Berlin Institute of Technology • Fasanenstr. 89 • 10623 Berlin

Paul Scherrer Institute
Alexey Denisov
Villigen PSI
Villigen, Switzerland

September, 21st - 24th 2014
FLAME STRUCTURE 2014
8th International Seminar on Flame Structure

www.flame-structure-2014.com

Berlin Institute of Technology (TU Berlin)

Prof. Dr. Frank Behrendt
Fakultät III: Prozesswissenschaften,
Institut für Energietechnik

Chair Energy Process Engineering and Conversion Technologies for Renewable Energies (EVUR)
Fasanenstr. 89
10623 Berlin

Contact
info@flame-structure-2014.com
frank.behrendt@tu-berlin.de

Berlin, 2. Mai 2014

Confirmation of paper submission

Name:	Alexey Denisov
Email:	alexey.denisov@psi.ch
Co-author:	Giacomo Colmegna
2nd co-author:	-
3rd co-author:	-
Title of Paper:	Soot growth in a counterflow diffusion flame studied with time-resolved laser-induced incandescence
Program:	Kin-et-ics
Name of Institute:	Paul Scherrer Institute

September, 21st - 24th 2014
FLAME STRUCTURE 2014
8th International Seminar on Flame Structure

www.flame-structure-2014.com

Berlin Institute of Technology (TU Berlin)

Prof. Dr. Frank Behrendt
Fakultät III: Prozesswissenschaften,
Institut für Energietechnik

Chair Energy Process Engineering and Conversion Technologies for Renewable Energies (EVUR)
Fasanenstr. 89
10623 Berlin

Contact
info@flame-structure-2014.com
frank.behrendt@tu-berlin.de

Soot growth in a counterflow diffusion flame studied with time-resolved laser-induced incandescence

Abstract

Soot formation in a counterflow diffusion flame was studied with time-resolved laser-induced incandescence for a variety of gas compositions. Temperature of soot particles, determined by two-color pyrometry, could be fitted by a single exponential decay attributed to convective cooling. Thus, soot particle diameters were determined across the flame. The soot growth rate and the final particle size do not depend on oxygen content in the oxidizing stream. The highest growth rate was observed for propane as a fuel.

Introduction

Soot formation in laminar diffusion flames was extensively studied in past decades [1]–[4]. Counterflow flames have been used as a well-controlled environment with good optical access for studying effects of gas composition [5] and strain rate [6] on sooting properties of diffusion flames.
Time-resolved laser-induced incandescence (LII) was proposed [7] as a method to determine soot particle size based on its heating by a laser pulse and subsequent cooling. Here we apply the method to study soot particle growth in counterflow diffusion flames for a variety of fuels to provide data for validation of soot growth models.

Approach

The measurements were done on a rectangular counterflow diffusion flame burner, described in details elsewhere [8]. We used air enriched up to 60% with oxygen as an oxidizing flow and methane, ethane, propane and ethylene as a fuel. The output of an Nd:YAG laser at 1064 nm was focused into a thin sheet with average fluence of 0.4 J/cm^2. It was shown before [9] that such fluence is high enough to heat all soot particles to the boiling temperature, while still avoiding significant losses through vaporization.
An image-intensified CCD camera recorded LII emission at 450 nm at 650 nm using two dichroic bandpass filters at a varied delay with respect to the laser pulse and the images were averaged along the horizontal axis.
Temperature of soot particles has been determined by the pyrometric method from the ratio of LII signals at two wavelengths, after they have been corrected for different filter transmittance and camera sensitivity.
Temperature of soot particles during LII measurement has been modelled using equations of heat balance as suggested by Melton[7]. He showed that soot particles cool down mostly through convective heat transfer to the ambient gas, which could be described with a closed-form heat transfer model by McCoy and Cha [11].
We used Cantera software [12] to model axisymmetric, opposed-flow diffusion flames using the GRI-Mech 3.0 kinetics mechanism [13]. It was shown before[8] that temperature profiles calculated with Cantera match well to measured temperature profiles with multiline NO LIF thermometry.

Results and Discussion

Temperature profiles measured with two-color pyrometry are close to what Goulay[14] reports for laser fluence of 0.4 J/cm^2 with peak temperatures reaching 4000 to 4500 K (Figure 1 shows one typical example). The convective cooling rate was determined by exponential fitting of temperature decay starting 20 ns after the temperature peak.

www.flame-structure-2014.com

Berlin Institute of Technology (TU Berlin)

Prof. Dr. Frank Behrendt
Fakultät III: Prozesswissenschaften,
Institut für Energietechnik

Chair Energy Process Engineering and Conversion Technologies for Renewable Energies (EVUR)
Fasanenstr. 89
10623 Berlin

