combustion," *Combustion and Flame*, **147**, 32-38 (2006).
2. R.K. Mohammed, M.A. Tanoff, M.D. Smooke, A.M. Schaffer and M.B. Long, "Computational and experimental study of a forced, time-varying, axisymmetric, laminar diffusion flame," *Proceedings of the Combustion Institute*, **27**, 693-702 (1998).
3. S.B. Dworkin, B.C. Connelly, A.M. Schaffer, B.A.V. Bennett, M.B. Long, M.D. Smooke, M.P. Puccio, B. McAndrews and J.H. Miller, "Computational and experimental study of a forced, time-dependent, methane-air coflow diffusion flame," *Proceedings of the Combustion Institute*, **31**, 971-978 (2007).
4. B.C. Connelly, "Quantitative Characterization of Steady and Time-Varying, Sooting, Laminar Diffusion Flames using Optical Techniques," Ph.D. Thesis, Yale University, http://guilford.eng.yale.edu/bcc_thesis.pdf (2009).
5. B.C. Connelly, B.A.V. Bennett, M.D. Smooke and M.B. Long, "A paradigm shift in the interaction of experiments and computations in combustion research," *Proceedings of the Combustion Institute*, **32**, 879-886 (2009).
6. P.B. Kuhn, B. Ma, B.C. Connelly, M.D. Smooke and M.B. Long, "Soot and thin-filament pyrometry using a color digital camera," *Proceedings of the Combustion Institute*, **33**, 743-750 (2011).
7. B. Ma, G. Wang, G. Magnotti, R.S. Barlow and M.B. Long, "Intensity-ratio and color-ratio thin-filament pyrometry: Uncertainties and accuracy," *Combustion and Flame*, **161**, 908-916 (2014).
8. E. Kristensson, A. Ehn, J. Bood and M. Aldén, "Advancements in Rayleigh scattering thermometry by means of structured illumination," *Proceedings of the Combustion Institute*,