Contact
info@flame-structure-2014.com
frank.behrendt@tu-berlin.de

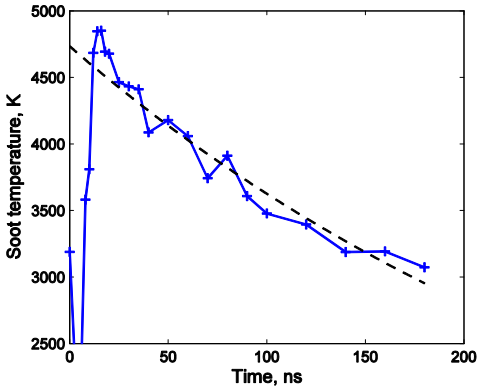

Figure 1. Soot temperature measured with two-color pyrometry at different time delays after the laser trigger. Dashed line shows exponential fit of temperature decay starting at 20 ns. Measurement done in propane flame with 30% oxygen enrichment.

Using the model of convective cooling, we calculated soot particle diameter that corresponds to the fitted decay time. By repeating the procedure for every point across the burner, we could obtain spatial distribution of soot particle size. Figure 2 shows such a profile in propane flame. Here, the fuel is coming from the left, the oxidizing flow from the right and the soot particles start to form as the gas temperature reaches about 1600 K.

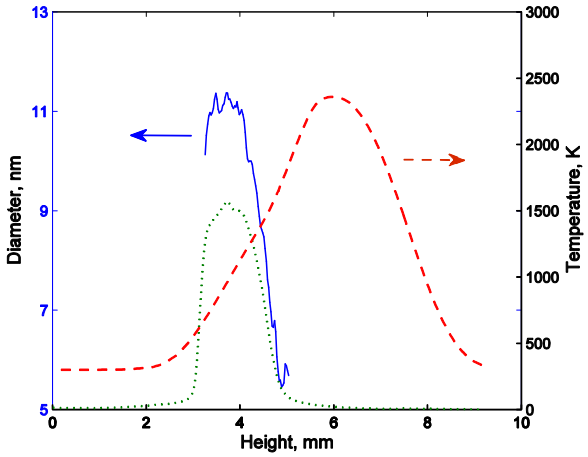

Figure 2. Soot particle diameter derived from the soot cooling rate (solid line). Raw LII taken through 650 nm filter is shown with dotted line (arbitrary units). Calculated gas temperature is plotted with dashed line. Measurement done in propane flame with 30% oxygen enrichment.

Using calculated velocity profiles across the burner, we could determine the time it takes for soot particles to reach specific height and thus reconstruct the time evolution of soot particle diameter. We observed that the diameter of soot particles increases quickly for several milliseconds and then stabilizes around 10-13 nm (see Figure 3). The flame temperature increases with higher oxygen content and so does the mass fraction of soot. However, measured growth rates and final soot particle sizes did not change significantly, so the increase of soot mass fraction can be attributed to a larger number of particles.

www.flame-structure-2014.com

Berlin Institute of Technology (TU Berlin)

Prof. Dr. Frank Behrendt
Fakultät III: Prozesswissenschaften,
Institut für Energietechnik

Chair Energy Process Engineering and Conversion Technologies for Renewable Energies (EVUR)
Fasanenstr. 89
10623 Berlin

Contact
info@flame-structure-2014.com
frank.behrendt@tu-berlin.de

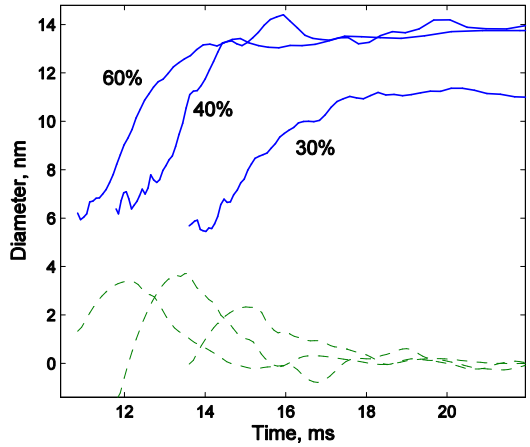

Figure 3. Soot diameter evolution over time (solid line), measured in propane flame with different oxygen enrichment. Dashed line shows smoothed gradient (growth rate) in nm/ms.

Multiplying the rate of diameter increase by the density of soot, we obtained specific soot growth rates: $3.1 \cdot 10^{-3}$ kg/m^{2}s for methane, $4.7 \cdot 10^{-3}$ kg/m^{2}s for ethane, $7.6 \cdot 10^{-3}$ kg/m^{2}s for propane and $6.1 \cdot 10^{-3}$ kg/m^{2}s for ethylene. Although the last value is approximately four times larger than the value reported for ethylene/air diffusion flame[2] it is still within the limits reported by other studies[3]. Notably, the specific soot growth rate in ethylene flame is lower than that for propane even though the mass of produced soot is about four times higher.

Conclusions

Time-resolved LII reveals growth of soot particles in counterflow flame. We saw that after initial linear growth the diameter of soot particles stabilizes at about 10 to 13 nm.

The soot growth rate increases with the number of carbon atoms in a fuel molecule. For a given fuel, the growth rate does not depend on oxygen content in the oxidizer, even though the soot mass fraction increases for higher oxygen content.

Our measurements provide an insight into the early stage of soot particle growth in a well-controlled environment that allows good comparison with chemical kinetics models.

Acknowledgements

This study is part of a project funded by Methanol Casale (www.casale.ch).

www.flame-structure-2014.com

Berlin Institute of Technology (TU Berlin)

Prof. Dr. Frank Behrendt
Fakultät III: Prozesswissenschaften,
Institut für Energietechnik

Chair Energy Process Engineering and Conversion Technologies for Renewable Energies (EVUR)
Fasanenstr. 89
10623 Berlin

Contact
info@flame-structure-2014.com
frank.behrendt@tu-berlin.de

References

[1] R. L. Axelbalim, W. L. Flower, and C. K. Law, "Dilution and Temperature Effects of Inert Addition on Soot Formation in Counterflow Diffusion Flames," *Combust. Sci. Technol.*, vol. 61, no. 1–3, pp. 51–73, 1988.

[2] R. Puri, T. F. Richardson, R. J. Santoro, and R. A. Dobbins, "Aerosol dynamic processes of soot aggregates in a laminar ethene diffusion flame," *Combust. Flame*, vol. 92, no. 3, pp. 320–333, Feb. 1993.

[3] F. Xu, P. B. Sunderland, and G. M. Faeth, "Soot formation in laminar premixed ethylene/air flames at atmospheric pressure," *Combust. Flame*, vol. 108, no. 4, pp. 471–493, Mar. 1997.

[4] C. H. Kim, A. M. El-Leathy, F. Xu, and G. M. Faeth, "Soot surface growth and oxidation in laminar diffusion flames at pressures of 0.1–1.0 atm," *Combust. Flame*, vol. 136, no. 1–2, pp. 191–207, Jan. 2004.

[5] D. X. Du, R. L. Axelbaum, and C. K. Law, "Soot formation in strained diffusion flames with gaseous additives," *Combust. Flame*, vol. 102, no. 1–2, pp. 11–20, Jul. 1995.

[6] V. Huijnen, A. V. Evlampiev, L. M. T. Somers, R. S. G. Baert, and L. P. H. de Goey, "The Effect of the Strain Rate on PAH/Soot Formation in Laminar Counterflow Diffusion Flames," *Combust. Sci. Technol.*, vol. 182, no. 2, pp. 103–123, 2010.

[7] L. A. Melton, "Soot diagnostics based on laser heating," *Appl. Opt.*, vol. 23, no. 13, p. 2201, Jul. 1984.

[8] A. Denisov, G. Colmegna, and P. Jansohn, "Temperature measurements in sooting counterflow diffusion flames using laser-induced fluorescence of flame-produced nitric oxide," *Appl. Phys. B*, pp. 1–8.

[9] H. A. Michelsen, P. O. Witze, D. Kayes, and S. Hochgreb, "Time-Resolved Laser-Induced Incandescence of Soot: The Influence of Experimental Factors and Microphysical Mechanisms," *Appl. Opt.*, vol. 42, no. 27, pp. 5577–5590, Sep. 2003.

[10] R. Hadef, K. P. Geigle, J. Zerbs, R. A. Sawchuk, and D. R. Snelling, "The concept of 2D gated imaging for particle sizing in a laminar diffusion flame," *Appl. Phys. B*, vol. 112, no. 3, pp. 395–408, Sep. 2013.

[11] B. J. McCoy and C. Y. Cha, "Transport phenomena in the rarefied gas transition regime," *Chem. Eng. Sci.*, vol. 29, no. 2, pp. 381–388, 1974.

[12] D. Goodwin, *Cantera: Object-Oriented Software for Reacting Flows*. .

[13] G. P. Smith, D. M. Golden, M. Frenklach, N. W. Moriarty, B. Eiteneer, M. Goldenberg, C. T. Bowman, R. K. Hanson, S. Song, W. C. Gardiner, V. V. Lissianski, and Z. Qin, "GRI-Mech 3.0," *GRI-Mech 3.0*. [Online]. Available: http://www.me.berkeley.edu/gri_mech/.

[14] F. Goulay, P. E. Schrader, X. López-Yglesias, and H. A. Michelsen, "A data set for validation of models of laser-induced incandescence from soot: temporal profiles of LII signal and particle temperature," *Appl. Phys. B*, vol. 112, no. 3, pp. 287–306, Sep. 2013.